Glass Transition and the Coulomb Gap in Electron Glasses

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We establish the connection between the presence of a glass phase and the appearance of a Coulomb gap in disordered materials with strongly interacting electrons. Treating multiparticle correlations in a systematic way, we show that in the case of strong disorder a continuous glass transition takes place whose Landau expansion is identical to that of the Sherrington-Kirkpatrick spin glass. We show that the marginal stability of the glass phase controls the physics of these systems: it results in slow dynamics and leads to the formation of a Coulomb gap.

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The relation between the slow dynamics of Coulomb glasses and the appearance of a soft ''Coulomb'' gap in their density of states (DOS) due to strong electron-electron repulsion [1,2] has been a mystery for a long time. Efros and Shklovskii [3] showed that the long-range Coulomb interaction between localized electrons in semiconductors leads to a soft gap and to the crossover in the temperature dependence of the conductivity from Mott's law $\ln(\rho) \sim (T_M/T)^{1/4}$ to the Efros-Shklovskii law $\ln(\rho) \sim (T_{ES}/T)^{1/2}$ at low temperatures [4]. The latter was verified in semiconductors, alloys, and granular metals, and recently the gap itself was observed in semiconductors [5]. In such materials, the presence of disorder frustrates the Coulomb interactions and leads to glassy behavior [6], as was first evidenced by the slow relaxation of charge injected into compensated semiconductors [7]. Later, Ovadyahu's group established that the slow dynamics in indium oxides is indeed due to electron-electron interactions [8], and recently they found memory and aging effects similar to those of spin glasses [9].

Despite the experimental progress a thorough understanding of the glass phase is still missing. The difficulty is due to many particle correlations that are essential to describe glassy phenomena [10] but are not captured in the single particle theory by Efros and Shklovskii. An increasing number of experiments suggests that multiparticle processes and correlations are also crucial for a *quantitative* description of the hopping conductivity [11].

The mean field solution of a model of uniformly interacting electrons in a disordered medium indicates that the glass transition and the formation of a pseudogap in the DOS are driven by the same mechanism, and a similar relation has been conjectured for the Coulomb glass [12].

The goal of this Letter is to develop a formalism that accounts for the correlations between the electrons in a realistic model for Coulomb glasses in 3D and allows one to study the properties of the vitreous state. Our approach is based on the locator approximation developed for spin glasses in Refs. [13–15]. The idea is to map the original lattice problem into an effective single-site problem that encodes correlations by the distribution of a fluctuating local field, which gives exact results for infinite range models. In the limit of strong disorder, the Coulomb interactions are essentially unscreened, so that the effective number of neighbors is large and the locator approximation is parametrically well justified. In this regime, we find a replica symmetry breaking glass transition (at finite T_c), which belongs to the same universality class as the transition in the Sherrington-Kirkpatrick (SK) spin glass [16]. Below T_c, ergodicity is broken since the phase space divides into an exponential number of metastable states, only a few of which can be explored in finite time. As in any generic glass, the vast majority of those states is only marginally stable and thus exhibits soft modes that lead to the slow relaxation dynamics observed in experiments. We show that the genuine Coulomb gap starts to form only below T_c; in particular, we derive the asymptotics of the DOS at low temperatures (parabolic in 3D) from the condition of marginal stability.

We consider the classical model [4] for strongly localized electrons occupying a fraction $K = 1/2$ of a given set of impurity sites i,

$$H = \frac{1}{2}\sum_{i\neq j}(n_i - 1/2)J_{ij}(n_j - 1/2) + \sum_i n_i\epsilon_i, \qquad (1)$$

where $n_i \in \{0, 1\}$ is the occupation number of the site i. For simplicity, we take them to be arranged on a cubic lattice with lattice spacing $\ell \equiv 1$. The unscreened Coulomb interactions are described by $J_{ij} = 1/r_{ij}$ in units where $e^2/\ell \equiv 1$, and the ϵ_i's denote random on-site energies. The particle-hole symmetry of the $K = 1/2$ case suggests to introduce *pseudospin* variables $s_i = n_i - 1/2 = \pm 1/2$. Further, we assume a Gaussian distribution of width W for the on-site energies ϵ_i. Their randomness emulates the effect of the disorder in the site positions which is present in all physical electron glasses and generates rather large site-to-site fluctuations of the Coulomb potential. We focus on dimension $D = 3$ and on the limit of strong disorder, $W \gg 1$, which reduces the bare density of states and thus suppresses the screening on short scales so that the effective interactions remain long range; this justifies the use of the locator approximation [17]. We

show that at low temperatures the self-generated disorder becomes very large. We thus expect that in this regime the locator approximation becomes valid even in the case of weak disorder.

In the case of long-range interactions, diagrammatic expansions (see, e.g., [2]) can be efficiently resummed since the large number of effective neighbors allows one to approximate the self-energy by an average local term, as shown in Fig. 1. This also suggests to replace the interactions of a given site with its environment by an effective local field described by couplings of the site to its replicas. This reduces the model to a single-site problem, while the disorder average allows one to translate the complexity of the environment into a nontrivial replica structure of the one-site Hamiltonian [12,15],

$$\beta H_0(\{s_\alpha\}) = \frac{1}{2}\sum_{a,b} s_a(\mathcal{B} - \beta^2 W^2 \mathcal{I})_{ab} s_b. \tag{2}$$

Here $\mathcal{I}$ denotes a $n \times n$ block matrix with all entries equal to 1 (as usual, the number of replicas n is implicitly assumed to tend to zero in the end of calculations). We note that this is similar to the way in which the SK model is transformed into a one-site problem. Indeed, the locator approximation applied to the SK model sums all treelike diagrams with doubled interaction lines which becomes exact in the large N limit.

To make contact between the Hamiltonians (1) and (2), we require that they both yield the same single-site correlation functions,

$$\langle s_a s_b\rangle = \sum_{\{s_\alpha\}} s_a s_b e^{-1/2\sum_{\alpha,\gamma} s_\alpha \tilde{\mathcal{B}}_{\alpha\gamma} s_\gamma} = \left[\frac{1}{\tilde{\mathcal{B}} - \Sigma}\right]_{ab}$$
$$= \frac{1}{N}\sum_i \langle s_{i,a} s_{i,b}\rangle = \frac{1}{V}\mathrm{Tr}\left[\frac{1}{\beta J - \beta^2 W^2 \mathcal{I} - \Sigma}\right]_{ab}. \tag{3}$$

We have averaged over the random energies ϵ_i and defined $\tilde{\mathcal{B}} = \mathcal{B} - \beta^2 W^2 \mathcal{I}$. In Eqs. (3) we approximated the full propagator for either model as a simple geometric series with a *local* self-energy insertion $\Sigma_{ab}\delta_{ij}$, as motivated above. Since the mapping is to preserve correlations, the self-energy has to be the same for both models [15]. From (2), we obtain the free energy

$$n\beta F(\mathcal{B}) = -\ln\left[\sum_{s_\alpha} e^{-1/2\sum_{\alpha,\gamma} s_\alpha \tilde{\mathcal{B}}_{\alpha\gamma} s_\gamma}\right] + \frac{U(\mathcal{B})}{2}, \tag{4}$$

where $U(\mathcal{B})$ has to be determined such that the saddle point equations with respect to $\mathcal{B}$ yield back Eqs. (3). Up to a function of temperature, we find

$$U(\mathcal{B}) = \mathrm{tr}\left[\ln(\tilde{\mathcal{B}} - \Sigma) - \frac{1}{V}\mathrm{Tr}\ln(\beta J - \beta^2 W^2 \mathcal{I} - \Sigma)\right], \tag{5}$$

where tr denotes the trace in replica space. We emphasize that in this expression the self-energy Σ has to be considered as an implicit function of $\mathcal{B}$ as defined through Eq. (3). These equations are very similar to the ones obtained in [18] for the regime $W \lesssim 1$ where the locator approximation is difficult to justify, however. In the following we need spatial traces like $g_n(x) = V^{-1}\mathrm{Tr}\frac{1}{(\beta J + x)^n}$ which we evaluate in Fourier space, $g_n(x) = \int \frac{d^3k}{(2\pi)^3} \times \frac{1}{(\beta J_k + x)^n}$, with $J_k = 4\pi/k^2$ at small k. We assume some cutoff procedure that regularizes the small scale physics so that $\int_k = 1$ and $\int_k J_k = 0$. For $x \gg \beta$ we obtain $g_n(x) \approx x^{-n}[1 + C_n(\beta/x)^{3/2}]$, where $C_1 = 2\sqrt{\pi}$ and $C_2 = 5\sqrt{\pi}$.

Let us first discuss the replica symmetric (RS) solution of Eqs. (3) for which we assume $\Sigma_{ab} = -\Sigma_0\delta_{ab} + \Sigma_I \mathcal{I}_{ab}$, and $\mathcal{B}_{ab} = -B_0\delta_{ab} + B_I \mathcal{I}_{ab}$. For $W \gg 1$, we find $\Sigma_0 \approx \sqrt{2\pi}\beta W$, suggesting the interpretation of Σ_0^{-1} as the fraction of thermally active sites (for $T_c < T \ll W$). The distribution of local fields obtained from this RS solution agrees well with numerical data for $T \gtrsim T_c$ and $W \lesssim 1$ [18]. A depletion of sites in small fields is found due to strong correlations in this "Coulomb plasma." However, a closer analysis reveals that there is no true pseudogap on the replica symmetric level, the depletion disappearing completely for strong disorder. This is also reflected in the *pseudoequilibrium* charge susceptibility defined by $\chi_{\mathrm{RS}} = \beta(1/4 - \langle s_a s_b\rangle_{a\neq b}) \approx \beta/\Sigma_0$ which tends to a finite constant ($\sim 1/W$) within this solution. Physically, χ measures the charge response to a local potential change when the particles on other sites are allowed to readjust to the induced charge, without driving the system out of its local free energy minimum. The genuine Coulomb gap is formed only when the replica symmetry is broken. For $W \gg 1$, the RS solution indeed exhibits an instability (the extremum in F_{RS} becomes a saddle point) when the condition

$$\int_{-\infty}^{\infty} dy \frac{e^{-y^2/2(W^2 + B_I/\beta^2)}}{\sqrt{2\pi(W^2 + B_I/\beta^2)}} \frac{1}{[2\cosh(\beta y/2)]^4}$$
$$= [g_1^{-2}(\Sigma_0) - g_2^{-1}(\Sigma_0)]^{-1} \approx (\pi\beta^3\Sigma_0)^{-1/2} \tag{6}$$

is met, from which we extract the critical temperature $T_c \approx W^{-1/2}/[6(2/\pi)^{1/4}] \ll 1 \ll W$. We emphasize that the difference $g_1^{-2}(\Sigma_0) - g_2^{-1}(\Sigma_0)$ is controlled by the contribution from large scales, $1/k \sim \sqrt{W}$, which justifies

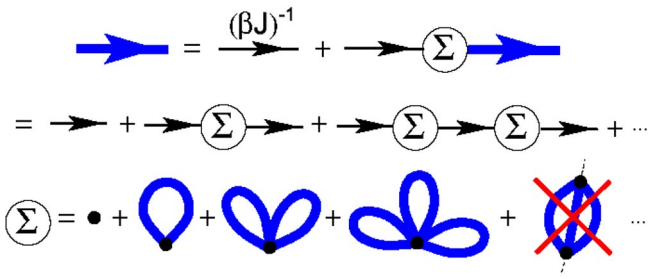

FIG. 1 (color online). For long-range interactions the self-energy Σ can be approximated by a local operator. The full propagator is obtained as a simple geometric series. The (thick) lines represent the (renormalized) interaction, the dots one-site correlators.

our assumption of a large number of effective neighbors. To verify that the transition is not an artifact of the locator expansion we calculated the disorder average of the squared correlator $\langle s_i s_j\rangle_c^2$ in the original lattice model and checked that it shows critical behavior at the same T_c.

The instability (6) signals a continuous glass transition with full replica symmetry breaking (RSB). We may analyze it further by expanding the free energy with respect to the replicon mode $\delta\mathcal{B}$ (with $\delta\mathcal{B}_{aa}=0$ and $\delta\mathcal{B}\mathcal{I}=0$) around the RS solution $n\beta\delta F \sim W^{-3/2}[\mathrm{tr}(-\tau\delta\mathcal{B}^2 + c_2\delta\mathcal{B}^3) + c_3\sum_{a,b}\delta\mathcal{B}_{ab}^4]$, where $\tau = 1 - T/T_c$. This shows that the glass transition in Coulomb glasses belongs to the same universality class as the one in the SK model: the order parameter develops continuously. Hence, many results for infinite range spin glasses [19] should be directly applicable to the present case, providing, e.g., a description of the aging and memory effects observed in experiments [9]. Further, the electron glasses present a testing ground for many theoretical ideas developed for the SK model.

We now turn to a more detailed analysis of the physics far below T_c. Since we expect that $\Sigma_0 \approx \beta\chi^{-1} \gg \beta$ we may expand the free energy (5) in $\beta J/\Sigma$. Using Eqs. (3) we eliminate Σ and obtain $U(\mathcal{B}) = -\mathrm{tr}(\mathcal{B}^3)/12\pi\beta^3$, resembling the SK model where $U(\mathcal{B}) \sim -\mathrm{tr}(\mathcal{B}^2)/\beta^2$. The exponent reflects the spatial dimension $D=3$ and is responsible for the shape of the pseudogap [$\rho(E) \sim E^{D-1}$]. For the following it is more convenient to keep the self-energy in the formalism. Let us suppose that the replica symmetry is broken at the level of K steps. We represent the Parisi matrices as $\Sigma = -\Sigma_0 + \sum_{k=1}^{K}\Sigma_k R_{m_k}$, where R_{m_k} consist of blocks of size m_k on the diagonal with all entries equal to 1. Let us focus on the set C of the m_1 pseudospins corresponding to one of the innermost blocks. They experience an effective field y created by all other replicas. We describe its fluctuations by a distribution $P(y)$, which was a simple Gaussian in the RS case [see Eq. (6)]. We note that $P(y)$ corresponds to the distribution of *pseudoequilibrium* local fields y_i defined by $\langle s_i\rangle = m_i = \tanh(\beta y_i/2)/2$ on the original lattice [20]. $P(y)$ is a functional of $\mathcal{B}$ which could be obtained by integration of Parisi's differential equation using the methods of Ref. [20]. Here, we exploit only the well-known fact that a full RSB glass is in a marginally stable state at all $T<T_c$ [19,20] (i.e., the Hessian $\partial^2 F/\partial\mathcal{B}^2$ has a vanishing eigenvalue in the replicon mode $\delta\mathcal{B}$ characterized by $\delta\mathcal{B}_{aa}=0$ and $\delta\mathcal{B}R_{m_k}=0$ for all k). This imposes the following constraint on $P(y)$:

$$\int_{-\infty}^{\infty} dy P(y)\frac{1}{[2\cosh(\beta y/2)]^4} = \frac{1}{g_1^{-2}(\Sigma_0) - g_2^{-1}(\Sigma_0)}. \quad (7)$$

Further, the innermost component of Eqs. (3) read

$$\chi \equiv \beta\left[\frac{1}{4} - \langle s_a s_b\rangle_{a\neq b\in C}\right] = \beta g_1(\Sigma_0) = \beta\int_{-\infty}^{\infty} dy P(y)\frac{1}{[2\cosh(\beta y/2)]^2}. \quad (8)$$

Expanding g_n for $\Sigma_0/\beta \gg 1$, one can see that at low temperatures these two equations admit a solution only if $\Sigma_0 \sim \beta^3$ and $P(y)$ takes the scaling form

$$T^{-2}P(y \equiv zT) \to p(z) \qquad (T\to 0), \quad (9)$$

with $p(z) \sim z^2$ for $z \gg 1$. This implies that the susceptibility obeys the scaling $\chi \sim T^2$, and the (static) screening length diverges at low temperatures as $l_{\mathrm{sc}} = (4\pi\chi)^{-1/2} \sim T^{-1}$. Note that χ is associated with the *pseudoequilibrium* local fields y_i that account for relaxations of other charges within the local free energy minimum. While we expect χ to control the hopping conductivity, tunneling experiments [5] probe the system on short time scales, sampling the distribution $\tilde{P}(h)$ of *instantaneous* local fields $h_i = \sum_j J_{ij}s_j$. The *thermal average* of these fields, $\langle h_i\rangle = \sum_j J_{ij}m_j$, is related to the field y_i via a Thouless-Anderson-Palmer (TAP) equation [21], $\langle h_i\rangle = y_i + \langle s_i\rangle h_O$, where the Onsager term

$$h_O = \beta\int_k \frac{J_k^2}{\beta J_k + \Sigma_0} \approx 2\sqrt{\pi\beta/\Sigma_0} \approx 2\sqrt{\pi\chi} \quad (10)$$

accounts for the extra polarizations induced by the presence of the charge $\langle s_i\rangle$. For consistency with the locator approximation, we have retained only terms corresponding to a local self-energy. The deviation of the local field h_i from its mean $\langle h_i\rangle$ is essentially a Gaussian variable with width h_O. More precisely, the relation

$$\tilde{P}(h) = \int dy P(y)\frac{\cosh(\beta h/2)}{\cosh(\beta y/2)}\frac{e^{-\beta(h-y)^2/2h_O}}{\sqrt{2\pi h_O/\beta}e^{-\beta h_O/8}} \quad (11)$$

holds [17], which generalizes a known result for the SK model [22]. The tunneling density of states at zero bias then follows from $\nu_0 = \beta\int dh\tilde{P}(h)[2\cosh(\beta h/2)]^{-2}$. Equation (11) implies that $\tilde{P}(h)$ obeys a scaling analogous to Eq. (9), and hence $\nu_0 \sim T^2$. Generally, in order to make quantitative predictions, one needs to know the functional form of the field distributions. It turns out, however, that certain parameters are not very sensitive to their details. It is convenient to assume a simple form $\overline{P}(\langle h\rangle) = \alpha(\langle h\rangle^2 + \gamma T^2)$ for the distribution of average fields, obtain $P(y)$ via the TAP equations, and solve Eqs. (7) and (8). This yields χ, ν_0, and α as slowly varying functions of γ [23]: $\alpha \approx 0.204 - 0.0067\gamma$, $\chi \approx (22.27 - 0.81\gamma)T^2$, $\nu_0 \approx (2.178 - 0.008\gamma)T^2$. The tunneling DOS ν_0 is roughly an order of magnitude smaller than the full susceptibility χ, as is also evident from the typical distributions shown in Fig. 2. This agrees well with the experimental observation [11] that the susceptibilities governing tunneling and hopping transport differ significantly. The value of α should be compared to the Efros-Shklovskii prediction $\alpha_{\mathrm{ES}} = 3/\pi \approx 0.95$ [3] which is larger than our estimate because their self-consistency argument imposes stability only with respect to single electron hops. By contrast, our estimate includes multiparticle constraints that decrease α

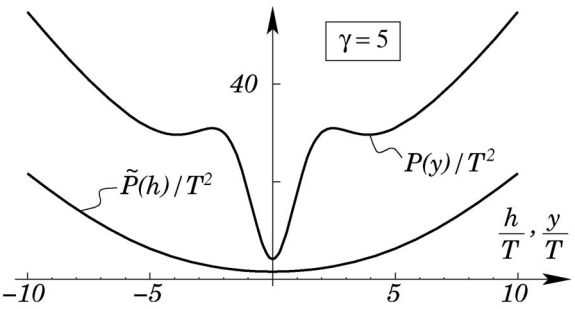

FIG. 2. The distributions $\tilde{P}(h)$ and $P(y)$ of the instantaneous and pseudoequilibrium fields, respectively. Since the latter include the relaxation of the environment, the gap is narrower.

below α_{ES} in agreement with large-scale numerical simulations [24].

In conclusion, we have developed the locator approximation for Coulomb glasses, allowing us to include multiparticle correlations. We have used this formalism to provide evidence for a continuous glass transition below which the Coulomb glass gets stuck in a marginally stable state, resulting in subexponential relaxation dynamics and the universal Coulomb gap at low temperature. Formally, the locator approximation is justified for large disorder. However, in the absence of crystallization, a structural glass transition provides sufficient self-generated disorder, so that we expect our results to hold at low temperatures even in the case of weak external disorder. We verified [17] that in this limit the local observables are controlled by large scales and reveal the Efros-Shklovskii gap. Further, as noticed in [18], the locator approximation gives a significant decrease of the DOS with temperature already above T_c, in agreement with numerics. Moreover, it predicts a discontinuous glass transition at a scale of $T_c \approx 0.030$ which depends, however, on the details of the cutoff at small scales. The validity of this prediction remains unclear.

The locator approximation not only provides new insight into classical Coulomb glasses but also allows for quantitatively new predictions that go beyond the single particle theory, setting the stage for further developments of the theory of correlated transport and glassy relaxation in these systems. For instance, it allows one to study the collective modes of electrons that induce fluctuations in the local electric fields [10] and thus allow resonant tunneling in the absence of phonons. This provides an alternative mechanism for electron transport as observed at low temperatures when phonons freeze out [25]. Finally, extensions of the locator approximation to describe quantum electron glasses [12,26] may be envisioned.

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