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Evidence for many-electron composite charge excitations in a Coulomb glass

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We present experimental evidence that the elementary charge excitations in a Coulomb glass are composite entities comprised of a correlated motion involving many electrons, rather than the ordinary single-point particles previously believed. The Coulomb interaction energy in localized boron-doped silicon is measured by dc transport and electron tunneling. The transport excitations are found to have a significantly lower Coulomb energy than single-particle point charges introduced by tunneling. This reduction in the transport Coulomb energy is clearly inconsistent with the conventional model of single-particle excitation but agrees semiquantitatively with new theories of many-electron composite excitations.

The influence of strong, long-ranged interactions on a many-particle quantum system remains a central problem in condensed matter and statistical physics. A Coulomb glass, such as a disordered, localized doped semiconductor near its $T=0$ K ground state, is an archetypical example of such a system. When the dopant concentration n is below the critical density n_c that separates insulating and metallic phases, both the disorder from random dopant sites and the electron-electron Coulomb interaction have profound effects on all electronic properties near the ground state. Surprisingly, even in a solid as thoroughly researched as doped bulk silicon, there is still no generally acceptable understanding of the fundamental ground-state and elementary excitation properties in the Coulomb glass state.

A central question, debated for over 20 years, is whether the elementary charge excitations in a Coulomb-correlated localized semiconductor are ordinary single-point particles or large many-body composite particles. In the benchmark theory of Efros and Shklovskii (ES),[1] the Hartree interaction redistributes the localized state energies near the Fermi energy ε_F so that a quadratic [in three dimensions (3D)] Coulomb gap opens in the single-particle density-of-states $N(\varepsilon)$ near ε_F:

$$N(\varepsilon)=\frac{3}{\pi}\left(\frac{\kappa}{e^2}\right)^3\varepsilon^2, \tag{1}$$

where κ is the dielectric constant, e is the electron charge, and we have defined $\varepsilon_F=0$. This quadratic gap persists up to an energy Δ where $N(\Delta)=N_0$, the noninteracting value of the density of states, giving a Coulomb gap width $\Delta=e^3(N_0/\kappa^3)^{1/2}$. This Coulomb gap subsumes all effects of the interaction; the charge excitations are assumed to be single electronlike or holelike point particles hopping independently among these renormalized states. From this assumption and Eq. (1), the variable-range hopping resistivity ρ as $T\to 0$ takes the form:

$$\ln\rho\propto(T_0/T)^{1/2}, \tag{2}$$

where T_0, a measure of the characteristic Hartree interaction strength, is given by

$$T_0=C\frac{1}{k_B}\frac{e^2}{\kappa\xi}, \tag{3}$$

where ξ is the localization length and C is a numerical constant. In the ES theory, $C=2.8$ (in 3D). Experiments on many disordered electronic solids[2] show a $\rho(T)$ obeying Eq. (2). Because of such measurements, the single-particle ES picture has been the most widely accepted description of charge excitations in a Coulomb glass system.

Through the 1980s, several theories[3–5] challenged the ES single-particle assumption. These works adopted a dynamical picture in which any charge hop from an initial to a final site changes the local potentials of all nearby sites, altering the occupancy of those states. In these models, current is composed of spatially large composite excitations formed by the correlated motion of many charges; in the literature this has been called a charge polaron or a composite quasiparticle. However, these early efforts failed to generate a $\rho(T)$ compatible with experiment. As a result, the applicability of many-particle excitation models was not generally accepted.

Recently, the theory of many-particle excitations was revisited by Pérez-Garrido *et al.*,[6] Meir,[7] and Lamba and Kumar.[8] Reference 6 introduced a calculation where dc current flow is carried by sequential reconfigurations of site occupancies involving correlated motion of many charges in a Coulomb glass. Simulations generated a hopping resistivity the same as Eq. (2) for such many-electron excitations. In Ref. 7, analytical calculations of the Coulomb gap were made assuming the net interaction is between large composite quasiparticles, rather than between single-point particles. This also yielded the hopping form of Eq. (2). In contrast, the linear response calculations of Ref. 8 concluded that the relaxation time for other charges to respond to the hop of a single-point charge is so slow that the background distribution of charges is effectively frozen, keeping intact the single-particle ES picture. While both single- and many-particle theories now explicitly predict Eq. (2), there is one

major physical difference between the two models. A reconfiguration or relaxation involving many charges will reduce the net Hartree energy difference between initial and final many-body states of a charge hop, compared to an independent single-particle hop. Therefore, the Hartree energy as measured by T_0 should be smaller for many-electron excitations compared to the single-particle ES theory. In fact, Refs. 6 and 7 estimated that T_0 for many-electron excitations should be roughly an order of magnitude smaller than the ES value.

The experimental consequence of these recent theoretical works is that previous experiments[2] simply showing a $\rho(T)$ that follows Eq. (2) can no longer be interpreted as evidence for single-particle excitations. The distinction between single- and many-particle excitations must rely on measuring the Coulomb interaction energy through a quantity related to T_0, and then comparing with the single-particle value. T_0 is easily measured by fitting $\rho(T)$ data to Eq. (2). However, obtaining only the transport T_0 is insufficient because the single-particle ES value, to which the transport T_0 must be compared, requires independent knowledge of at least one other parameter, such as the product $\kappa\xi$ in Eq. (3), which is usually difficult to determine reliably. The best way to delineate between single- and many-particle excitations is to measure the interaction energy in both a transport process and a known single-particle excitation process.

In this paper, we present a set of experimental data that clearly distinguishes single-particle and many-particle excitations in the Coulomb glass state of boron-doped silicon (Si:B). We accomplish this by comparing transport measurements of T_0 to the purely single-particle interaction energy obtained by electron tunneling. In tunneling, a point-particle electron is the excitation injected into the system through a classically forbidden potential barrier, yielding the single-particle density of states[9] $N(\varepsilon)$ used in the ES theory. The tunneling process is physically distinct from dc transport, in which an applied electric field excites a dressed quasiparticle excitation from just below to just above ε_F. Only in the noninteracting case are the excitations necessarily the same in both measurements, so that the same noninteracting density of states N_0 governs both processes. In the presence of strong interactions, tunneling and dc transport involve different densities of states with possibly different interaction energy scales.

Three energy scales relevant to hopping conductors are measured independently. At relatively high temperatures the system is noninteracting because thermal energy overcomes the Coulomb correlations. Here the Mott temperature $T_M = 18/k_B N_0 \xi^3$ can be extracted using the expression for noninteracting variable range hopping transport:[10]

$$\ln \rho \propto (T_M/T)^{1/4}. \tag{4}$$

Since T_M is a property of the system without interactions, its value is independent of whether interactions result in single- or many-particle excitations at lower temperatures. The Hartree correlation temperature T_0 is obtained from fitting Eq. (2) in the low-temperature interacting regime. The single-particle Coulomb gap width Δ is measured directly by electron tunneling spectroscopy. Since both T_0 and Δ measure the Hartree interaction strength, they are related and can be expressed in terms of each other by eliminating κ and ξ from the definitions of T_0 [Eq. (3)], Δ, and T_m given above, resulting in

$$\Delta = B k_B (T_0^3/T_M)^{1/2}, \tag{5}$$

where $B = (18)^{1/2}/C^{3/2}$, where C is the constant in Eq. (3). Thus Eq. (5) makes possible a direct comparison between the measured transport and tunneling interaction energies.

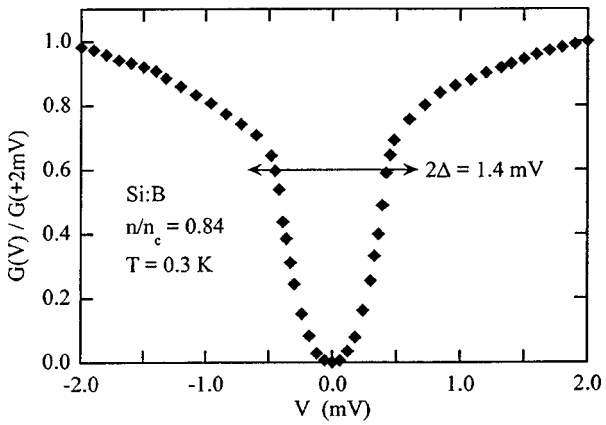

FIG. 1. Tunneling conductance spectrum $G(V)$ between ± 2 mV bias taken on a Si:B crystal doped at $n/n_c = 0.84$. The spectrum is normalized to $G(+2\ \text{mV})$. The temperature is 0.3 K.

Bulk single-crystal samples of Si:B with dopant density $n/n_c = 0.84$ were prepared for electron tunneling and dc resistivity measurements. Measurements reported in the literature[11,12] on doped silicon crystals within a few percent of this density show that the Coulomb glass state sets in at temperatures below ~ 1 K, making it convenient to observe the full transition from noninteracting to interacting behavior. Samples with higher density do not show interaction effects until a significantly lower temperature and have physics complicated by the approaching phase transition to the metallic state.[13] Conversely, samples of significantly lower density are impractical because the strongly diverging ρ and $\partial\rho/\partial T$ of very insulating samples makes it extremely difficult to maintain the bias conditions and temperature stability required to achieve a reasonable signal-to-noise tunneling spectrum at low temperatures. Details of the processing and characterization have been described previously.[12] The tunnel junctions were formed by growing an ultrathin (estimated 1.5 to 2 nm) SiO_2 potential barrier on the front surface of the crystal samples, and then depositing a thin film of Al as the tunnel junction counterelectrode. Each sample also had four cryogenically compatible Ohmic contacts fabricated on the back surface, so that both tunneling and dc transport could be measured on the same crystal.

Figure 1 shows the low-bias tunneling conductance spectrum $G(V)$ taken on a Si:B crystal at a temperature of 0.3 K. A 1 kG magnetic field suppressed the superconductivity in the Al electrode, so that all nonlinear features in $G(V)$ on this energy scale are features of the Si:B single-particle density of states. Ordinary thermal broadening is insignificant on the scale of the main features of the spectrum at this low temperature, so the data can be treated as proportional to the single-particle density-of-states $N(eV)$. The prominent feature is the smooth dip in conductance around $V=0$, which

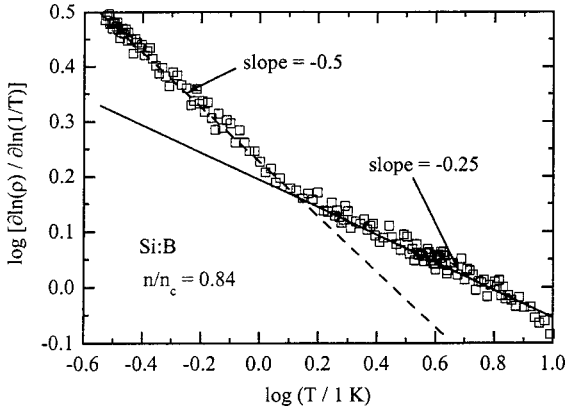

FIG. 2. Plot of $\log[\partial \ln \rho / \partial \ln(1/T)]$ vs $\log T$. The slope of a linear fit to the data in a temperature range gives the negative of the hopping exponent. The solid line is a linear fit to the range 1.5 K $<T<9$ K. The dashed line is a linear fit to the range 0.3 K$<T<1$ K.

defines ε_F in tunneling experiments. This dip is the single-particle Coulomb gap in $N(\varepsilon)$, and can be fit to a power law, $G(V) \propto |V|^{2.3}$, close to the quadratic dependence of Eq. (1). From this spectrum, the Coulomb gap width can be directly measured. The shoulder-to-shoulder full width of the gap, marked in Fig. 1, gives a value $2\Delta = 1.4 \pm 0.2$ meV for the single-particle Coulomb energy scale measured by tunneling.

The hopping temperatures T_M and T_0 for this same Si:B crystal can be obtained from low-temperature transport data. First, the temperature regimes for Mott and ES-type hopping must be established definitively. Using the hopping resistivity form $\ln \rho = (A/T)^{\upsilon}$ for a constant A, the hopping exponent υ can be determined without further assumptions by noting that

$$\ln\left[\frac{\partial \ln \rho}{\partial \ln(1/T)}\right] = \ln(\upsilon A^{\upsilon}) - \upsilon \ln(T). \tag{6}$$

So when plotting the data as $\partial \ln(\rho)/\partial \ln(1/T)$ as a function of T on a log-log plot, the slope of a linear fit over some temperature range gives $-\upsilon$.[14] This is shown in Fig. 2. Even though the temperature range 0.3–10 K is not especially large, there are clearly two different hopping regimes. From ~1.3 through 10 K the data are well fit to a line with slope of -0.25, indicative of noninteracting Mott hopping. Below ~1 K, the data are best fit to a slope of -0.5, the expected value for ES-type hopping.

Fits of the $\rho(T)$ data to obtain T_M and T_0 are shown in Fig. 3. In Fig. 3(a), resistivity data in the Mott hopping regime are plotted on a logarithmic scale against $T^{-1/4}$. On such a plot, noninteracting Mott hopping of Eq. (4) appears as a straight line. The slope of a linear fit gives $T_M = 1480 \pm 20$ K. Figure 3(b) shows the resistivity data in the ES hopping regime plotted logarithmically against $T^{-1/2}$. Here the variable-range hopping form of Eq. (2) is represented by a straight line, which reasonably fits the data through this temperature range. The slope of the fit gives $T_0 = 12 \pm 1$ K.

The interaction energies T_0 from transport and Δ from tunneling on the same sample can now be compared using Eq. (5). Inserting the single-particle values $\Delta = 0.7$ meV and $C = 2.8$ into Eq. (5) yields a single-particle (sp) correlation

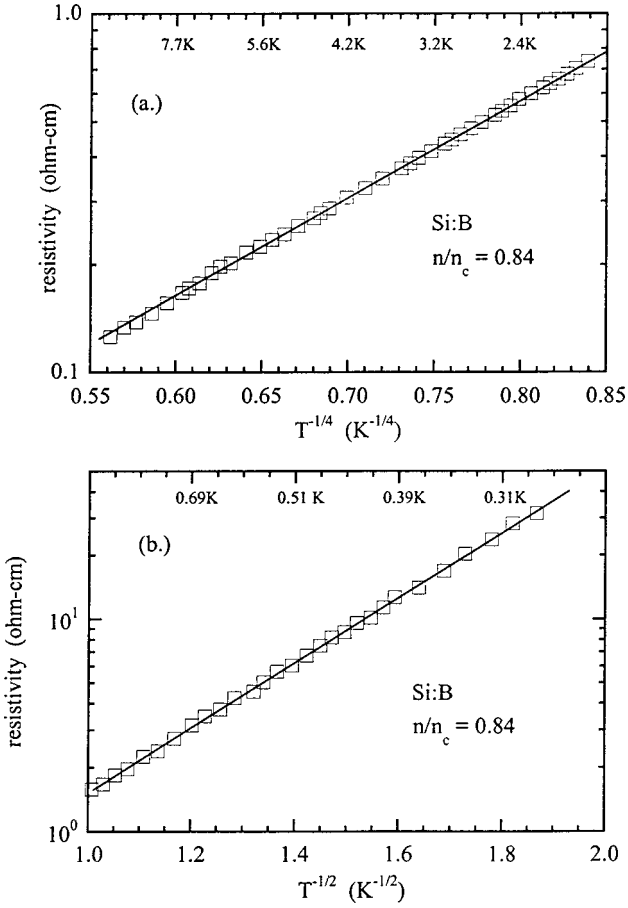

FIG. 3. (a) Resistivity of a Si:B crystal (the same crystal used to generate the data of Fig. 1) plotted on a logarithmic scale against $T^{1/4}$ over a temperature range 10 K$>T>2$ K. The line is a fit to Eq. (4), the slope of which gives $T_M = 1480$ K. (b) Resistivity of the same sample plotted logarithmically against $T^{1/2}$ over a lower temperature range (1 K$>T>0.3$ K). The line is a fit to Eq. (2), the slope of which gives $T_0 = 12$ K.

temperature $T_0^{\mathrm{sp}} = 49$ K. This is the Hartree interaction energy that should enter into Eq. (2) if single-particle excitations carry the transport current. However, this value is over four times larger than the $T_0 = 12$ K actually measured by transport and is much too large to be consistent with the ES single-particle transport theory. In fact, the ratio $T_0/T_0^{\mathrm{sp}} = 0.25$ is consistent with the roughly order-of-magnitude reduction in Hartree energy obtained in the numerical simulations of Pérez-Garrido *et al.*[6] In addition, for a sample very similar to the one used in this paper, Meir's calculation[7] of composite quasiparticle transport results in a value of $T_0 = 10$ K, in close agreement with the measured value but much smaller than the single-particle result.

The existence of many-particle excitations can resolve a puzzling anomaly in the literature. Roy *et al.*[15] first noted that values of T_0 for many different semiconductors[16–19] obtained by fitting Eq. (2) to variable-range hopping resistivities would, using the ES form of Eq. (3), imply an anomalously long localization length $\xi > R$, the mean hopping radius (taking κ to be known and fixed by the host lattice material). This is anomalous because variable-range hopping conduction occurs only when charges seek out energetically accessible impurity states by hopping distances beyond ξ,

making $R>\xi$ a requirement for Eq. (2) to describe $\rho(T)$. The resolution of this anomaly has not been clarified either experimentally or theoretically until now. This discrepancy can be explained by our findings. A large ξ was used to correct the T_0 of Eq. (3) to accommodate a smaller measured T_0. As demonstrated above, a small transport T_0 is the result of a reduction in the Hartree energy magnitude by many-particle excitations. Therefore, rather than making ξ unusually large, the proper adjustment is to make C in Eq. (3) smaller than the ES value. Reducing C by a factor of ~ 4 returns ξ to sensible values in most of the literature cited. This factor is fully consistent with the data and conclusions presented here.

In summary, the many-particle nature of elementary charge excitations in a Coulomb glass have been experimentally demonstrated. The Hartree energy measured by transport is much smaller than the energy measured by single-particle tunneling. This reflects the reduction in interaction energy resulting from many-particle excitations. The magnitude of the transport Hartree energy agrees semiquantitatively with recent theories of many-particle composite excitations.

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