

Dielectric susceptibility of the Coulomb-glass

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Abstract

We derive a microscopic expression for the dielectric susceptibility χ of a Coulomb glass, which corresponds to the definition used in classical electrodynamics, the derivative of the polarization with respect to the electric field. The fluctuation–dissipation theorem tells us that χ is a function of the thermal fluctuations of the dipole moment of the system. We calculate χ numerically for three–dimensional Coulomb glasses as a function of temperature and frequency.

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I. INTRODUCTION

The subject of this paper is the calculation of the dielectric susceptibility in Coulomb glasses, a term used for Anderson insulators with Coulomb interactions between the localized electrons. We consider situations deep in the insulating phase, when quantum energies t arising from tunneling are much smaller than the other important energies in the problem, i.e., Coulomb interactions and random energy fluctuations. The model also applies to systems in the quantum Hall regime far away from the peaks, for which the conductivity is exponentially small as compared to e^2/h and the conduction mechanism is by variable range hopping between localized states. The model can be easily extended to granular metals in the insulating regime.

Previous calculations of the dielectric susceptibility of the Coulomb glass have used an expression directly obtained from the analogy between Coulomb and spin glasses [1]. However, the non-local character of the processes involved in Coulomb glasses makes this expression of the dielectric susceptibility inappropriate. Furthermore, it does not correspond to the standard definition of the dielectric susceptibility. The first aim of this work is to present a microscopic expression of the dielectric susceptibility χ valid for the Coulomb glass, and to apply the fluctuation–dissipation theorem to this expression. We also want to calculate χ at very low temperatures. To this end we have to take into account that interactions, and specially those of long–range character, drastically change the properties of systems with localized states [2,3]. Most properties of these systems are affected by electron correlations, and such effects cannot be described by one–particle densities of states or excitations. To deal with complex excitations, methods were developed [4–12] to obtain the low lying states and energies of Coulomb glasses.

The paper is organized as follows: Section II introduces the Coulomb glass models used for the numerical calculations. Section III presents the derivation of a microscopic expression for the dielectric susceptibility of Coulomb glasses. Section IV describes how the low-energy many-particle states are obtained numerically and gives a method to calculate the depen-

dence of the dielectric susceptibility with the frequency. In section V, we present the results obtained for the dielectric susceptibility of Coulomb glasses at very low temperatures and the dependence on temperature, and frequency. Finally, in section VI we extract some conclusions.

II. MODELS

Our next results apply to any model of a Coulomb glass but to be definite we performed numerical simulations for the two most common models: the standard model with a uniform random potential distribution and the classical impurity band model (CIB).

Efros and Shklovskii proposed a practical model to represent Coulomb glass problems with localized electronic states, which has been widely used and extended [2,3]. This model is represented by the standard tight-binding Hamiltonian:

$$H = \sum_i \phi_i n_i + \sum_{i < j} \frac{e^2}{4\pi\epsilon_0} \frac{(n_i - K)(n_j - K)}{r_{ij}} \quad (1)$$

where $n_i \in \{0, 1\}$ denotes the occupation number of site i . We use rationalized units, unlike in most works on the Coulomb glass, because in this problem they constitute the most convenient choice. We will consider sites at random positions, with a density ρ equals to 1, and simulate the disorder by a random potential in each site ϕ_i , uniformly distributed between $-W/2$ and $W/2$. r_{ij} is the distance between sites i and j according to periodic boundary conditions in the sense of [13] in the perpendicular directions to the applied electric field. Charge neutrality is achieved by a background compensation charge $-K$ at each lattice site.

The classical impurity band model (CIB) is a realistic representation of a lightly doped semiconductor in which the random potential arises from the minority impurities [14]. Here we consider an n-type, partially compensated semiconductor with donor concentration N_D , and acceptor concentration $N_A = KN_D$. The Hamiltonian is given by:

$$H = \frac{e^2}{4\pi\epsilon_0} \left(\frac{1}{2} \sum_{i \neq j} \frac{(1 - n_i)(1 - n_j)}{r_{ij}} - \sum_{i\nu} \frac{(1 - n_i)}{r_{i\nu}} \right) \quad (2)$$

where the donor occupation number n_i equals 1 for occupied donors, and 0 for ionized donors. The index ν runs over the acceptors and $r_{i\nu} = |\mathbf{r}_i - \mathbf{r}_\nu|$, with \mathbf{r}_i being the donor coordinates and \mathbf{r}_ν the acceptor coordinates. We chose the donor density ρ equal to 1 and imposed again periodic boundary conditions. For numerical reasons we constrained the nearest neighbor distance to be larger than 0.5. We used $K=0.5$ because the interaction effects are largest there.

III. SUSCEPTIBILITY OF THE COULOMB GLASS

We now proceed to obtain a proper microscopic expression for the dielectric susceptibility of Coulomb glasses. It is applicable to an arbitrary three-dimensional model of Coulomb glass as long as the interaction between charges goes as $1/r$.

A certain analogy between the spin glass and the Coulomb glass lead to an incorrect expression for the dielectric susceptibility of the Coulomb glass. In some sense the local spin s_i in the spin glass is analogous to the site occupation n_i in the Coulomb glass. If only $n_i = 0$ and $n_i = 1$ are allowed due to strong on-site interaction, then the analogy is with spins $s_i = 1/2$. The magnetic field in the spin glass is then analogous to the chemical potential in the Coulomb glass. However, this analogy should not be pushed too far and does not apply to polarizabilities. The magnetic polarizability is the change of total spin induced by the magnetic field, but the electric polarizability is the change in the electric polarization induced by an electric field, not the change in the total occupation number induced by a change of a global potential as the direct analogy would have it. The basic difference between the two susceptibilities can be understood more clearly by realizing that the magnetic polarization $\langle s_i \rangle_T$ comes from field-induced flips of spatially fixed spins, where $\langle \dots \rangle_T$ refers to thermal average. An analogous electric polarization can come from flips of local dipoles, but in many systems it involves a field induced displacement of charges (there is no magnetic equivalent to this because there are no magnetic charges). Such a polarizability is thus not represented by $\langle n_i \rangle_T$ responding to a potential (as in [1] and [15])

but by $\sum_i x_i \langle n_i \rangle_T$ responding to a field (as in [16]). The dielectric susceptibility χ then is:

$$\chi = \frac{1}{\epsilon_0} \frac{\partial P}{\partial E} \quad (3)$$

where E is the total electric field.

Our first aim is to obtain a microscopic expression for Coulomb glasses of the classical definition of the dielectric susceptibility. Let us assume that we apply an electric displacement D . This will induce a polarization P equal to

$$P = \frac{e}{N} \sum_i x_i \langle \Delta n_i \rangle_T \quad (4)$$

where x_i is the position of site i , N is the number of sites, and $\langle \Delta n_i \rangle_T$ is the change in the average occupation of site i due to the applied electric displacement. In the linear approximation, the change in the average occupation of site i due to a general change in the potential is given by:

$$\langle \Delta n_i \rangle_T = \sum_j \frac{\partial \langle n_i \rangle_T}{\partial \phi_j} \Delta \phi_j \quad (5)$$

where $\Delta \phi_j$ is the change in potential at site j . The partial derivative appearing in this expression is proportional to the local susceptibility χ_{ij} :

$$\frac{\partial \langle n_i \rangle_T}{\partial \phi_j} = \frac{e \epsilon_0}{T} \chi_{ij} \quad (6)$$

where T is the temperature, and the Boltzmann constant k_B is taken to be 1 throughout the paper.

The change in potential corresponding to a uniform electric displacement is $\Delta \phi_i = D x_i / \epsilon_0$, so the ratio between P and D is:

$$\frac{\partial P}{\partial D} = \frac{e^2}{TN} \sum_{ij} x_i \chi_{ij} x_j \equiv \chi_0. \quad (7)$$

To calculate the dielectric susceptibility numerically it is convenient to apply the fluctuation-dissipation theorem to χ_0 in order to rewrite it in terms of thermal fluctuations of the dipole moment. Taking into account the expression for the thermal average of

the site occupation, $\langle n_i \rangle_T$, it is easy to obtain that its derivative with respect to the potential in j , i.e., the local susceptibility χ_{ij} , is equal to the fluctuation in the electron occupancy of the two sites involved:

$$\chi_{ij} = \langle n_i n_j \rangle_T - \langle n_i \rangle_T \langle n_j \rangle_T \quad (8)$$

Using this equation in expression (7) for χ_0 , we arrive at

$$\begin{aligned} \chi_0 &= \frac{e^2}{TN} \sum_{ij} x_i (\langle n_i n_j \rangle_T - \langle n_i \rangle_T \langle n_j \rangle_T) x_j \\ &= \frac{1}{TN} (\langle d^2 \rangle_T - \langle d \rangle_T^2) \end{aligned} \quad (9)$$

where $d = e \sum_i x_i n_i$ is the dipolar moment of the sample. The dielectric susceptibility is a function of the thermal fluctuation of the dipolar moment.

Our computer simulations can per force involve only systems of mesoscopic size. For the macroscopic susceptibility we can imagine building a macroscopic system of many mesoscopic cubes of linear size L arranged to fill the space. Each of these samples corresponds to a particular realization of the random positions and energies of the sites involved. The total electric field that a given microscopic sample feels is the sum of the applied field, D , and the induced field. If the applied field is uniform, and the polarizabilities of all the samples were the same, the polarization would also be uniform and the induced field would come only from the boundary of the sample. We then have (in our units):

$$\epsilon_0 E = D - P \quad (10)$$

and get from Eqs. (9) and (10):

$$\chi = \frac{\chi_0}{1 - \chi_0}. \quad (11)$$

At very low frequencies most samples are conducting and we have an effectively uniform distribution of χ_0 over the computer “samples”. At lower frequencies there is no mechanism mitigating the effect broad distribution of χ_0 and the argument leading to Eq. (11) fails and the problem becomes Clausius-Mossotti-like. A general approach to this problem for

random media has been given in [17,18]. Here we shall avoid the inherent complications of such a computation and assume that Eq. (11) is approximately valid even at higher frequencies if we use for χ_0 a value averaged over many computer realizations. The relation $\sigma = i\omega\epsilon$ means that a finite dc conductivity implies an infinite dc dielectric susceptibility. A proper calculation of the DC conductivity can be done by percolation in configuration space, but it is a difficult problem requiring huge numerical efforts so that for three-dimensional systems we could only consider very small samples. Our approximate calculation of the divergence of the susceptibility allows us to estimate the variation of the DC conductivity with temperature as we will see.

IV. NUMERICAL PROCEDURE

A. Low-energy configurations

We calculate the dielectric susceptibility at very low temperatures making use of the ground state and the very low-energy configurations of the systems. With the procedure that we briefly discuss below we obtain the first 5.000 many-particle configurations and calculate their dipole fluctuations, Eq. (9).

We find the low-energy many-particle configurations by means of a three-steps algorithm [10]. This comprises local search [4,5], thermal cycling [11], and construction of “neighbouring” states by local rearrangements of the charges [4,5]. The efficiency of this algorithm is illustrated in Ref. [10]. 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. At the end we only keep configurations with a fix number of electrons, so we work with canonical ensembles.

B. Frequency dependence

At finite frequencies only transitions with characteristic time τ_{IJ} shorter than the inverse of the frequency contribute to the susceptibility. Thus, for a given frequency, we consider two configurations as connected if their τ_{IJ} is shorter than the inverse of the frequency, and we group the configurations in clusters according to these connections.

The characteristic transition time between configurations I and J is [2],

$$\tau_{IJ} = \omega_0^{-1} \exp\left(2 \sum r_{ij}/a\right) \exp(E_{IJ}/T) / Z \quad (12)$$

In this equation, the quantity ω_0 is a constant of the order of the phonon frequency, $\omega_0 \sim 10^{13} \text{ s}^{-1}$. 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 , and Z is the partition function.

We calculate the susceptibility of each cluster through Eq. (9), assuming thermal equilibrium in the cluster. The glassy nature of our systems is responsible for the existence of the clusters, which indicate the non-ergodicity of the systems for times shorter than the critical time connecting all the configurations in a single cluster. Each realization of the systems will be in a given cluster and will not see the other clusters. The probability to be in a cluster depends on the history of the system and is very difficult to estimate. In order to obtain averages of the susceptibility, we will assume that the weight of each cluster is proportional to its partial partition function, which constitutes the simplest possible assumption. The results are finally averaged over many different disorder realizations.

V. RESULTS

If we take into account all types of transitions, including the slowest ones, Coulomb glass behaves like a conductor and is able to screen fully as its susceptibility diverges. But small samples may not have excitations which carry electrons across the entire sample and produce nearly equipotential surfaces at the two opposite edges. So we must consider samples above

a certain critical size at which the steady state susceptibility χ diverges. We found that this critical size is about 200 sites for both models (Eqs. (2) and (1)). Above this size the results are practically independent of size.

Fig. 1 shows the average value of χ_0 as a function of frequency for several temperatures. The plots are for the standard model of size $N = 256$. The localization radius is $a = 0.2$, which is maintained throughout the paper. At low frequencies χ_0 increases with T while at high frequencies it decreases with T . The reason is that at small ω hopping extends over many configurations, and the main effect of T is to enhance the transition rates. At large ω hopping is between two optimal configurations for that frequency (or even two sites) and the main effect of T is to equalize the occupation probabilities. This bears analogy with uncorrelated hopping conductivity which increases strongly with T as $\omega \rightarrow 0$ and behaves as $1/T$ at high frequency.

The result for the CIB model are very similar to those for the standard model and so we do not show them explicitly. The results for the CIB model roughly correspond to those for the standard model with an effective disorder energy of approximately 3.

As already mentioned, an accurate calculation of the frequency dependent macroscopic susceptibility requires the distribution $f(\chi_0)$, not only the average value of χ_0 . Fig. 2 shows the integrated distribution $P(\chi_0) = \int_0^{\chi_0} f(\chi'_0) d\chi'_0$ for $N = 256$ at $T = 0.01$ for different values of the frequency, $\omega \rightarrow 0$ (dotted curve), $\omega = 10^3 \text{ s}^{-1}$ (dashed curve), $\omega = 10^7 \text{ s}^{-1}$ (long dashed curve). The solid curve is a plot of the function $1 - \exp\{-(x\lambda)^\alpha\}$ with the parameters $\lambda = 1.5$ and $\alpha = 0.6$. In the range examined this form of integrated distribution fits (with varying λ and α) fairly well our data for all T and ω . The broad character of the distribution indicates large mesoscopic fluctuations and shows a need to examine in the future the accuracy of our approximation by taking proper account of the distribution of χ_0 .

We define a critical time for saturation τ_c as the inverse of the frequency for which the value of the susceptibility is 95 % of the asymptotic value for extremely long times. We studied the T dependence of this critical time τ_c . For all temperatures considered the values of τ_c are extremely large, which is a sign of the glassy nature of our systems. Since τ_c is

long and close to the saturation of χ_0 , we expect our results to be rather accurate for this study. Fig. 3 plots the logarithm of τ_c *vs.* $T^{-1/2}$ for four sizes of the standard model and for two sizes of the CIB model. The data are fitted quite well by straight lines indicating that a similar mechanism which gives rise to the $T^{-1/2}$ conductivity [4] is also effective in the dielectric susceptibility. This should of course be expected because of the close connection between the two properties. The square root of the slope of each straight line yields a characteristic temperature T_0 which has been often associated with variable range hopping theory in a Coulomb gap. In that theory T_0 is given by:

$$T_0 = \beta \frac{e^2}{4\pi\epsilon_0 k_B a} \quad (13)$$

where $\beta = 2.8$ in three dimensions [19]. Such a theory does not take into account correlations, so comparing our results with the theory can assess the importance of many-body effects. The values of β obtained from Fig. 3 are 0.9 ± 0.2 for the standard model and 0.9 ± 0.1 for the CIB model, i.e., about three times less than in the one-particle theory. This indicates the importance of correlations. The results follow the same trend we found for two dimensional systems by a numerical simulation study of the conductivity [4] where we also found β systematically smaller than was predicted by the one-particle theory, and were able to identify specific many-body processes.

To evaluate, at least approximately, the frequency behavior of the macroscopic susceptibility we calculated $\chi(\omega)$ using Eqs. (9) and (11). The results are exhibited in Fig. 4 where χ is plotted *vs.* frequency for several temperatures. The data correspond to the data for χ_0 of Fig. 1. Note that the susceptibility diverges in the static limit at all temperatures examined, but at a different rate at each temperature. Coulomb glasses will screen like a metal if where not for their glassy nature, which produces typical times for the divergence of the dielectric function much larger than measurement times.

VI. CONCLUSIONS

We derived a microscopic expression for the dielectric susceptibility χ applicable to hopping systems including systems where interactions are important. The expression is particularly suitable for low frequencies. It corresponds to the expression used in classical electrodynamics. Si and Varma [20] have recently study the same problem in the metallic limit of the metal-insulator transition for two dimensional systems, and obtain than the static compressibility vanishes at the transition. Some previous works [1,15] used expression based on an analogy between spin and Coulomb glasses. We argues that these analogies cannot be extended to the susceptibility. The fundamental reason is that unlike in spin glasses the susceptibility in the hopping systems arises from non-local processes.

The fluctuation–dissipation theorem tells us that the dielectric susceptibility is a function of the thermal fluctuations of the dipole moment of the system, instead of the fluctuations of the charge density, result that one obtains when using analogy between Coulomb and Spin glasses. We calculate χ numerically for three–dimensional Coulomb glass systems as a function of temperature and frequency. We found that χ diverges as the frequency tends to zero. One has to consider sizes larger than a critical one of approximately $N = 200$ for the CIB model and for the standard model with $W = 3$.

The logarithm of the critical time for saturation varies proportionally to $T^{-1/2}$, the same dependence as in variable range hopping. The characteristic temperature for this dependence is approximately equal to 0.9, a factor of three smaller than the theoretical predictions for the equivalent constant appearing in variable range hopping.

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FIGURES

FIG. 1. Averaged values of χ_0 obtained for the standard model, plotted against frequency, for several values of the temperature as follows: 0.006 (\bullet), 0.008 (\blacksquare), 0.01 (\blacklozenge), 0.012 (\blacktriangle), 0.014 (\blacktriangleleft), 0.016 (\blacktriangledown), 0.018 (\blacktriangleright), and 0.02 (\circ). The disorder energy is $W = 2$ and the localization radius is $a = 0.2$.

FIG. 2. Accumulated distribution probability of χ_0 for $N = 256$ and $\omega \rightarrow 0$ (dotted curve), $\omega = 10^3 \text{ s}^{-1}$ (dashed curve), $\omega = 10^7 \text{ s}^{-1}$ (long dashed curve). The solid curve corresponds to the fit explained in the text. χ_{av} is the average value at each frequency.

FIG. 3. Logarithm of critical time τ_c for reaching the static susceptibility as a function of $T^{-1/2}$ for four sizes of the standard model, $N = 64$ (\bullet), 128 (\blacksquare), 216 (\blacklozenge), 512 (\blacktriangle), and for two sizes of the CIB model, $N = 216$ (\blacktriangleleft) and 512 (\blacktriangledown).

FIG. 4. Dielectric susceptibility χ as a function of frequency for several temperatures, $T = 0.006$ (\bullet), 0.008 (\blacksquare), 0.01 (\blacklozenge), 0.012 (\blacktriangle), 0.014 (\blacktriangleleft), 0.016 (\blacktriangledown), 0.018 (\blacktriangleright), and 0.02 (\circ). The other parameters and the model considered are the same as in Fig. 1.

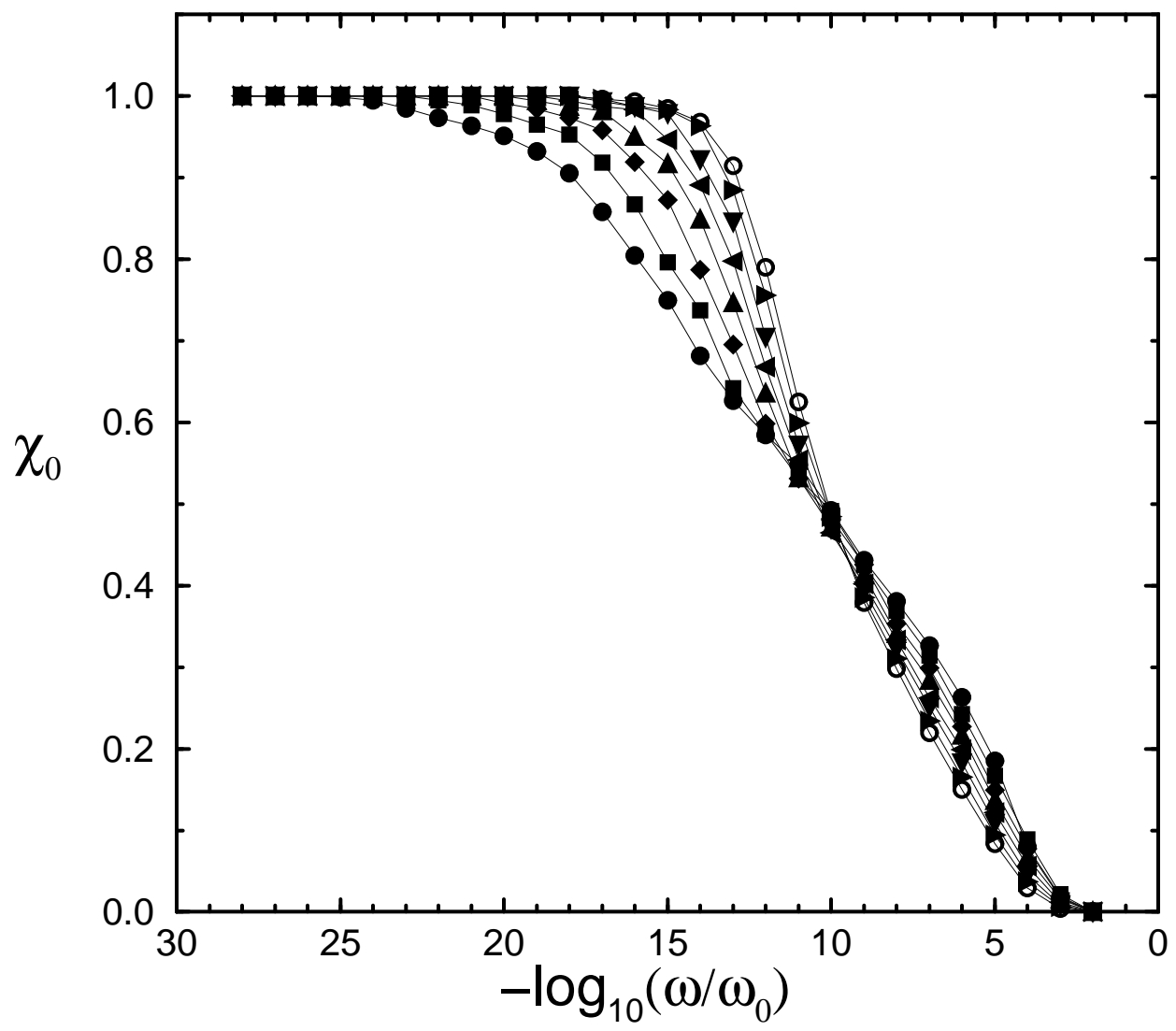


Fig. 1

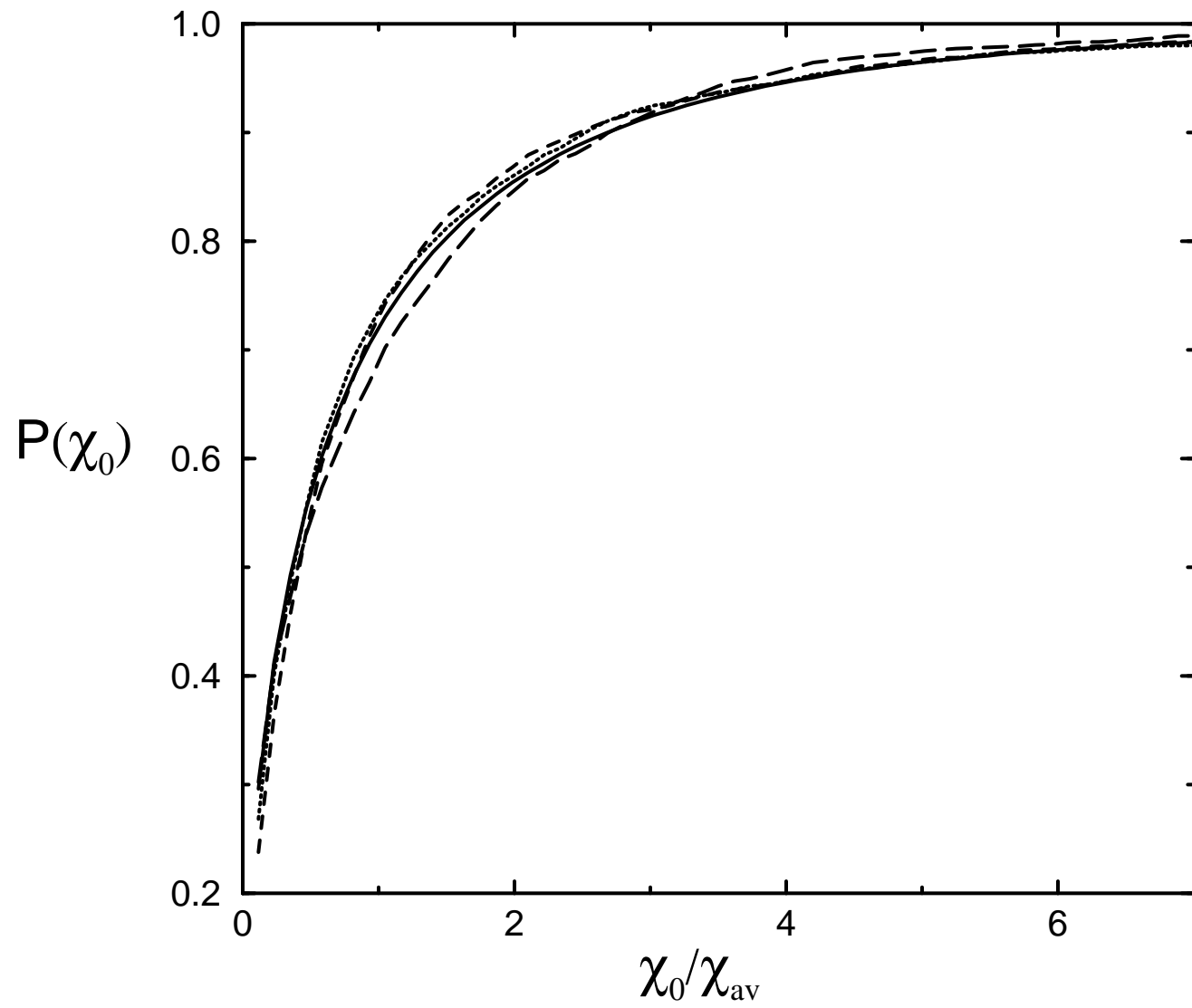


Fig. 2

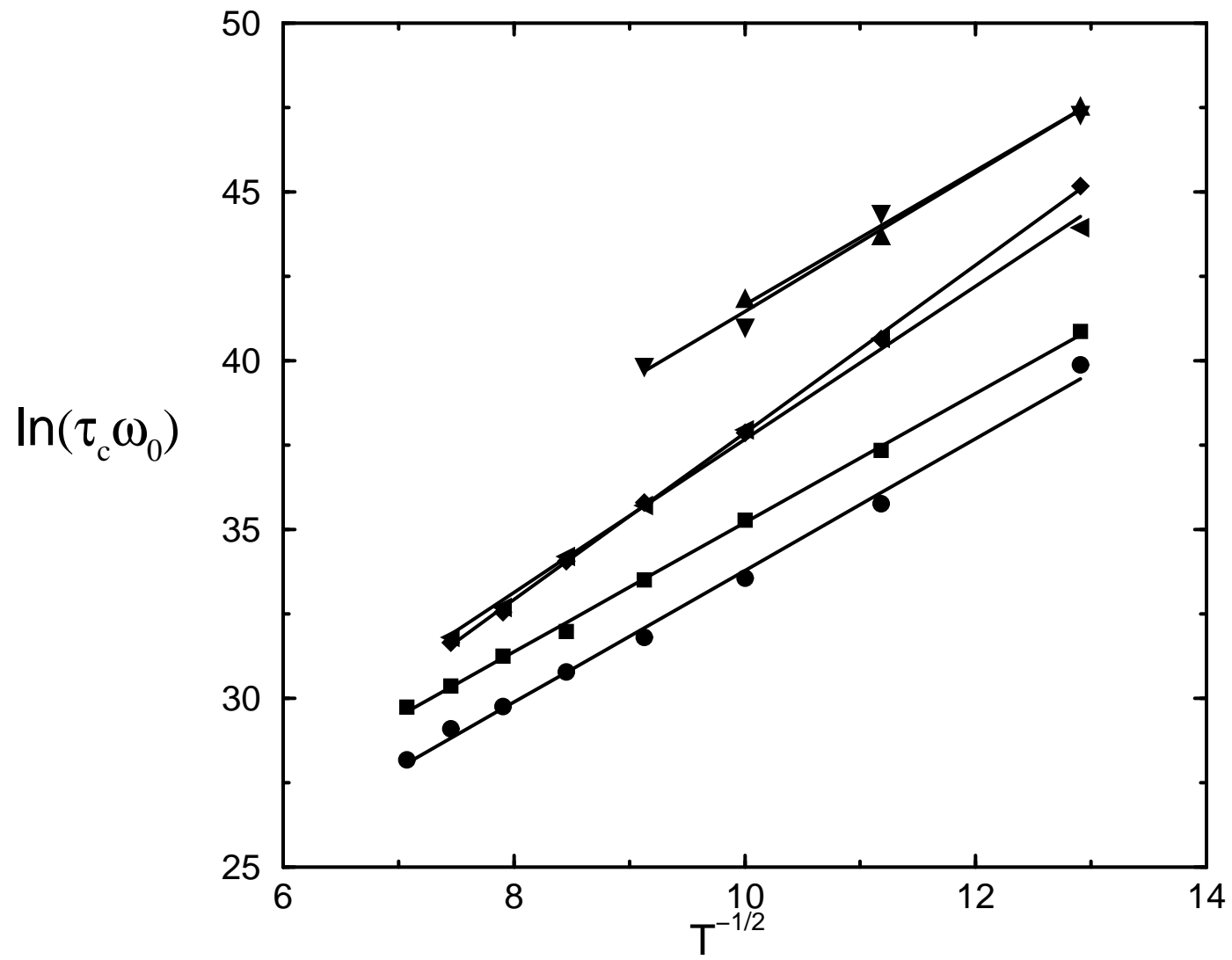


Fig. 3

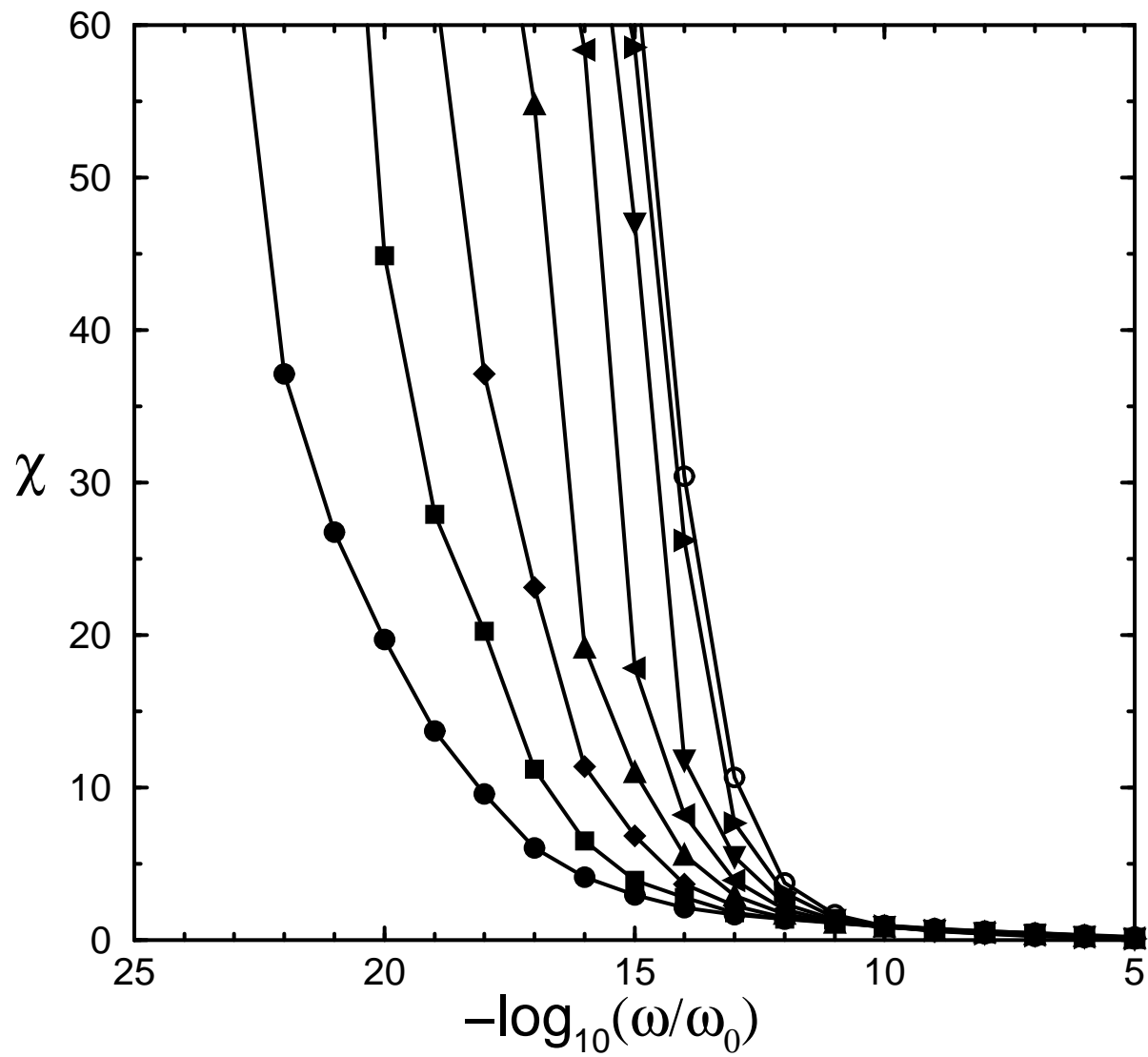


Fig. 4