Glassy properties and localization of interacting electrons in two-dimensional systems

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Abstract

We present a computer simulation study of a disordered two-dimensional system of localized interacting electrons at thermal equilibrium. It is shown that the configuration of occupied sites within the Coulomb gap persistently changes at temperatures much less than the gap width. This is accompanied by large time dependent fluctuations of the site energies. The observed thermal equilibration at low temperatures suggests a possible glass transition only at $T = 0$. We interpret the strong fluctuations in the occupation numbers and site energies in terms of the drift of the system between multiple energy minima. The results also imply that interacting electrons may be effectively delocalized within the Coulomb gap. Insulating properties, such as hopping conduction, appear as a result of long equilibration times associated with glassy dynamics. This may shine new light on the relation between the metal-insulator transition and glassy behavior.

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The existence of glassy properties in a system of strongly disordered localized interacting electrons was predicted some time ago [1], thus introducing the terms “Coulomb glass”, or “Electron glass”. Recently, experimental confirmation of this concept has been obtained through the observation of very slow relaxation times characteristic of glassy dynamics [2,3]. Another important manifestation of glassy properties, namely the existence of multiple low energy minima in the energy landscape of the system, has been known since the first computer simulations of the Coulomb glass [4]. However, while there is strong evidence for glassy behavior, no finite temperature thermodynamic glass transition has been observed either experimentally or numerically. Furthermore, the two dimensional (2D) Coulomb glass has much in common with various 2D spin glass (SG) models, where there is strong numerical evidence that no finite temperature phase transition occurs [5]. We wish to point out that our results also support the absence of any finite temperature phase transition in the 2D Coulomb glass.

The goal of this Letter is to study whether and how the multi-minima structure of the energy landscape, which results from electron-electron interaction, affects the fluctuation properties of the Coulomb glass. Previous finite temperature computer simulations have confirmed the existence of a robust and stable gap in the density of states (DS) around the Fermi level, known as the Coulomb gap. However, as far as we know, the following question has not been studied: Is the configuration of occupied sites within the gap time independent, or does it change persistently with time due to equilibrium fluctuations?

Such fluctuations may be very important for transport if the equilibration time of the system is not too long. For example, near a metal non-metal transition the equilibration time decreases sharply [3]. It should be remembered that the standard theory of hopping conduction in localized systems is based upon percolation theory, and it assumes that the basic electronic configurations and site energies near the Fermi level are time independent. This means that the percolation paths are also time independent. It has long been suggested that this single particle picture of transport may be altered by electron-electron interactions [6], however, as yet no alternate picture has emerged. For clean systems and systems with...
weak disorder, it was recently demonstrated [7], that the persistent change of the occupancy configurations may cause the mechanism of the conductivity to change from percolation to diffusion.

We present here computer simulations designed to study the effect of thermodynamic fluctuations on the site occupation numbers and energies in the Coulomb glass. Our results indeed show a persistent change of the occupation configuration within the Coulomb gap, even at temperature well below the gap width. This persistent change creates a time dependent random potential, which causes fluctuations of the site energies. The fluctuations are much larger than the temperature, and for sites within the gap, are of the order of the gap width. We interpret these results in terms of a drift of the total energy of the system between different minima of the energy landscape. It can also be considered as some kind of classical delocalization effect.

For the purpose of our study we use the standard Coulomb glass Hamiltonian given by

\[ H = \sum_i \phi_i n_i + \frac{1}{2} \sum_{i \neq j} \frac{e^2}{r_{ij}} (n_i - \nu) (n_j - \nu). \]  

(1)

The electrons occupy sites on a 2D lattice, \( n_i = 0, 1 \) are the occupation numbers of these sites and \( r_{ij} \) is the distance between sites \( i \) and \( j \). The quenched random site energies \( \phi_i \) are distributed uniformly within an interval \([-A, A]\). To make the system neutral each site has a positive background charge \( \nu e \), where \( \nu \) is the average occupation number, i.e. the filling factor of the lattice. We concentrate here on the case \( \nu = 1/2 \), where the Fermi level is zero due to electron hole symmetry. It is expected that the features of this model relevant for our purpose are dimensionality, Coulomb interactions, and strong diagonal disorder. Hereafter we take the lattice constant \( a \) as a length unit and \( e^2/a \) as an energy unit. Then, the single particle energy at site \( i \) is given by

\[ \epsilon_i = \phi_i + \sum_j \frac{1}{r_{ij}} (n_j - \nu). \]  

(2)

It has long been established that near the Fermi level the long range Coulomb interaction cannot be considered as a perturbation. This results in a soft gap in the density of single-particle states (DS), known as the Coulomb gap.
In this letter we study numerically the equilibrium fluctuations of the occupation numbers and site energies within the gap, at temperature well below the gap width. To this end we use the standard Metropolis algorithm, where the rate of a hopping transition depends only on the energy difference between the initial and final configurations. Specifically, the rate does not depend on the hopping distance, which is limited only by the system size. The use of such transition rates greatly decreases the equilibration time of the system compared to short range hopping transitions. Note that the Hamiltonian of Eq. (1) does not contain any dynamics in itself, and therefore the simulation time does not reflect any physical time. However, averaging over the simulation time is equivalent to ensemble averaging, assuming the system is in thermal equilibrium. Related approaches are commonly used to study the thermodynamics of various SG models [8].

The simulations were performed on a square lattice of \( L \times L \) sites with periodic boundary conditions. In this torus geometry, the distance between two sites is taken as the length of the shortest path between them. All results obtained were averaged over \( P \) different sets of the random energies \( \{ \phi_i \} \). Unless stated otherwise, the values \( L = 50 \) and \( P = 100 \) were used throughout. Also, we use the notation whereby \( \langle x \rangle \) means time averaging of the quantity \( x \), whereas \( \overline{x} \) denotes averaging over sets of random energies \( \{ \phi_i \} \).

The dynamics within the Coulomb gap can be seen in two ways. One is the time dependence of the single particle energies, which we call spectral diffusion. The other is the time dependence of the configuration of occupied sites within the gap. These two phenomena are closely related since the average occupation number of all sites with energy \( \epsilon \) is given in thermal equilibrium by the Fermi function [9]. Thus, at low enough temperatures, the occupation number of a site changes when it’s energy crosses the Fermi level.

To study the spectral diffusion, we mark after \( t_w \) Monte Carlo (MC) steps all sites whose single particle energies are in a narrow interval \( [-W, W] \) within the gap, and then observe the evolution of the distribution of these energies as the simulation proceeds. We find that after some number of MC steps, this distribution reaches an asymptotic form which is independent of \( t_w \), unless \( t_w \) is shorter than the equilibration time. Fig. [1] shows the final
energy distribution, averaged over sets of \( \{ \phi_i \} \), for \( A = 1 \) and various values of the width \( W \) and the temperature \( T \). The DS of the entire system, which exhibits the Coulomb gap, is also shown. One can see that for any given \( T \), the final distribution does not depend on \( W \) for small \( W \), and it’s width is much larger than \( W \). Furthermore, the width of the distribution is much larger than the temperature.

Another way to observe spectral diffusion is to measure the time average of the single-particle energy at site \( i \), \( \langle \epsilon_i \rangle \), and the standard deviation at the same site, \( \Delta_i = \sqrt{\langle \epsilon_i^2 \rangle - \langle \epsilon_i \rangle^2} \). We perform this calculation for all sites and create a function \( \Delta(\langle \epsilon \rangle) \). Fig. 2 shows this function for \( A = 1 \) and various temperatures. From this figure it is clear that the standard deviation for all sites is much larger than the temperature, while for sites near the Fermi level the standard deviation is \( 2 - 3 \) times larger than for other sites. Sites with large \( \Delta \) are expected to be “active” sites, meaning they often change their occupation numbers as their energies cross the Fermi level. The occupation number changes of these sites are accompanied by a reorganization of the local configuration of occupied sites, which in turn is responsible for the larger value of \( \Delta \). On the other hand, the sites with smaller \( \Delta \) are “passive” sites, and change their energies only in response to the random time dependent potential created by the active sites.

Fig. 3(a) shows the maximum and minimum values of the standard deviation \( \Delta(\langle \epsilon \rangle) \), as a function of temperature. From the results it appears that these functions tend to a finite values as \( T \to 0 \). It is also interesting to consider the width of the curves in Fig. 2 as a function of temperature. We define this width, \( E_w \), as the energy at which \( \Delta(\langle \epsilon \rangle) = \langle \epsilon \rangle \). The meaning of \( E_w \) is that sites which satisfy \( \langle \epsilon \rangle < E_w \) have energy fluctuations larger then their average energy, and therefore are active. From Fig. 2 it is also apparent that these sites have larger value of \( \Delta \), thus supporting our understanding that these are indeed the active sites of the system. The width \( E_w \) as a function of temperature is plotted in Fig. 3(a), and it also appears to tend to a finite value as \( T \to 0 \).

The above results may indicate that the active sites are predominantly within the Coulomb gap. This is reasonable, since the occupation number of sites within the gap
is strongly affected by interactions. However, at $A = 1$ all characteristic energies, including the gap width, are of order unity. Thus, to check whether the active sites are indeed within the gap, it is necessary to simulate $A > 1$. Then, the width of the gap decreases with $A$ as $E_g \sim 1/A$. The results from these simulations are also presented in Fig. 2, where $\Delta(\langle\epsilon\rangle)\sqrt{A}$ is plotted as a function of $\langle\epsilon\rangle A$. For these plots we use $L = 200$ and $P = 20$, and the temperature for each $A$ is $T = 0.05/A$, keeping it constant in units of the gap width. Using this scaling, the curves for $A > 1$ collapse into one, and correspond to the curve for $A = 1, T = 0.05$. Thus, the energy region containing active sites scales as $1/A$, and the active sites are indeed within the Coulomb gap. Moreover, since the number of active sites decreases with increasing $A$, the values of $\Delta$ should also decrease. This is indeed supported by the data in Fig. 2.

Thus, it appears that the configuration of occupied sites within the Coulomb gap persistently changes in thermodynamic equilibrium. To obtain more information about this motion, one can study the correlation function of occupation numbers. We do this by constructing a vector $D(t_w)$ after $t_w$ MC steps have been performed, whose components are the occupation numbers $n_i$ of all sites within a given energy range $[-W, W]$. The vector is normalized so that $D(t_w) \cdot D(t_w) = 1$. As the simulation proceeds, we check the occupation number of these same sites, construct the vector $D(t_w + t)$, and calculate $C(t_w, t) = D(t_w) \cdot D(t_w + t)$. Correlation functions analogous to $C(t_w, t)$ are commonly used to measure the similarity between two configurations. For identical configurations $C(t_w, t) = 1$, while if there is no correlation $C(t_w, t) = 0.5$. Basically, we are interested in $C_\infty = \lim_{t_w \to \infty} \lim_{t \to \infty} C(t_w, t)$, which is a measure of the similarity of two arbitrary states of the system at thermal equilibrium. For a non-interacting system,

$$C_\infty = \frac{\int_W^W f^2(\phi) d\phi}{\int_{-W}^W f(\phi) d\phi},$$

where $f(\phi)$ is the Fermi function. Thus, for the non-interacting system $C_\infty = 1 - T/W$ at $W \gg T$ and $C_\infty = 0.5$ at $T \gg W$.

In order to evaluate $C_\infty$ from the simulation, we measure $C(t_w, t)$ as a function of $t$ for
a given $t_w$, and wait long enough so that $C(t_w, t)$ becomes independent of $t$. We denote this saturated value as $C(t_w, \infty)$. We then increase $t_w$ until $C(t_w, \infty)$ becomes independent of $t_w$, and thus obtain our estimate of $C_\infty$. The value of $t_w$ at which $C(t_w, \infty)$ becomes independent of $t_w$, is the equilibration time $\tau_{eq}$ of the system.

In this light, an important question is whether the Hamiltonian of Eq. (1) exhibits a finite temperature glass transition in 2D. If so, then below the transition temperature the equilibration time should increase with system size $L$, and our results may also depend on $L$. To test this we have studied the size dependence of the equilibration times \cite{10}, and found that within the temperature range studied here, namely $T \geq 0.05$, the equilibration times (measured in MC steps/site) saturate as a function of $L$. The value of $L$ at which the equilibration times become $L$-independent is the correlation length of the system, and is smaller than the system size we use to produce the results. The $L$-independent equilibration times strongly increases with decreasing temperature, making it difficult to study temperatures below $T = 0.05$. However, since at $A = 1$ the temperature $T = 0.05$ is well below any relevant energy scale, we conclude that there is no finite temperature phase transition.

The results for $C_\infty$ for the interacting system are shown in Fig. 3(b) as a function of temperature, for $A = 1$ and $W = 0.3$. The corresponding function for the non-interacting system, calculated directly from Eq. (3), is also shown. Attempting to extrapolate the results to lower temperature, we obtain $\lim_{T \to 0}(1 - C_\infty) \approx 0.15$. Thus, for any two different configurations in thermal equilibrium as $T$ approaches zero, about 15% of the sites within the energy interval $[-0.3, 0.3]$ will have different occupation numbers. This also means that about 30% of sites in this interval are active. Note that by increasing $W$ we include more sites in the correlation function $C_\infty$, but according to our results for the spectral diffusion, most of these sites remain passive as $T \to 0$. Thus, $C_\infty$ should increase as $W$ increases, as we indeed observe for $W = 0.6$.

We view the large values of $1 - C_\infty$ at low $T$ as a manifestation of the multiple minima of the total energy landscape of the system. These minima, known as pseudoground states (PS’s) in the context of the Coulomb glass \cite{4}, are close in energy to the ground state, but
differ from each other by a finite fraction of the site occupation numbers. Their properties have been widely studied theoretically [11], and they have been used to explain [12] the experimentally observed long relaxation time [2,3]. The details of our interpretation are as follows: The Gaussian fluctuations of the total system energy are $T\sqrt{C_V L^2}$, where the specific heat $C_V$ is of the order of $T/E_g$. At finite temperature the system drifts through all PS’s that are inside the fluctuation interval of the total energy. This drift causes a persistent change of the occupation number configuration, and creates a time dependent random potential which is responsible for the spectral diffusion of the site energies. The picture should remain $T$-independent at very low $T$, providing the fluctuation interval contains many PS’s. Since the number of PS’s increases exponentially with the volume of the system [11], this last condition should be fulfilled down to zero temperature in a macroscopic system.

The following example may clarify our interpretation. Suppose there is no disorder in the system, so that $A = 0$. Then at low temperature the system forms a Wigner crystal, which is two-fold degenerate on a square lattice at half filling. These two states represent our PS’s. If transitions between them are permitted, then all sites in the system continuously change their occupation number and the energy of each site fluctuates with a standard deviation of order unity. Note that a major difference between the Wigner crystal and the disordered system is that the former has a gap in the excitation spectrum, while the latter possesses a continuous spectrum of PS’s.

It is important to point out that our results cannot be explained by assuming that the excitations of the system are separated pairs of sites, with electrons hopping back and forth between the sites of each pair. This assumption would mean that electrons are effectively localized in space. Since the energy density of such excitations is constant at low energies, meaning the number of available excitations decreases linearly with temperature, one immediately obtains that $\lim_{T \to 0} (1 - C_\infty) \approx T/W$, like in the non-interacting system. The same temperature dependence is obtained even if excitations involve a few electrons that change their positions simultaneously (so called many electron excitation [6]). In fact, any picture based upon confined separated excitations which do not interact with each other would
mean that \( \lim_{T \to 0} (1 - C_{\infty}) \approx T/W \). Since our data definitely contradicts this temperature dependence, we conclude that such excitations cannot explain our results.

Thus, the results presented in this work may indicate the existence of a classical delocalization effect which is related to the glassy properties of the system. The formal criterion for the existence of this effect is that \( \lim_{T \to 0} (1 - C_{\infty}) \) is nonzero. The limit \( T \to 0 \) should be taken in such a way that the thermodynamic fluctuations of the total energy are larger than the energy distance between PS’s. In a macroscopic system such a limit should always be possible. It is important, however, that the equilibration time of the system increases greatly as the temperature goes to zero. This increase should be even more pronounced if one takes into account the exponential dependence of the tunneling rate on the tunneling distance. Therefore, as the temperature decreases, the system will be frozen in phase space during times shorter than the equilibration time. We believe this to be the cause of the insulating properties of the Coulomb glass, such as hopping conduction. A possible confirmation of this view may be found in the experimental observation that the equilibration time decreases sharply near the metal-insulator transition. A similar view was also suggested by Pastor and Dobrosavljevic, however, they considered a dynamic mean field theory with infinite range interaction, thus neglecting the effect of dimensionality.

In summary, we have presented strong computational evidence that in a disordered 2D system of localized interacting electrons, the configuration of occupied sites within the Coulomb gap persistently changes with time. This effect persists down to temperatures well below the Coulomb gap width, and is accompanied by a time dependent random potential responsible for fluctuations of the site energies within the gap. We view this as a classical delocalization effect, which may be suppressed at low temperature due to very long equilibration times. This suggests that the transport properties of the system may be intimately related to glassy behavior. Thus, the system may be considered as a “slow metal”. The increase of the localization radius due to quantum mechanical overlap may transform it into a normal metal. We thank Z. Ovadyahu and A. Vaknin for many helpful discussions. A.E. would also like to acknowledge fruitful discussions with A. I. Larkin and B. I. Shklovskii.
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REFERENCES

[1] J. H. Davies et al., Phys. Rev. Lett. 49, 758 (1982); Phys. Rev. B 29, 4260 (1984).

[2] M. Ben-Chorin et al., Phys. Rev. B 48, 15025 (1993); A. Vaknin and Z. Ovadyahu, Phys. Rev. Lett. 81, 669 (1998); G. Martinez-Arizala et al., Phys. Rev. Lett. 78, 1130 (1997); C. J. Adkins et al., J. Phys. C 17, 4633 (1984).

[3] Z. Ovadyahu and M. Pollak, Phys. Rev. Lett. 79, 459 (1997).

[4] S. D. Baranovskii et al. J. Phys. C 12, 1023 (1979).

[5] E. Marinari et al., in “Spin Glasses and Random Fields”, edited by P. Young (World Scientific, Singapore 1998).

[6] M.L Knotek and M. Pollak, J. Non-Cryst. Solids, 8-10, 505 (1972); M.L Knotek and M. Pollak, Phys. Rev. B, 9, 664 (1974); M. Pollak and M. Ortuno, in “Electron-Electron Interaction in Disordered Systems”, Ed. by A.L. Efros and M. Pollak, (North-Holland 1985).

[7] A. L. Efros, Phys. Rev. Lett., 68, 2208 (1992).

[8] K. Binder and D.W. Heerman, “Monte-Carlo Simulation in Statistical Physics.” (Springer, Berlin 1997).

[9] A. L. Efros and F. G. Pikus, Phys. Rev. B, 48, 14694 (1993).

[10] D. Menashe, O. Biham, B.D. Laikthman and A.L. Efros, To be published elsewhere.

[11] Sh. Kogan, Phys. Rev. B 57, 9736 (1998)

[12] A. Perez-Garrido et al., Proc. of the 7th Int. Conf. on Hopping and Related Phenomena, Phys. Stat. Sol.(b) 205, 31 (1998); A. Dias-Sanches et al., ibid p.17; Clare C. Yu, Phys. Rev. Lett., 82, 4074 (1999)

[13] A. A. Pastor and V. Dobrosavljevic, preprint cond-mat/9903272
FIGURES

FIG. 1. Final energy distribution of sites initially in the energy range $[-W, W]$. Results are shown for $A = 1$ and different $T$ and $W$. Only positive energies are shown due to electron-hole symmetry. The DS of the entire system is also shown for different $T$.

FIG. 2. Site energy standard deviation as a function of site average energy $\Delta \langle \epsilon \rangle$, for various values of $A$ and $T$. For $A > 1$ the temperature is given by $T = 0.05/A$.

FIG. 3. (a) Maximum and minimum values of $\Delta \langle \epsilon \rangle$ ($\Delta_{\text{max}}$ and $\Delta_{\text{min}}$), and the width $E_w$ as a function of temperature for $A = 1$. (b) The correlation function $1 - C_\infty$ as a function of temperature for $A = 1$ and $W = 0.3$. The solid line shows the correlation function for the non-interacting system.
Figure 1
Figure 2

The graph shows the relationship between $\Delta \sqrt{A}$ and $\langle \epsilon \rangle A$. Different symbols represent different values of $A$ and $T$. The plot includes points for $A = 1, T = 0.15$, $A = 1, T = 0.1$, and $A = 1, T = 0.05$. Additionally, there are symbols for $A = 2$, $A = 3$, and $A = 4$.
Figure 3