Microwave ac Conductivity Spectrum of a Coulomb Glass

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We report the first observation of the transition between interacting and noninteracting behavior in the ac conductivity spectrum $\sigma(\omega)$ of a doped semiconductor in its Coulomb glass state near $T = 0$ K. The transition manifests itself as a crossover from approximately linear frequency dependence below $\sim$10 GHz, to quadratic dependence above $\sim$15 GHz. The sharpness of the transition and the magnitude of the crossover frequency strongly suggest that the transition is driven by photon-induced excitations across the Coulomb gap, in contrast to existing theoretical descriptions.

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A spatially random arrangement of localized charges becomes a Coulomb glass when the long-range Coulomb interaction energy between charges exceeds the thermal energy. Coulomb glasses are archetypes of the strongly interacting many-particle problem at the forefront of solid-state and statistical physics. Coulomb glasses can display a rich variety of many-body physics, including slow relaxation dynamics [1], and a ground state and excitation structure dominated by an interplay between disorder and strong quantum correlations [2]. Crystalline semiconductors doped at a density n below the critical density n_c of a metal-insulator transition are a common realization of a Coulomb glass. Several recent experiments [3,4] probing interactions in various Coulomb glass materials have revealed that the transition from interacting to noninteracting behavior and the influence of interactions as the metal-insulator phase transition is approached are complicated problems that remain inadequately described by existing theory.

The Coulomb glass is nonmetallic and so has zero dc electrical conductivity at $T = 0$ K. However, it can have significant ac conductivity at $T = 0$ K by photon, rather than phonon, assisted hopping among localized states. Using a pair approximation, in which charge transitions occur between pairs of occupied and unoccupied sites separated in energy by a photon resonance $\hbar\omega$ and separated in space by a mean distance $r_\omega = \xi \ln(2I_0/\omega)$, where ξ is the localization length and I_0 is the hopping attempt rate, Shklovskii and Efros [5] calculated the real part of the ac conductivity $\sigma(\omega)$ of a Coulomb glass at $T = 0$ K:

$$\sigma(\omega) = \frac{\pi^2}{3}\frac{e^2}{\hbar} N_0^2 \xi^5 \ln^4(2I_0/\omega)\hbar\omega[\hbar\omega + U(r_\omega)], \quad (1)$$

where N_0 is the noninteracting density of states at the Fermi level, $U(r_\omega) = e^2/\kappa r_\omega$ is the mean Hartree energy between two sites forming a resonant pair, and κ is the dielectric constant. I_0 is generally taken to be between 10^{12} to 10^{14} s^{-1} in localized semiconductors. In the noninteracting limit $U(r_\omega) = 0$, Eq. (1) reproduces an earlier calculation by Mott [6].

Apart from logarithmic corrections, Eq. (1) predicts a transition from linear frequency dependence in the interacting regime when $\hbar\omega \ll U(r_\omega)$ to quadratic behavior (the Mott result) when $\hbar\omega \gg U(r_\omega)$. Equation (1) assumes that the states forming a pair have small enough average spatial separation so that $U(r_\omega) > 2\Delta$, the full width of the "soft" Coulomb correlation gap [7], so that only states outside the gap edge contribute to $\sigma(\omega)$. The situation for pairs deep inside the gap edge, i.e., $U(r_\omega) \ll 2\Delta$, differs from Eq. (1) only by logarithmic frequency factors [5]. In particular, Δ does not explicitly enter into the description in either limit.

The range of validity of Eq. (1) is open to question. First, it is unclear how well Eq. (1) works in the important and experimentally relevant case when the state pairs are near the gap edge, i.e., $U(r_\omega) \approx 2\Delta$. Second, it is believed [8] that the pair approximation generates an unreliable number of logarithmic factors in Eq. (1) as n approaches n_c. The linear and quadratic frequency factors, however, are believed to be robust even in more general cluster approximations that apply closer to n_c [9]. In practice, logarithmic corrections to power-law dependencies are difficult to measure accurately. Thus it is conventional in the experimental literature to write $\sigma(\omega) \propto \omega^s$ with $s \sim 1$ in the interacting case at low frequency, and $s \sim 2$ in the noninteracting case at high frequency. Logarithmic corrections appear as deviations from exact linear and quadratic behavior.

The $T = 0$ K or "zero-phonon" result assumes the absence of thermal phonon-driven hopping processes. Equation (1) should remain valid at nonzero temperatures as long as the inelastic hopping is dominated by photon, rather than thermal, activation. Thus we expect Eq. (1) should be temperature independent to leading order for $T > 0$ K as long as $kT \ll \hbar\omega$.

There are few reported measurements of zero-phonon ac conductivity in a Coulomb glass, and none conclusively support or refute Eq. (1). As reviewed by Castner [10], the main difficulty is the required combination of a low enough kT to be in the zero-phonon Coulomb glass state, i.e., $kT < \hbar\omega$ and $kT < \Delta$ which realistically means $T < 0.1$ K in most doped semiconductors, and a broad enough frequency bandwidth to cover $\hbar\omega \sim 2\Delta$, which practically means tens of GHz. Experiments on Si:P [11,12] and Si:As [13] deep into the Coulomb glass state ($T < 0.05$ K) have been limited to a small bandwidth ($\omega/2\pi < 400$ MHz) where the zero-phonon assumption is marginal and the transition to noninteracting behavior cannot be reached. These works found a sublinear frequency dependence $\sigma(\omega) \propto \omega^{0.9}$ consistent with Eq. (1), but an anomalous temperature dependence was reported [12]. Deri and Castner [14] made measurements to 9 GHz, but only at relatively high temperatures ($T > 1$ K), where the system is probably not in its Coulomb glass state and the zero-phonon assumption is also not valid. They found a density-dependent superlinear frequency dependence $\sigma(\omega) \propto \omega^s$ with $1.0 < s < 1.5$.

In this Letter, we present measurements of microwave ac conductivity on localized Si:B samples at temperatures low enough (0.08 K base temperature) to be clearly in the zero-phonon Coulomb glass state and with bandwidth large enough (0.1 to 20 GHz) to cover the Coulomb correlation energy. We show the first clear experimental measurement of the crossover between interacting conductivity at low frequency and noninteracting conductivity at higher frequency.

The samples used in this experiment are single-crystal silicon chips doped with boron (Si:B) at densities of $n/n_c = 0.85$ and 0.88, where $n_c = 4.0 \times 10^{18}$ cm^{-3} is the critical density of the metal-insulator transition [15]. The dc characteristics of samples from the same wafers have been reported extensively [3,16]. The dc resistivity (ρ) data [3] show that they exhibit Coulomb correlated hopping of the form $\ln\rho \propto (T_0/T)^{1/2}$ for $T < 1$ K, where $T_0 = 12$ and 10 K are the hopping correlation temperatures for $n/n_c = 0.85$ and 0.88, respectively, and the respective Coulomb gaps $2\Delta = 0.06$ and 0.05 meV obtained from fits to dc transport [17]. Hence these samples become Coulomb glasses at temperatures below $\sim$0.5 K. The size of 2Δ means that the characteristic photon frequency of the correlation should be between 6 to 18 GHz. In this frequency range the zero-phonon assumption becomes valid below $\sim$0.3 K. Hence it is necessary to reach measurement temperatures $\leq$0.1 K to satisfy the Coulomb glass and zero-phonon conditions. With our current experimental capability, only samples in this narrow density range are suitable to study Coulomb glass ac conductivity. At significantly lower densities Δ becomes large enough to exceed our measurement bandwidth. At much higher densities Δ is small enough to require lower temperatures than achievable in our dilution refrigerator with the broadband microwave system in operation. For example, a sample with $n/n_c = 0.82$ showed essentially linear ac conductivity all the way to 20 GHz, whereas a sample with $n/n_c = 0.93$ showed a superlinear but subquadratic ac conductivity similar to that reported in Ref. [14].

The wafers were diced into $\frac{1}{4}$ in. $\times$ $\frac{1}{4}$ in. $\times$ 200 μm thick chips, and Ohmic contacts were patterned on the polished face using annealed aluminum. The contact geometry used was a Corbino disk [18], where a center contact matched the inner conductor of a 2.92 mm microwave plug, and a concentric outer contact matched the outer conductor. This geometry has the advantages of preserving the symmetry of the coaxial transmission line feeding the sample, suppressing fringe fields, and mating easily with the sample with the coaxial transmission line.

Each sample terminated the end of a coaxial transmission line that reached into the mixture of a dilution refrigerator with a demountable epoxy mixing chamber. Magnitude and phase of the reflection coefficient $S_{11}(\omega/2\pi)$ in the coax were measured using a vector network analyzer (VNA) in step-sweep mode. All measurements were at low enough power to be in the linear response regime. With the VNA set to output -20 dBm of power, we estimate the power delivered to the sample to be near 250 nW at 10 GHz, accounting for line attenuation and impedance mismatches. The load impedances measured were independent of VNA output power within a factor of 2 above and below this power, so there is no evidence for electron heating. The samples' conductivity is low enough that there is significant microwave field throughout the sample thickness. There is no sign of a decreasing surface conductance with rising frequency that might indicate a skin depth limiting effect. Also, the index difference between Si:B and liquid He is large enough that negligible power is radiated into the He mixture. Using standard transmission line equations [19], the load impedance $Z(\omega/2\pi)$ of the sample and the ac conductivity $\sigma(\omega)$ were computed from S_{11}.

The major experimental difficulty involves imperfections in the long coaxial line. Stringent thermal load considerations required the use of a 1.2 m length of 0.086 in. o.d. all-stainless steel coax. These lines have large temperature- and frequency-dependent attenuation and phase shifts. However, proper calibration of the line using three known loads across the temperatures and frequencies of interest can correct systematic errors due to transmission line imperfections. In our system, calibration and measurement of a known NiCr film standard shows that we can measure absolute load impedance to an accuracy of better than 2%. Details of such a calibration procedure developed by us are described elsewhere [20]. The bandwidth of the calibration used in this experiment is 20 GHz.

Figure 1 shows the real part of the ac conductivity as a function of frequency for samples with dopant density $n/n_c = 0.85$ and 0.88 at $T = 0.080$ K. The increasing noise and spikes above 18 GHz are due to resonances in

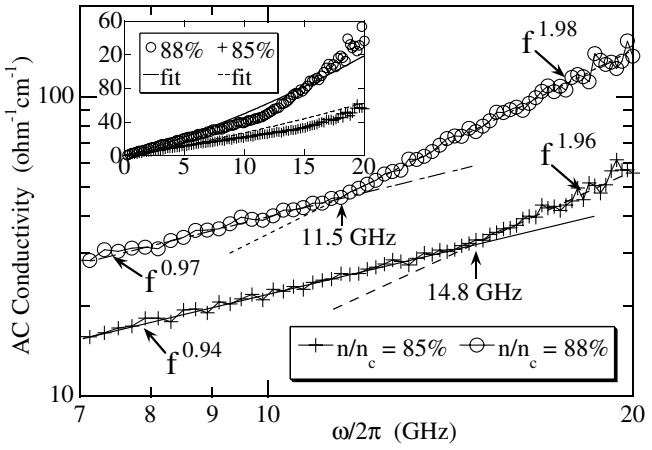

FIG. 1. Real part of the ac conductivity as a function of frequency for Si:B samples with dopant densities $n/n_c = 0.85$ and 0.88. Data from 7 to 20 GHz are plotted on a log-log plot. The various lines are fits of the high- and low-frequency parts of the data to power laws. The crossover frequency for each sample is identified by the intersection of the lines from high and low frequency. Inset: The data plotted on a linear scale over the whole band 0.1 to 20 GHz. The solid and dashed lines are fits of the data to Eq. (2), where prefactors have been chosen to match the low frequency data.

the transmission lines that are incompletely removed by the calibration procedure because of degrading signal-to-noise. The main figure shows the data from 7 to 20 GHz plotted on a log-log scale to identify power laws. A crossover between two power law behaviors can clearly be identified in the spectrum for each sample. At lower frequencies, the best power-law fit gives $\sigma(\omega/2\pi) \propto (\omega/2\pi)^s$ with $s = 0.94$ and 0.97 (both ± 0.05) for the $n/n_c = 0.85$ and 0.88 samples, respectively. At the highest frequencies, $\sigma(\omega/2\pi) \propto (\omega/2\pi)^{s'}$ with $s' = 1.96$ and 1.98 (both ± 0.1) for $n/n_c = 0.85$ and 0.88, respectively. If we simply take $s = 1$ and $s' = 2$, then the best fits are $\sigma(\omega/2\pi) = (2.2\ \Omega^{-1}\,\mathrm{cm}^{-1}/\mathrm{GHz})(\omega/2\pi)$ and $(4.0\ \Omega^{-1}\,\mathrm{cm}^{-1}/\mathrm{GHz})(\omega/2\pi)$ at low frequency, and $\sigma(\omega/2\pi) = [0.14\ \Omega^{-1}\,\mathrm{cm}^{-1}/(\mathrm{GHz})^2](\omega/2\pi)^2$ and $[0.34\ \Omega^{-1}\,\mathrm{cm}^{-1}/(\mathrm{GHz})^2](\omega/2\pi)^2$ at high frequency for $n/n_c = 0.85$ and 0.88, respectively. The crossover frequency ω_c is identified by extrapolating the fits in both regimes to find an intersection, which yields $(\omega_c/2\pi) = 14.8$ and 11.5 GHz for $n/n_c = 0.85$ and 0.88, or photon energies $\hbar\omega_c = 0.061$ and 0.048 meV.

The inset of Fig. 1 shows the conductivity plotted linearly over the whole frequency range 0.1 to 20 GHz. The low frequency quasilinear frequency dependence seen in the main figure extends down to 0.1 GHz. The solid and dashed curves in the inset are fits for both samples to the form of Eq. (1), using the measured values of the linear slopes to determine the prefactors, and treating $\ln(2I_0)$ and $U(r_\omega)$ as fitting parameters. Because the linear coefficients are obtained from experiment, the data at the lowest frequencies automatically fit well. However, it is obvious that Eq. (1) is a poor fit to the data over the whole frequency range. In particular, for both samples the measured crossover from quasilinear to quasiquadratic behavior is much sharper than the very gradual transition predicted by Eq. (1).

Figure 2 shows the temperature dependence of the linear slope, from 0.1 to 10 GHz, as a function of T from 0.08 to 1.0 K. Below $\sim$0.25 K, the slope is temperature independent to within measurement uncertainty, which is evidence that the ac conductivity is in the zero-phonon regime. Above roughly 0.25 K, the slope starts to increase approximately linearly with increasing temperature, indicating that phonon-assisted hopping is becoming important. The approximately linear temperature dependence is consistent with the Austin-Mott model [21] for ac hopping conductivity with phonon activated processes.

Looking at the crossover photon energy $\hbar\omega_c$, we reiterate that Eq. (1) predicts a transition when $\hbar\omega_c$ equals the pair electrostatic energy $e^2/\kappa r_{\omega_c}$, not when $\hbar\omega_c$ equals 2Δ. However, a connection between $\hbar\omega_c$ and the many-body correlation energies 2Δ and T_0 emerges from an analysis of the data. First, fits to a first-principles theory [17] of the crossover from noninteracting to correlated dc hopping for these samples gives $2\Delta = 0.06$ and 0.05 meV (both ± 0.01 meV) for $n/n_c = 0.85$ and 0.88, respectively. In both cases, 2Δ equals the measured $\hbar\omega_c$ to well within experimental uncertainty. Also, the related correlation temperature T_0 can be estimated from $\sigma(\omega)$. For $n/n_c = 0.85$, we evaluate $r_{\omega_c} = \xi \ln(2I_0/\omega_c)$ using $\omega_c/2\pi = 14.8$ GHz and $I_0 = 10^{12}$ to 10^{14} s^{-1} to obtain $3.1\xi < r_{\omega_c} < 7.7\xi$. Using the mean $r_{\omega_c} = 5.4\xi$ gives $e^2/\kappa r_{\omega_c} = 0.18e^2/\kappa\xi = 0.066T_0$, where $T_0 = 2.8e^2/\kappa\xi$ is the Efros-Shklovskii definition. Using $e^2/\kappa r_{\omega_c} = \hbar\omega_c = 0.061$ meV then gives $T_0 = 10.7 \pm 4$ K, where the uncertainty reflects the range of r_{ω_c} values. This T_0, obtained using only zero-temperature ac conductivity data, agrees well with the $T_0 = 12 \pm 0.5$ K obtained from dc $\rho(T)$ data [3]. A similar conclusion holds for the $n/n_c = 0.88$ sample. Finally, for the two samples measured, $\hbar\omega_c$ scales with n/n_c in a manner consistent

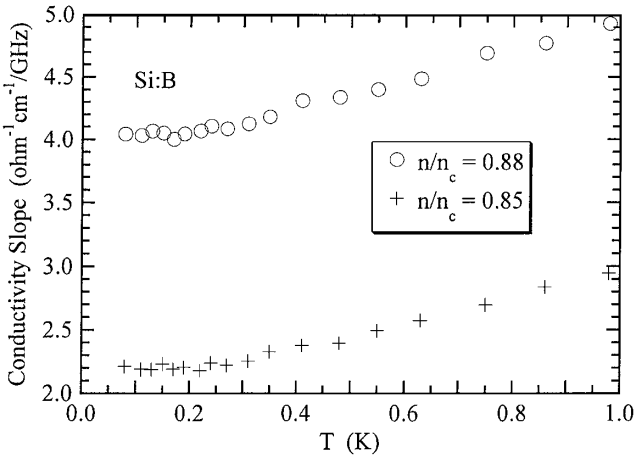

FIG. 2. Temperature dependence of the slope of the linear part of the ac conductivity (0.1 to 10 GHz) for the samples whose lowest temperature conductivity spectra are depicted in Fig. 1.

with the density dependence of Δ and T_0 expected from scaling theory and obtained from tunneling and transport experiments previously reported on Si:B samples in the same range of n/n_c [16].

Thus we find that although Eq. (1) provides a *qualitative* guide that predicts a transition from quasilinear to quasiquadratic frequency dependence, it fails to describe *quantitatively* the conductivity spectrum in Fig. 1, at least for samples this close to n_c. The fact that the observed transition is much sharper than predicted by Eq. (1) indicates that an energy threshold more sharply defined than $e^2/\kappa r_\omega$ controls the transition. The equality of $\hbar\omega_c$ and 2Δ and the reasonable value for T_0 obtained from $\sigma(\omega)$ strongly suggest that the relevant energy threshold is, in fact, the Coulomb gap width 2Δ, so that the transition from interacting to noninteracting conductivity is driven by photon-induced excitations across the Coulomb gap edge where $e^2/\kappa r_{\omega_c} \approx 2\Delta$. Such gap edge excitations are not included in the derivation of Eq. (1). Hence the challenge for understanding the physics of the ac conductivity lies in building a quantitative description of $\sigma(\omega)$ that takes into account gap edge excitations and that applies closer to n_c.

In summary, we have shown the first experimental evidence for a transition from interacting to noninteracting behavior in the zero-phonon ac conductivity spectrum of a Coulomb glass, Si:B. The form of the data is qualitatively described by existing pair approximation theory, but the theory cannot provide a quantitative fit. Analysis of the data strongly indicates that gap edge excitations are most important to the transition, something not anticipated in existing theory.

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