Critical Behavior of the Coulomb Glass

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We have performed a Monte Carlo study of a classical three dimensional Coulomb glass. We use finite size scaling to demonstrate the existence of a spin-glass-like phase transition and determine its critical exponents. We find that the transition temperature is well below the bare energy scale; we attribute this to pairs of nearby sites binding into weakly interacting effective dipoles. We have also studied the density of states as a function of temperature; we find that the Coulomb gap begins to develop well above the transition temperature and is nearly fully formed at T_c.

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The competition between disorder and interactions arises in a variety of systems. In the case of doped semiconductors, the disorder is produced by the random placement of donor and acceptor sites. As a result, electrons occupy sites in a random environment, and interact with one another via a long-range Coulomb potential [1]. It has been presumed that classically such a system undergoes a phase transition analogous to the spin-glass transition [2,3]. However, the nature of the transition has not been determined, and in fact, there has been no direct demonstration of a finite temperature transition. The features which have been seen in previous simulations, such as a broad peak in the specific heat and an increase in the nonlinear susceptibility, have not been sufficient to establish the existence of a finite temperature transition.

In this Letter we present the results of a Monte Carlo study of the Coulomb glass in three dimensions, in which we demonstrate for the first time the existence of a phase transition and determine its critical exponents. We also monitor the formation of the Coulomb gap [4] in the single particle density of states as the temperature is lowered. Even though the gap begins to develop at temperatures on the order of the characteristic interaction energy, we find that the phase transition occurs at a much lower temperature; we attribute this to the pairing of nearby sites into weakly interacting effective dipoles.

The essential physics of the Coulomb glass is the presence both of disorder and of long-range Coulomb interactions between electrons. The Hamiltonian often studied for the Coulomb glass is [2,5]

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

where we set the charge $e=1$, n_i is the number operator for site i, ϕ_i is a random onsite energy, $r_{ij}=|r_i-r_j|$, and K is a compensating background charge making the whole system charge neutral. Such a Hamiltonian describes a lightly doped semiconductor, in which the impurity sites are far enough apart that the overlap between sites can be neglected. In formulating simple theoretical models of this system, there are different ways of introducing the randomness; for the purposes of our paper, we feel that the exact nature of the disorder is not so crucial. In most of the early work on the Coulomb glass (e.g., Refs. [2,3,5]), the sites are chosen to form a periodic lattice, and the disorder is present in the form of random onsite energies.

However, the presence of random onsite energies makes numerical analysis difficult, since even in the high temperature state the average occupation of a site is not zero. This makes the search for a phase transition difficult; there is no obvious order parameter which becomes nonzero at the transition. For our numerical analysis, it is more convenient to take the disorder to be entirely in the location of the sites. Although this changes the symmetry of the Hamiltonian, we expect the physics to be qualitatively the same. We note that a previous simulation in two dimensions also has taken this form for the Hamiltonian [6]. In the case of half filling there is a particle-hole symmetry, and we expect a phase transition to be associated with the development of a nonzero Edwards-Anderson order parameter. We therefore rewrite the Hamiltonian (taking $K=\frac{1}{2}$) to look like that of a spin glass,

$$\mathcal{H}=\frac{1}{4}\sum_{i>j}\frac{S_iS_j}{r_{ij}}\,. \tag{2}$$

$S_i=1$ (-1) will denote an occupied (unoccupied) site.

We have simulated three dimensional systems of linear size $L=4$, 6, and 8. We randomly place $N\equiv L^3$ sites in the system; the position of each site is independently chosen uniformly over the volume. We use periodic boundary conditions, in which the distance between each pair of sites is taken to be the minimum distance between them in the full repeated lattice. We have only considered the case of half filling, in order to take advantage of the spin-flip symmetry.

We have used a Monte Carlo heat bath algorithm. We keep a table of the potential energy at each site. Each electron is looked at sequentially and moved to one of the available $N/2+1$ sites (its own site or one of the $N/2$ unoccupied sites), chosen with a Boltzmann probability. If the site chosen is the electron's original location, the

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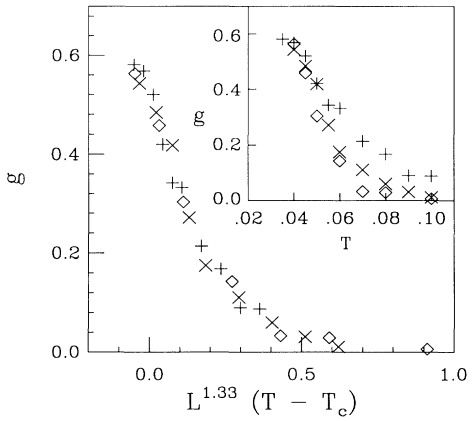

FIG. 1. $g(L,T)$ scaled using $\hat{g}(L^{1/\nu}(T-T_c))$, $T_c=0.043$, and $\nu=0.75$, for systems of size 4 (+), 6 (×), and 8 (◇). The inset is the unscaled $g(L,T)$.

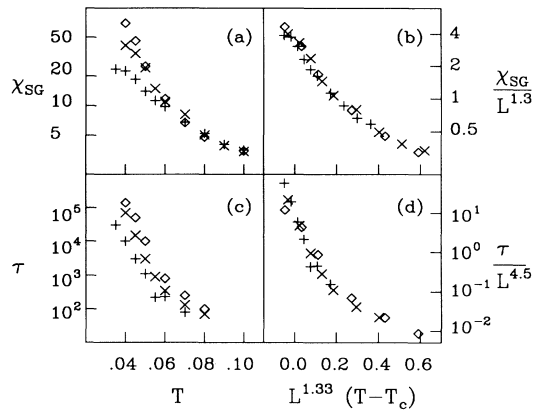

FIG. 2. (a) $\chi_{SG}(L,T)$. (b) χ_{SG} scaled using $L^{2-\eta}\times\hat{\chi}(L^{1/\nu}(T-T_c))$ with $\eta=0.7$. (c) $\tau(L,T)$. (d) τ scaled using $L^z\hat{\tau}(L^{1/\nu}(T-T_c))$ with $z=4.5$. Symbols denote systems of size 4 (+), 6 (×), and 8 (◇).

potential energies are unchanged; if the electron decides to hop to a new site, we update all the potential energies. If the electron chooses its initial site, which it does with high probability at low temperatures, we do not have to recompute the fields. This speeds up the simulation considerably, partially compensating for the much longer equilibration times needed at low temperatures.

The Edwards-Anderson order parameter alluded to above is defined as $q\equiv[\langle S_i\rangle^2]$; we will denote thermal averages by $\langle\cdots\rangle$ and disorder averages by $[\cdots]$ [7]. The spin-glass susceptibility is defined as

$$\chi_{SG}\equiv\frac{1}{N}\sum_{ij}[\langle S_iS_j\rangle^2]=[N\langle q^2\rangle]\,.$$

For an infinite system χ_{SG} diverges at the phase transition as $\chi_{SG}\sim(T-T_c)^{-\gamma}$; the scaling hypothesis implies that χ_{SG} satisfies the finite size scaling form $\chi_{SG}=L^{2-\eta}\times\hat{\chi}(L^{1/\nu}(T-T_c))$ [using the scaling relation $\gamma=(2-\eta)\nu$]. Another parameter related to the transition is $g\equiv\frac{1}{2}(3-\langle q^4\rangle/\langle q^2\rangle^2)$. At high temperatures, the distribution of q becomes Gaussian and g tends to 0, whereas at low temperatures g becomes nonzero. Since g is dimensionless, we expect that it should satisfy a scaling form $g(L,T)=\hat{g}(L^{1/\nu}(T-T_c))$, and thus at the critical temperature, $g(L,T_c)$ should have the same value independent of the system size L (as long as L is sufficiently large for finite size scaling to apply) [8,9].

We have monitored equilibration by simulating two replicas which have the same placement of sites but different spin configurations [9]. We can measure q either by computing overlaps between the two replicas, $q_r=(1/N)\sum_i S_i^{(1)}S_i^{(2)}$, or we can use the same replica at different times,

$$q_t(t,t')=\frac{1}{N}\sum_i S_i^{(1)}(t)S_i^{(1)}(t')\,. \tag{3}$$

If the time difference $t'-t$ is sufficiently large that the configurations are essentially uncorrelated, this will give the same result as the replica overlap. The value of χ_{SG} determined by the replica overlap increases with time to the equilibrium value, whereas the value given by the time overlap decreases to the equilibrium value. The two methods agree when the system has reached equilibrium. Our longest run (for $L=8$ at $T=0.04$) had 262000 Monte Carlo steps per electron, although most of the runs were much shorter. Depending on system size and temperature, the sample averages involved between 20 and 160 disorder configurations.

We have determined the critical exponents of the phase transition through finite size scaling [9]. In the inset of Fig. 1, we show $g(L,T)$ for $L=4$, 6, and 8. We note that the curves for all three sizes cross at a value near $T=0.043$, showing the validity of the scaling hypothesis. In Fig. 1, we show a scaling collapse of these data, using $T_c=0.043$ and $\nu=0.75$. In Fig. 2(a) we show $\chi_{SG}(L,T)$; 2(b) shows a scaling collapse using the same values of T_c and ν as before, with $\eta=0.7$. We have estimated the errors in the critical temperature and exponents by how well the curves can be made to collapse. Since we are unable to perform simulations much below our estimate of T_c, getting a lower bound on T_c is more difficult than an upper bound, although our data are clearly incompatible with a zero temperature transition. We estimate the uncertainty in the values to be $T_c=0.043^{+0.003}_{-0.006}$, $\nu=0.75^{+0.2}_{-0.1}$, and $\eta=0.7\pm0.2$.

It is interesting that T_c is so much lower than the characteristic energies in the system, which are of order unity. To understand this, note that at the temperatures of our simulation, nearby pairs of sites will with high probability consist of an occupied and an unoccupied site. Since these strongly coupled pairs of sites are close together, they are guaranteed to have small dipole moments. Therefore, they will interact weakly with the rest of the system, remaining active down to temperatures

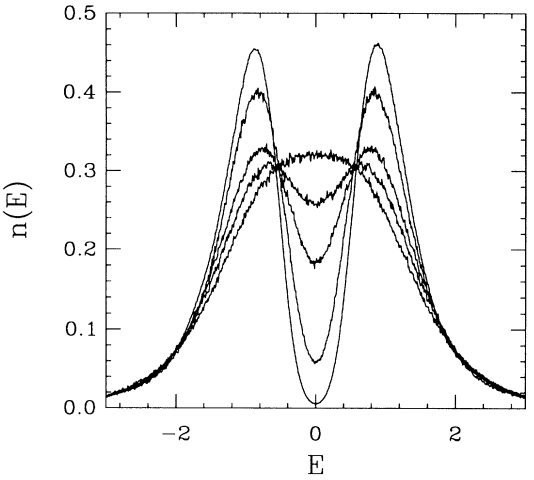

FIG. 3. $n(E)$ for the 6^3 system at temperatures of 0.05, 0.1, 0.2, 0.3, and 0.5. $n(0)$ is decreasing with decreasing temperature.

much lower than the bare interaction energy. To test this hypothesis, we have performed a numerical renormalization procedure in which the most strongly coupled pair of sites is replaced by a single effective spin [10]. We find that after removing all couplings greater than $J_{ij}=1$, for example, the distribution of couplings generated is nearly symmetric about zero with both ferromagnetic and antiferromagnetic bonds. The width of the new distribution is greatly reduced compared to the original width and is on the scale of T_c. This is consistent with our explanation of the reduced transition temperature.

We have also studied how the system relaxes by using a dimensionless parameter $R(t)$ defined by

$$R(t)=\frac{[\langle Q(t)\rangle]}{\sqrt{[\langle Q^2(t)\rangle]}} \quad \text{where} \quad Q(t)=\frac{4}{t}\sum_{t'=3t/4+1}^{t} q_t\left[\frac{t}{2},t'\right]. \tag{4}$$

For $t\to 0$, $R(t)\to 1$, while for $t\to\infty$, $R(t)\to 0$. As discussed in Ref. [11], since $R(t)$ is dimensionless, we expect it to have the scaling form $R(t)=\hat{R}(t/\tau)$, where τ is the relaxation time. Thus we can determine $\tau(L,T)$ by monitoring the time it takes for $R(t)$ to drop to some specified value (we have used both 0.7 and 0.5 for this value, with similar results). This can be used to deduce the dynamic exponent z defined by $\tau\sim\xi^z$. In Fig. 2(c) we show $\tau(L,T)$; Fig. 2(d) shows τ scaled onto one curve, using a scaling form $\tau(L,T)=L^z\hat{\tau}(L^{1/\nu}(T-T_c))$, with $z=4.5$. We estimate $z=4.5^{+0.4}_{-0.8}$. Since our heat bath algorithm is nonlocal (recall that an electron can jump to any available site in the whole system), it is likely that a local algorithm would give a larger value for z, since if electrons are allowed only local moves, it would presumably take much longer to relax locally unfavorable regions.

We have also calculated the specific heat and we find that, as in spin glasses, there is fairly broad bump. This has a maximum around $T=0.07$, a temperature somewhat higher than the transition temperature. Above $T=0.07$, there is a slow decrease in the specific heat with increasing temperature, and below the maximum the specific heat decreases roughly linearly with decreasing temperatures, although we did not measure the specific heat at very low temperatures.

In Fig. 3, we show the density of states for single particle excitations $n(E)$ at various temperatures. Because of strong electron-electron correlations, the density of states at zero energy starts to decrease at $T\sim 0.4$, almost an order of magnitude higher than the transition temperature. By $T=0.05$, still above the transition temperature, $n(0)$ has decreased by a factor of more than 50. We find that for moderately low temperatures, but above the transition temperature, $n(0)$ decreases roughly linearly with temperature, and extrapolates to a gap of zero at roughly the transition temperature. [We have not measured $n(0)$ below the transition temperature, although at sufficiently low temperatures $n(0)$ should vary quadratically with temperature [12].] This is in qualitative agreement with capacitance experiments which have indirectly measured $n(0)$ versus T [13]. To the best of our knowledge, tunneling experiments have not looked at the density of states as a function of temperature in three dimensions [14].

An indirect way of monitoring the development of electron-electron correlations comes from recent experiments [15], which looked at the inhomogeneously broadened linewidth $\sigma(T)$ of the $1s\to 2p_0$ transition at neutral donor sites in GaAs. The linewidth is essentially temperature independent both below ≈ 4 K and above ≈ 8 K, and decreases from the high temperature value to the low temperature value as the temperature is lowered. The linewidth is primarily broadened due to the distribution of gradients in the electric field at the donor sites, and the decrease in the linewidth has been attributed [16] to the development of correlations among the occupied sites.

In an effort to explain these experiments, Baranovskiĭ, Thomas, and Vaupel [16] performed simulations using a Hamiltonian similar to Eq. (1), but which had explicit donor and acceptor sites. They looked at the distribution of electric field gradients at unoccupied donor sites, and saw that the width of this distribution decreased with temperature as in the experiments, but unlike the experiments, the width did not level off at low temperatures. These simulations, however, were all performed at temperatures which we believe are above T_c (although since they were simulating a somewhat different model, their temperatures cannot be compared directly with our simulation).

For our simulation, a corresponding quantity is the electric field gradient at an occupied site due to the other occupied sites,

$$V_{zz}(j)=\sum_{i\neq j}\frac{1}{2}(S_i+1)\frac{\partial^2}{\partial z^2}\left(\frac{1}{r_{ij}}\right), \quad (5)$$

for sites j where $S_j=1$. We expect the distribution of V_{zz} to narrow as the temperature is lowered, since sites will be occupied randomly at high temperatures, but at lower temperatures it is unlikely that two nearby sites will both be occupied. Indeed, we find that the width $\sigma(T)$ of the distribution of V_{zz} decreases as the temperature is lowered, and begins to flatten out below the phase transition. It is possible then that the low temperature plateau seen experimentally in $\sigma(T)$ is due to an actual Coulomb glass transition. More careful experiments, monitoring the time or frequency dependence of the plateau as well as the development of the Coulomb gap, are necessary to see if it is a nonequilibrium effect or an equilibrium phase transition.

In summary, we have performed simulations studying the three dimensional Coulomb glass transition and have determined its critical exponents. We find $T_c=0.043^{+0.003}_{-0.006}$, $\nu=0.75^{+0.2}_{-0.1}$, $\eta=0.7\pm0.2$, and $z=4.5^{+0.4}_{-0.8}$. Strong correlations are manifested in two ways. First, pairs of nearby sites form weakly interacting dipoles that result in a low transition temperature. Second, the Coulomb gap starts to develop well above T_c.

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