EXTENSION OF EQUILIBRIUM CONCEPTS TO NON-EQUILIBRIUM IN GLASSY CHALCOGENIDES: NUMERICAL SIMULATIONS IN THE COULOMB GLASS MODEL

M. CARAVACA[a*], F. GIMENO[a], J. L. MESEGUER- VALDENEBRO[b]
[a]*University Centre of Defence at the Spanish Air Force Academy, Base Aerea de San Javier, Coronel Lopez Peña st., n/n, 30720 Santiago de la Ribera, Murcia, Spain*
[b]*Department of Applied Physics and Materials Engineering, School of Mechanical Engineering, Technical University of Madrid, José Gutiérrez Abascal st., 2, 28006 Madrid, Spain*

In this paper, we investigate the generalization of some equilibrium properties of the Coulomb glass model to characterize the thermal behavior of glassy chalcogenides out of equilibrium. Concretely, we focus on the minimum of the one-particle density of states and the fluctuation-dissipation theorem. The former can be employed as a good thermometer out of equilibrium while the latter reveals the existence of more than one temperature in glassy chalcogenides. The approaches applied in this work are suitable to explore the nature as well as the thermal properties of other glassy systems, such as spin glasses.

1. Introduction

The extension of thermal equilibrium properties to non equilibrium is a fundamental topic to investigate the nature of glassy systems in physics. In fact, in the vitreous state, even the concept of temperature is not well defined [1]. Since statistical physics is fundamentally applied to systems in equilibrium, it is required to generalize some concepts inherent to this discipline to the studies out of equilibrium, in order to achieve a deep comprehension of this kind of materials.

Recently, El-Hakim and Morsy have shown evidences of the relationship between amorphous non-oxide chalcogenides and the Coulomb glass model (also known as the *electron glass model*) [2]. In fact, at very low temperatures, glassy non-oxide chalcogenides behave similar to their oxide counterparts, and they exhibit some features common to most glassy systems [3]: memory effects [4], aging [5,6] or slow relaxation rates [7], among others [8].

In this fashion, it is possible to generalize some equilibrium properties of Coulomb glasses in order to characterize the thermal behavior of glassy non-oxide chalcogenides. The key ingredient is the analysis of the deviations from the ideal equilibrium expressions when the system is placed out of equilibrium. Numerical simulations appear as a good approach to perform the study, given the complexity of the system. Concretely, in our work we employ Monte Carlo simulations.

In the present work we will focus on the analysis of the so called *one-particle properties* in equilibrium of Coulomb glasses. Among them, we will discuss two important phenomena: the dependence on the temperature of the minimum of the one-particle density of states (DOS) and the fluctuation-dissipation theorem (FDT). The first one will reveal that the minimum of the DOS is a

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*Corresponding autor: manuel.caravaca.upct@hotmail.es

good thermometer to characterize glassy systems, both in an out of equilibrium, and can be employed with experimental purposes. The second one will reveal the existence of more than one temperature in glassy chalcogenides, by means of the deviations from the ideal FDT curve.

The Coulomb glass Hamiltonian, which describes the system, is given by the expression [5, 9]

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

where 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 structural disorder of the sample, and K is the compensation, which ensures the electrical neutrality of the system. For the study of conductivity, which we also discuss in this paper, the Hamiltonian has and added term, and is equal to:

$$H = \sum_i \phi_i n_i + \sum_{i<j} \frac{(n_i - K)(n_j - K)}{r_{ij}} + \sum_i E x_i \qquad (2)$$

In this expression, E is the applied electric field (pointing through X direction) and x_i is the horizontal component of the position vector of the i-th impurity. The product of both magnitudes has dimensions of energy.

1.1 Minimum of the one-particle density of states

The one-particle density of states of a physical system is defined as the number of individual energy levels per unit of energy and volume. We usually denote this function as $g(\varepsilon)$, where ε is, in general, a generic variable that represents the energy of the state of one particle. Function $g(\varepsilon)$ is well defined for non interacting systems, where individual energy levels are well determined. For a quantum system of non interacting fermions in equilibrium, with a random disorder within an interval of width W, the DOS takes a constant value over the entire range of energies [10]. In this case, ε_i is the i-th individual energy level. This situation is equivalent to the Coulomb glass model without the interaction term, second addend in Hamiltonian defined in Eq. (1), with the change $\varepsilon_i \rightarrow \phi_i$. If we introduce a new electron in the system in a generic site indexed by k, the change in the Hamiltonian is just the potential ϕ_k.

For the interacting system the argument is similar. In this case, we define the *site energy* as [3,9]

$$\epsilon_i = \phi_i + \sum_{j \neq i} \frac{(n_j - K)}{r_{ij}}$$

The definitions of the variables are given above. If we now introduce a new electron in the system, it is easy to calculate that the change in the Hamiltonian is just the site energy of the new particle added [9]. In analogy with the non interacting situation, for the interacting Coulomb glass model we can identify the variable ε_i of the DOS with ϵ_i.

Due the combined effect of disorder and interaction the Coulomb glasses exhibit frustration, and the DOS presents a gap, following the next equation at zero temperature [10]

$$g(\epsilon) = k|\epsilon - \epsilon_{\mathrm{F}}|^{d-1}$$

In this expression, ϵ_{F} is the site energy of the Fermi level, d is the dimension of the system and k is a constant. Numerical simulations at $T = 0$ verify this dependence, in both two and three dimensions [11]. For $T \neq 0$, the shape of the DOS is qualitatively equal, but it presents a minimum greater than zero, approximately constant in a small region around the Fermi Level [12]. In

numerical simulations it is usual to calculate the DOS with respect to the Fermi level, that is, the function $g(\epsilon - \epsilon_F)$.

1.2 Fluctuation-dissipation theorem

In equilibrium, the FDT quantifies the equivalence of the spontaneous fluctuations of a system in equilibrium with the evolution through such state when the system is slightly displaced from it [13]. It is necessary to apply a small perturbation to the system through an external field, in order to place the system in a regime of linear response. This fact implies that the response function does not depend on the applied field. To model the system, we suppose that the Hamiltonian of Coulomb glasses (Eq. 1) can be perturbed in a quantity ΔH through the application of an external field f:

$$\Delta H = -fX$$

where X represents our observable, conjugated variable of f, so that the product of both magnitudes has dimensions of energy.

The FDT presents a simple relationship between the dynamical response of the system and the correlation in equilibrium [13]:

$$\langle \Delta X(t) \rangle = \langle X(t) \rangle - X_0 = -\beta f C(t)$$

In this expression, $\langle X(t) \rangle$ is the function total response of the system, averaged over different samples of the system under consideration, while X_0 is the value of variable X in equilibrium. So, $\langle \Delta X(t) \rangle$ is the variation in the response of the system. Function $C(t)$ is the correlation function associated to variable X, and β is equal to $1/k_B T$, where k_B is Boltzman's constant and T the temperature. The graphical representation of $\langle \Delta X(t) \rangle$ versus $C(t)$ in equilibrium shows a straight line of slope $-\beta f$, from which we can easily determine the value of T.

In 1982, Sompolinsky and Zippelius revealed the existence of a new temperature in spin glasses, besides of that of the phonon bath (room) T, which is commonly known in the literature as *effective temperature* (T_{eff}) [14]. Their theoretical study showed for the first time the violation of the FDT in glassy systems out of equilibrium. Over the years, the extension of the FDT to non-equilibrium dynamics became a main objective in the study of these materials. Works from Cugliandolo *et al.* [15] and Garriga and Ritort [16] have succeeded in extending theoretically such theorem for characterizing the thermodynamics of spin glasses in both relaxation and stationary regimes. The generalization to Coulomb glasses is immediate, as long as the model is mappable to a spin glass model with long range interaction and spin interchange [3]. Thus, the generalization of the FDT can be employed as an efficient tool via numerical simulations to study the non-equilibrium behavior in glassy chalcogenides. Its graphical representation show deviations from the ideal straight line in equilibrium revealing, in most cases, different regimes in which some key aspects of the glassy state can be directly determined, such as the effective temperature.

It is possible to generalize the FDT even when the system is placed out of equilibrium. The conditions are: (i) the system presents a regime characterized by small thermodynamic fluxes and (ii) the application of the external field, f, perturbs as less as possible this situation.

In our simulations, we have chosen the following perturbation, ΔH, for the Hamiltonian H:

$$\Delta H = -\sum_i f n_i \phi'_i = -fX$$

where ϕ'_i is a new random site potential, totally uncorrelated with variables n_i. The magnitude of the external field is f. We will apply this perturbation to the Hamiltonian described in Eq. (1) from a time t_w, called the *waiting time*, and follow the relaxation of the system via Monte Carlo simulations. Out of equilibrium the system presents slow relaxing rates and is history-dependent, so the total response and the correlation function will depend on both t_w (fixed) and t (variable). The key ingredient to extend the FDT to non-equilibrium is to obtain a graphical representation of

$\langle \Delta X(t, t_{\mathrm{w}}) \rangle$ versus its associated correlation function, $C(t, t_{\mathrm{w}})$. The deviation from the ideal behavior in equilibrium will be depicted from the existence of two different and well differentiated slopes, this time proportional to $-1/T$ and $-1/T_{\mathrm{eff}}$, respectively, as we will show.

2. Mathematical model and numerical details

We investigate the equilibrium properties of semiconductor samples doped with impurities randomly placed, in the regime of strong localization and very low temperatures. This fact is equivalent to assume that the system behaves as a dielectric material and transitions happen by electrons jumping between impurities, in the regime of variable range hopping [9]. We consider that the position of the electron matches that of the impurity, since the value of the localization length, ξ, is considered very small [3]. We study squared samples of lateral dimension L and implement periodic boundary conditions.

The units we employ in the numerical simulations are the following: $l_0 = L/\sqrt{N}$ is the length unit and $1/l_0$ the energy and temperature unit, if the electron charge, e, and Boltzmann constant, k_{B}, are taken as the unit. For this choice, our range of temperatures is [0.001, 0.05]. We consider systems ranging from 500 to 4000 sites, with a minimum distance between them of 0.2. The range of disorder is $W = 2$ and the localization length, $\xi = 1$. When any of these parameters takes another value, it will be properly noted. The characteristic electron-phonon time, τ_0, will be employed as the unit of time. We also choose $K = 1/2$, and consider that each particle only interacts with its nearest image. Therefore, we do not perform Ewald summation.

In order to obtain a wide range of temperatures we employ two kinds of algorithms of simulation. For the lowest temperatures we employ the lowest energy configurations of the system, which can be determined by specific optimization algorithms [17,18]. From these configurations we can calculate the magnitudes in equilibrium by applying the standards of statistical mechanics. For higher temperatures, we use Monte Carlo simulations by means of the Tsigankov *et al.* hybrid algorithm [19], at high enough temperatures so the system quickly reaches the thermal equilibrium. The simulations from the lowest energy configurations are valid in the range of very low temperatures, while those obtained by the Monte Carlo method are inefficient in the same region, since the system does not reaches the thermal equilibrium easily. However, the latter method works very well at higher values of *T*. The employment of both algorithms simultaneously allows us to study a wider range of temperatures.

For simulations using the lowest energy configurations, a statistical average has been carried out over 1000 samples, *i. e.*, different arrangements of sites. For the Monte Carlo algorithm, the total number of simulations, n_{tot}, is defined as $n_{\mathrm{tot}} = n_{\mathrm{s}} \cdot n_{\mathrm{occ}}$, where n_{s} is the number of samples and n_{occ} is the number of initial occupations of electrons per sample. In this work, we have set $n_{\mathrm{s}} = 100$ and n_{occ} ranging between 10 and 50.

3. Results and discussion

3.1 Minimum of the one-particle density of states

A good thermometer in equilibrium for Coulomb glasses is the minimum of the DOS of one particle, that we will denote from now as $g(\epsilon - \epsilon_{\mathrm{F}} = 0) \equiv g_0$. This magnitude can be determined experimentally, for example, through experiments of conduction by tunneling effect. Massey and Lee observed and quantitatively characterized in 3D the Coulomb gap in the density of states for the doped semiconductor Si:B, by measuring the tunneling conductance as a function of the bias voltage [20]. They obtained a pure regime of tunneling conductivity, and determined the value of g_0 from the experimental data obtained.

First, we can introduce an intuitive argument to justify that that g_0 depends linearly on temperature in two-dimensional systems in equilibrium, based on the qualitative information provided by Figure 1. Here, the straight (red) and curved (black) continuous lines represent the

shape of the DOS at zero and non-zero temperature, respectively. The vertical dashed lines delimit the width imposed by the thermal energy, which is proportional to T. At zero temperature the minimum of the DOS is strictly zero. The effect of temperature is traduced in the increasing of the thermal energy of the particles in a factor proportional to T, so they can *move* in a wider zone on the horizontal axis, which presents a width of the order of that thermal energy. Therefore, from the figure we can infer that the value of g_0 is similar to $g(\epsilon - \epsilon_F = T/2)$ at zero temperature. Besides, since the shape of the inner zone of the Coulomb gap in two dimensions is linear [3], we can conclude that g_0 presents a linear behavior in terms of T. Following the same argument, in the three-dimensional case, g_0 should present a quadratic dependence on T, since the shape of the gap is parabolic [3].
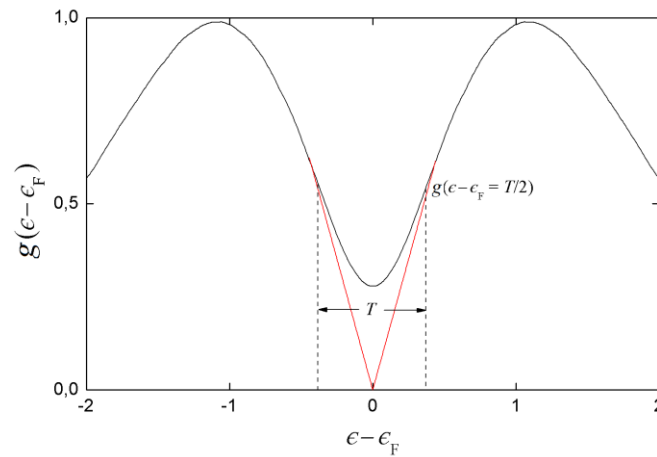

Fig. 1. Qualitative shape of the DOS at zero (red straight lines) and non-zero temperatures (black continuous line). Discontinuous vertical lines delimit the increase of thermal energy per particle by the effect of the temperature.

In our simulations we have calculated the dependence of g_0 on the temperature in both two and three dimensions, in equilibrium, for systems of size $N = 2000$. The surrounding region of g_0 is approximately constant over a small range of energies centered at zero [9]. Thus, we determine the value of g_0 by averaging the density of states in that interval. In each of the following figures the errors are of the order of the spot size, so they have been disregarded in the graphical representation. However, all calculations have been made taking them into account. In Figure 2 it is shown the dependence of g_0 on T in 2D. We note that this dependence is fairly linear. In order to check it, suppose first that g_0 follows a power law of the type T^γ. If we plot the variable $\ln(g_0)$ versus $\ln T$ and we perform a linear fit of the data, we obtain γ from the slope of the calculation. In the detail of Figure 2 we represent g_0 as a function of T, in double logarithmic scale. The slope of the line is $m = 1.011 \pm 0.012$, which reveals that the linear dependence between magnitudes is quite good. In the main graph of Figure 2, the linear fit of the data gives the result

$$g_0 = (-5.3 \pm 4.0) \times 10^{-4} + (0.72 \pm 0.01)T$$

The calculation is analogous for three-dimensional systems. Fig. 3 represents g_0 versus T in double logarithmic scale, this time for the 3D case. The slope of the linear fit has a value of 1.90 ± 0.10, which shows a quadratic dependence in very good approximation, as expected.
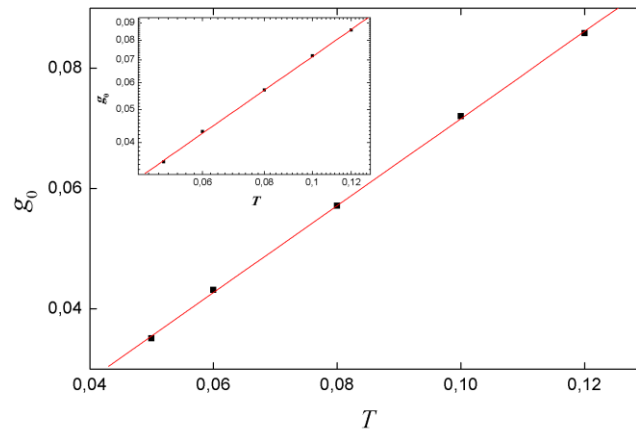
Fig. 2. Dependence of the minimum of the DOS, g_0, on T for a two-dimensional system of size 2000, in equilibrium. The inset represents the same data in double logarithmic scale. Within it, the slope of the linear fit is equal to 1.01 ± 0.01*, which confirms the expected linear dependence between both magnitudes.*
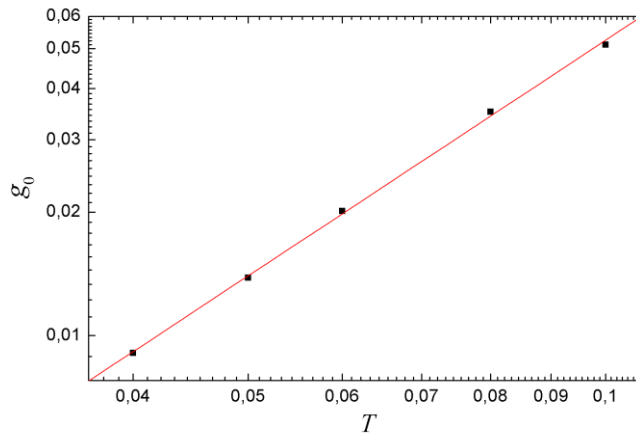

Fig. 3. Dependence of the minimum of the DOS, g_0, on T for a three-dimensional system of size 2000 in equilibrium, in double logarithmic scale. The slope of the linear fit is 1.90 ± 0.10*, which confirms the quadratic dependence expected between both magnitudes.*

The minimum of the density of states remains a valuable thermometer when the Coulomb glass is placed out of equilibrium, where it exhibits frustration: the inability of a physical system to minimize the energy corresponding to all their interactions. This fact generates the presence of significant effects in these materials, glassy behavior. The non-equilibrium dynamics of Coulomb glasses has been characterized by the employment of two temperatures: the temperature of the phonon bath (room), and an effective temperature, T_{eff}, greater than the previous one, which tends to it as the system approaches equilibrium, as commented in the introduction. In Coulomb glasses, the room temperature and the effective temperature correspond, respectively, to the degrees of freedom of the system that equilibrate slowly and rapidly [9, 21].

In this fashion, some of the authors found in a previous work that, in the relaxation process of Coulomb glasses, g_0 also exhibit a linear behavior, this time in terms of T_{eff} [21]. The results were obtained by reproducing a quench experiment at very low temperature by means of Monte Carlo simulations. The calculations were performed for several sizes and relaxation times, and the results revealed that de dependence of g_0 on T_{eff} is linear and scalable for all sizes:

$$g_0 = (0.9 \pm 0.2)T_{\text{eff}} + (-0.011 \pm 0.003)$$

Thus, as a natural extension of the equilibrium properties, g_0 represents an appropriate thermometer to measure T_{eff} experimentally, for example by tunneling effect experiments.

We here focus on another interesting and novel kind of study out of equilibrium, in which one can employ g_0 as a good thermometer: the conductivity of Coulomb glasses in a stationary state placed out of the linear regime, achieved by applying an electric field outside the linear regime. This field is usually applied pointing through one particular direction of the sample. The Hamiltonian which describes the system in this situation was defined in Eq. (2). The effect of a strong electric field produces a partial fill of the Coulomb gap, placing the system in a stationary state out of equilibrium, outside of the linear regime. This fact results in the presence of an effective temperature which depends on the applied electric field [22]. Since there will not be return to the equilibrium state, the effective temperature will not tend to that of the phonon bath along time. In fact, it will remain constant, on average. Numerical simulations of conductivity outside the linear regime are still scarce, and the extension of concepts inherent to equilibrium can be regarded as useful numerical tools to characterize the behavior of Coulomb glasses out of it.

To check the dependence of g_0 on T_{eff} we have performed dynamic Monte Carlo simulations, in which we determine the value of the effective temperature for the application of different electric fields outside the linear regime. As done in previous dynamic simulations, we have implemented periodic boundary conditions in the samples. The way to calculate the effective temperature is the same employed in previous studies of the authors [21, 22], and it is obtained from the determination of the density of states of a particle corresponding to the degrees of freedom that equilibrate slowly.

In our new simulations we calculate the effective temperature for several values of E. Operationally, the conductivity starts to deviate from the linear behavior when the electric field is greater than $T/20$. Looking at the minimum of the DOS at the Fermi level for the stationary case obtained from the results, we observe that it is also linearly dependent on the effective temperature, as in the case of pure relaxation. The behavior is also similar to that found in equilibrium (see Figures 2 and 3). The hypothesis that this is a general result due to the linearity of the Coulomb gap is strengthened from the results presented. In Figure 4 we present data of g_0 as a function of T_{eff} for a 2D system of size 2000 (blue dots) at $T = 0.05$, although the results are similar for other sizes. Errors have been computed assuming that the histogram of the density of states of one particle is a Poisson distribution [9], and are of the order of the dot size. From the linear fit (blue solid line) we obtain a slope equal to 0.60 ± 0.01. This value is similar to that found in equilibrium calculations in Fig. 2. Therefore, we have added to Figure 4 the representation of g_0 as a function of T in equilibrium (black squares). The dashed line represents the extrapolation of the linear fit of the equilibrium data. The results presented confirm the validity of g_0 as a thermometer for measuring T_{eff} in multiple situations and so, the usefulness of the extension of equilibrium properties to non equilibrium dynamics.
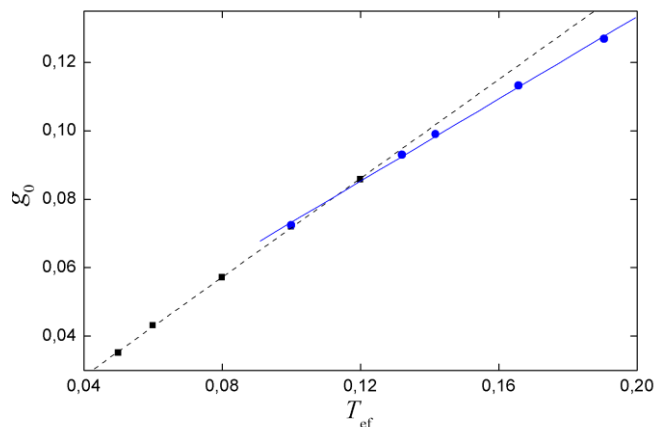

Fig. 4. Dependence of the minimum of the DOS, g_0, on T_{eff} for a 2D system of size 2000 (blue dots). The slope of the linear fit is 0.60 ± 0.01. The black squares represent the dependence of g_0 on T for the same system in equilibrium (same data that in Figure 2).

3.2 Fluctuation-dissipation theorem

In order to obtain an extension of the FDT we have performed two kinds of simulations for the same system. Firstly, we deal with the case of zero field amplitude, $f = 0$, and measure the electronic correlation associated to variable X as a function of two times, from time t_w. The expression for the correlation thus is [9]

$$C(t, t_w) = \frac{W^2}{12} \langle \sum_i n_i(t) n_i(t_w) \rangle$$

On the other hand, we must perform the simulations for the case $f \neq 0$ and measure the total response of the system to the applied perturbation, $\langle \Delta X(t, t_w) \rangle$, once the field is activated in t_w, which presumably will be done immediately. Our plots are based on the representation of $\langle \Delta X(t, t_w) \rangle$ versus $C(t, t_w)$ normalized to $C(t_w, t_w)$ (from now, C_0), when the field is connected. It is mandatory to take control over parameter f, which magnitude has to be small enough to maintain the linear response regime, but not so much in order to get reasonable statistics. A good test for it is to check that the degrees of freedom of the system that quickly equilibrate are thermalized at the room temperature T. This fact implies that those data fall into the ideal straight line which represents the FDT in equilibrium. We stated from our simulations that values of f higher than 0.008 lead to deviations from the ideal behavior and place the system in the non-linear regime. Below 0.008, we need too much statistics to show reasonable results. So, we have selected $f = 0.008$ in all our simulations, which remains valid for all sample sizes.

Under these circumstances, we represent the magnitude $\langle \Delta X(t, t_w) \rangle / C_0$, divided by βf, versus $C(t, t_w)/C_0$, for several values of t_w. In this fashion, the line associated to the FDT in equilibrium, which we will use as a visual guide, has a slope equal to -1 [9]. In Figure 5 we represent the previous variables for a system of size 1000 in relaxation from a quench at $T = 0.02$ and a waiting time $t_w = 10^4$. The total simulation continues until $t = 10^5$. The continuous line represents the ideal FDT in equilibrium (slope -1). From the graphical representation (blue dots) we can observe two differentiated behaviors, characterized by two different slopes. The right side of the plot shows a behavior similar to that of equilibrium, where the data fall into the solid line. This fact reveals that they are degrees of freedom in the system that quickly equilibrate at T. By contrast, the left side of the plot presents another slope, which is equal to $-T/T_{eff}$, showing the presence of slow relaxation degrees of freedom in the system, thermalized at T_{eff}.

This is the main ingredient of the extension of the FDT to non-equilibrium: the ability of showing different timescales for the glassy systems, characterized by slow or fast degrees of freedom.

The information contained in Figure 5 is consistent with the determination of T_{eff} by means of other methods. Concretely, we have employed the site occupation method [9, 21, 22], that some of the authors developed in previous publications, to compare the consistency of the FDT. From it, we have calculated T_{eff} at time t_w. The results are also showed in Figure 5, where we have plotted a dotted straight line of slope $-T/T_{eff}$. It can be directly depicted from the graph that both methods offer the same value of the effective temperature.
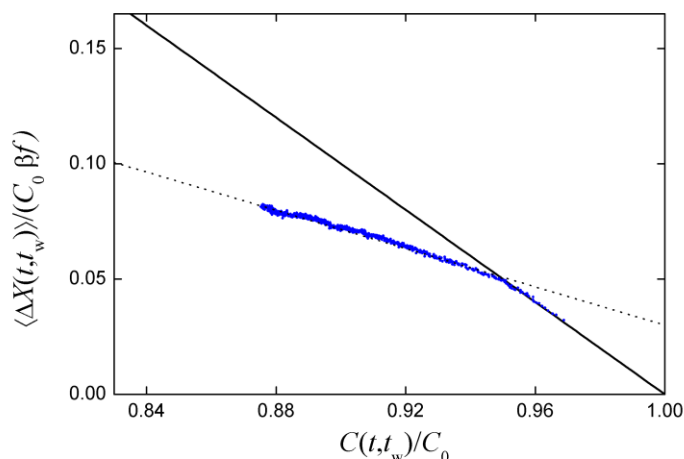
Fig. 5. Fluctuation-dissipation out of equilibrium for a system of size 1000 quenched from T=0.02. The waiting time is $t_w = 10^4$, and the total simulation time is $t = 10^5$. The straight continuous line shows the ideal behavior of the FDT in equilibrium, with slope equal to -1. The dashed line presents a slope $-T/T_{eff}$.

In general, the study of the FDT needs too much statistics to show clear results, without noise. However, it is a good tool to identify the different regimes of relaxation inside the system, and can be particularly useful for stationary regimes, when T_{eff} does not vary along time. Besides, the FDT is an excellent complement to other procedures to check the validity of the calculus of the effective temperature.

4. Conclussions

In this work we show the usefulness of the generalization of concepts inherent to equilibrium to study the non equilibrium dynamics in glassy chalcogenides. In particular, we point out the importance of the minimum of the one-particle density of states, whose characterization may have experimental implications.

The extension of equilibrium properties to non equilibrium is still an open field. In general, other concepts inherent to equilibrium can be generalized in order to study the thermodynamics of glassy chalcogenides. For example, the density of excitations of one particle. Other interesting phenomena which can be studied are related to the *many-particles* properties, such as the dependence of the total energy of the system on the temperature, the many-particle density of states or the absorbed and emitted power, topics in which we are currently working. This kind of studies may lead to a deeper comprehension of the glassy state, whose complete understanding is still far from being reached.

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