Electrodynamics of a Coulomb Glass in n-Type Silicon

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Measurements of the complex frequency dependent conductivity of uncompensated n-type silicon are reported. The experiments are done in the quantum limit, $\hbar\omega > k_BT$, across a broad doping range on the insulating side of the metal-insulator transition. The low energy linear frequency dependence is consistent with theories of a Coulomb glass, but discrepancies exist in the relative magnitudes of the complex components. At higher energies we observe a crossover to a quadratic frequency dependence that is sharper than expected. The concentration dependence gives evidence that the Coulomb interaction energy is the energy scale that determines this crossover.

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Doped semiconductors close to the metal-insulator transition (MIT) are systems ideally suited for the study of strong electron-electron interactions. Below a critical doping the material is an insulator; i.e., at zero temperature there is no dc conductivity. The charge carriers are localized in the Anderson sense, and screening is reduced from the metallic regime.

Anderson coined the term Fermi glass to describe a noninteracting disordered insulating system whose universal properties, independent of system specific details, are determined by Fermi statistics alone [1]. Mott's original treatment of such a system [2], which has a finite density of states at the Fermi level, but where localization creates the insulating state, did not address the issue of electron-electron interactions as pointed out by Pollak [3], and then Efros and Shklovskii (ES) [4]. A Fermi glass that includes interactions between localized electrons is referred to as a Coulomb glass. Among other things, such a system is typified by a depletion in the single particle density of states around the Fermi level which was termed by ES the Coulomb gap, Δ [5].

By taking into account the mean Coulomb interaction between two sites forming a resonant pair $U(r_\omega) = e^2/\varepsilon_1 r_\omega$, where $r_\omega = \xi[\ln(2I_0/\hbar\omega)]$ is the most probable hop distance between pairs, I_0 is the hopping attempt rate, and ε_1 is the dielectric constant, ES derived the real part of the ac conductivity of a Coulomb glass to be

$$\sigma_1 = \beta e^2 g_0^2 \xi^5 \omega[\ln(2I_0/\hbar\omega)]^4[\hbar\omega + U(r_w)]. \quad (1)$$

Here β is a constant of order one, g_0 is the noninteracting density of states, and ξ is the localization length. This formula takes on a different frequency dependence in two limits. When the photon energy $\hbar\omega > U(r_\omega)$, one recovers the same quadratic frequency dependence that Mott derived for a noninteracting Fermi glass. Here the Coulomb glass is indistinguishable from the Fermi glass in so far as the high frequency limit of the conductivity is concerned. In the opposite limit, $\hbar\omega < U(r_\omega)$, the conductivity of a Coulomb glass will show an approximately linear dependence on frequency, plus logarithmic corrections. The imaginary component of the complex conductivity, σ_2, as predicted by Efros [6], should be identical to σ_1 up to a logarithmic factor. We should note that Eq. (1) was derived for the case where $\hbar\omega > \Delta$, the Coulomb gap width. However, a linear dependence (albeit with additional logarithmic corrections) and an eventual crossover to Mott's noninteracting quadratic law is still expected even for the case where $\hbar\omega < \Delta$.

Despite its fundamental importance, there have been very few ac conductivity studies that have been done in the quantum limit $\hbar\omega > k_BT$, yet at low enough frequencies to probe the relevant energy ranges of Eq. (1). Only very recently have measurements been attempted that address these issues. Lee *et al.* found that for concentrations close to the MIT the expected linear to quadratic crossover occurs, but is much sharper than predicted [7]. Other recent work was done on amorphous NbSi, where the general systematics of a linear in frequency Coulomb glass response was found, but a crossover to the noninteracting quadratic dependence was not reached [8].

In this Letter we report the first measurements of the frequency dependent real and imaginary conductivity on the insulating side of the MIT for a doped, crystalline semiconductor. We find a concentration and frequency dependence of these two components consistent with predictions of a Coulomb glass, but a discrepancy arises in the ratio of their relative magnitudes. At higher frequencies a crossover to quadratic Mott-like behavior is observed and due to having measured across a broad dopant range, the concentration dependence of the crossover energy is shown to be consistent with the Coulomb interaction energy U and not the Coulomb gap width Δ.

Nominally uncompensated n-type silicon samples were obtained from Recticon Enterprises Inc. A boule of silicon was grown using the Czochralski method and sliced into 1 mm thick slabs. Room temperature resistivity was measured using an ADE 6035 resistivity gauge, and the concentration was determined using the Thurber scale [9]. A number of samples were etched with a 4%HF + 96%HNO_3 solution; this resulted in no difference in the results of frequency dependent conductivity measurements. The Si:P samples discussed in this Letter span a range from 39% to 69%, stated as $\frac{x}{x_c}$, a percentage of the sample's dopant concentration to the critical concentration at the MIT.

At 35 and 60 GHz the real part of the conductivity was evaluated from the measured loss of highly sensitive resonant cavities via the perturbation method. The technique and analysis is well established [10]. In the millimeter spectral range, 80 GHz to 1 THz, backward wave oscillators were employed as coherent sources in a transmission configuration [11]. Fabry-Perot–like resonances in the transmission were analyzed uniquely determining both components of the complex conductivity. Cavity measurements were performed down to 1.8 K and in the millimeter spectral range down to 2.8 K. With these base temperatures the quantum limit $\hbar\omega > k_B T$ (1 K $\approx$ 20 GHz) of the system was being investigated.

The real part of the frequency dependent conductivity, σ_1, at $T = 2.8$ K for two samples is shown in Fig. 1. These data, representative of all the samples, show an approximately linear dependence at low frequencies and then a sharp crossover to an approximately quadratic behavior at higher frequencies. This is the qualitatively expected behavior from Eq. (1). However, as seen by the overlayed fits, Eq. (1) provides only a rough guide. The solid lines are linear and quadratic fits to the low frequency and high frequency data, respectively. Individually these functions fit the data well; however, the full crossover function does not. The dotted line is a fit using the same method as Ref. [7], namely, forcing the linear portion to pass through the origin as well as the low frequency data and leaving the prefactor of the quadratic term as a free variable. The crossover between linear and quadratic portions is much more abrupt than the ES function predicts and is observed over our entire doping range as was observed previously in an analogous system, Si:B, for samples closer to the MIT [7].

Having shown that the frequency dependence is qualitatively consistent with a Coulomb glass, we focus on the low frequency regime. Both the real part and the magnitude of the imaginary part, $|\sigma_2|$ of the complex conductivity are plotted as a function of frequency in Fig. 2. The dotted lines are linear fits and show the relative increase in the magnitude of both components as the MIT is approached.

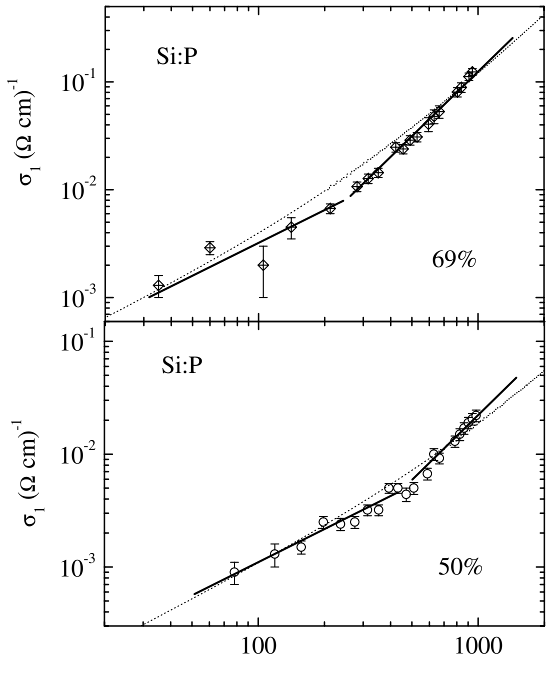

FIG. 1. $T = 2.8$ K real part of the frequency dependent conductivity versus frequency. The Si:P samples shown are 50% and 69% dopant concentration relative to the critical concentration, $x_c = 3.5 \times 10^{18}$ cm^{-3}. The solid lines are linear and quadratic fits to the lower and upper portions of the data, respectively. The dotted line is a fit to the form of Eq. (1).

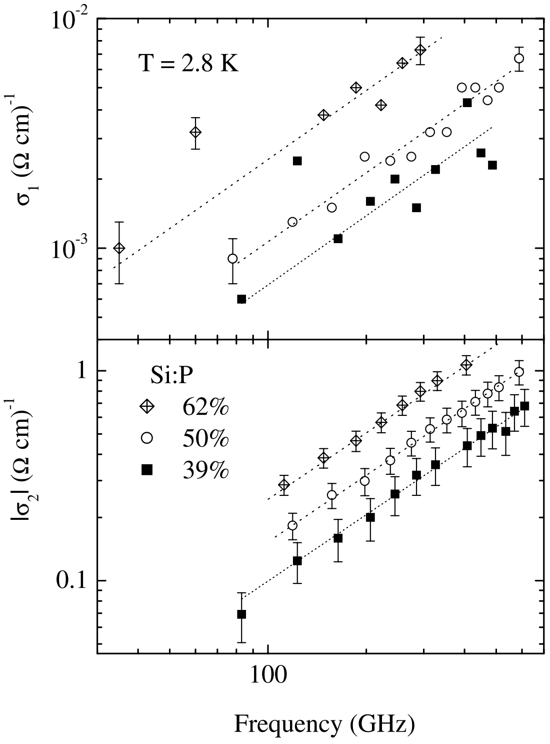

FIG. 2. The real part and the magnitude of the imaginary part of the optical conductivity for three different Si:P samples on the insulating side of the MIT is shown. The stated percentages are the sample's dopant concentration relative to the critical concentration. The dotted lines are linear best fits showing the trend of increasing conductivity as the MIT is approached. The error bars shown for σ_1 are representative of those for all the data points. Some of the lower frequency points were determined using resonant cavities at a base temperature of 1.8 K.

A complex conductivity obeying a power law can be expressed in a simple Kramers-Kronig compatible form, $\sigma_1(\omega) + i\sigma_2(\omega) = A(i\omega)^\alpha$, in order to have an independent means for determining the exponent. To determine the power α one can utilize the ratio of $|\sigma_2|$ versus σ_1 (with the frequency as a variable). The power α is given by

$$\alpha = \frac{2}{\pi}\tan^{-1}\left(\frac{|\sigma_2|}{\sigma_1}\right). \tag{2}$$

The uppermost panel in Fig. 3 shows the ratio of the imaginary to the real part of the conductivity for Si:P. NbSi is an amorphous insulating glass which is characterized by a vastly higher density of states (DOS) at E_F [8]. Disorder should give a somewhat similar functional dependence for the ac conductivity, but with very different energy scales. Data are included from it as another example of ac conductivity in a disordered system and for comparison purposes, as Si:P has a very small DOS in its impurity band. First we note that this ratio for Si:P remains large and approximately constant across our range of dopant concentrations. From theory, one expects $|\sigma_1|$ to be approximately equal to $|\sigma_2|$ to within a factor of 2–5 (with a reasonable estimate for I_0) as predicted by Efros [6]. Applied to Si:P, Eq. (1) in the $\hbar\omega < U(r_\omega)$ limit correctly predicts a linear correspondence between σ_1 and $|\sigma_2|$, but the theory incorrectly predicts the measured proportionality by at least a factor of 30. The proportionality is closer for NbSi but has a dependence on the doping concentration. This may be related to entering the quantum critical (QC) regime as discussed below. Here we have used the susceptibility due to the interacting electrons themselves $4\pi\chi = \varepsilon_1 - \varepsilon_{\mathrm{Si}}$, namely, the full measured dielectric constant, ε_1, minus the static background dielectric constant of the host silicon, $\varepsilon_{\mathrm{Si}}$, to determine the magnitude of the imaginary component of the conductivity shown in Fig. 3. The magnitude of the ratio becomes larger and the discrepancy even greater if the full dielectric constant ε_1 is considered instead.

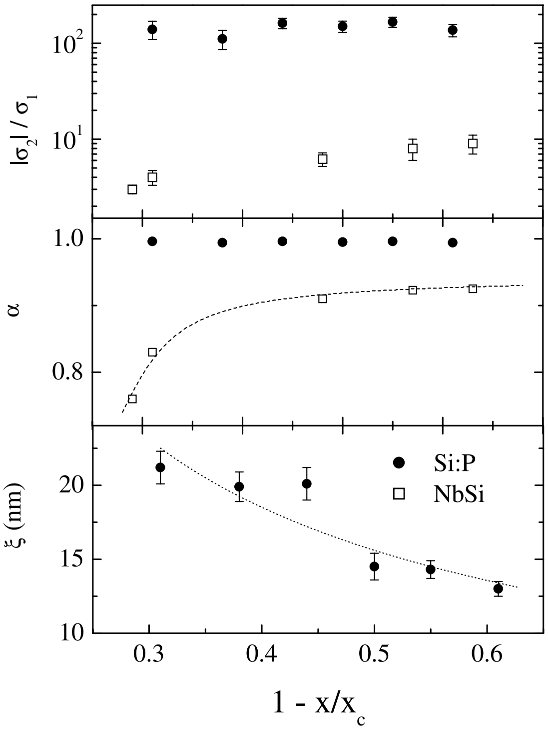

FIG. 3. The upper panel shows the ratios of the magnitude of the imaginary to the real part of the complex conductivity for samples of Si:P and amorphous NbSi. The NbSi data are adapted from Ref. [8]. The middle panel shows the calculated powers of α as determined from Eq. (2). The dashed line through the NbSi data is a guide to the eye. The bottom panel shows the divergence of the localization length (using $I_0 = 10^{13}\ \mathrm{s}^{-1}$), and the dotted line is a power-law fit of $(1 - x/x_c)^{-\beta}$, where β is found to be 0.95.

The middle panel of Fig. 3 shows the power α as determined by Eq. (2). The values for Si:P are approximately equal to, but slightly less than, one, consistent with Fig. 2. This indicates that the prefactor of the real and imaginary components of the complex conductivity has the same concentration dependence. When approaching the MIT, the frequency dependence is expected to cross over to the QC behavior [12,13], i.e., $\sigma_1 \propto \omega^{1/2}$ in NbSi, when the localization length ξ becomes comparable to the characteristic frequency dependent length scale, e.g., the dephasing length, ℓ_ω [14]. The crossover is not a phase transition and need not be sharp, therefore looking at a fixed window of frequencies, a broad, smooth crossover from $\omega \to \omega^{1/2}$ would show an averaged power of the frequency dependence similar to that measured for NbSi shown in the middle panel of Fig. 3. The fact that we see an $\alpha \approx 1$ across our whole doping range in Si:P, but an α that approaches 0.5 in NbSi indicates that the critical regime in Si:P is much narrower. Simple dimensional arguments [15] give a result similar to the noninteracting case [16] that the crossover should be inversely proportional to the dopant DOS. The vastly smaller (a factor of 10^3) dopant density in Si:P relative to NbSi is consistent with a narrower QC regime in Si:P.

Because our data span a large range of concentrations, the doping dependence of the crossover energy scale from linear to quadratic can be analyzed to see whether its dependence is consistent with the functional form of other energy scales, e.g., the Coulomb interaction energy U or the Coulomb gap width Δ. Figure 4 shows data of our experimentally determined crossover energy scale plotted versus the commonly used functional form, $1 - x/x_c$. Lee *et al.* performed ac conductivity measurements on a pair of Si:B samples [7], and in said measurements, a sharp crossover analogous to our own observations was seen. Our data extend the observed range of the sharp crossover deep into the insulating regime as the original measurements of Lee *et al.* were closer to the critical concentration.

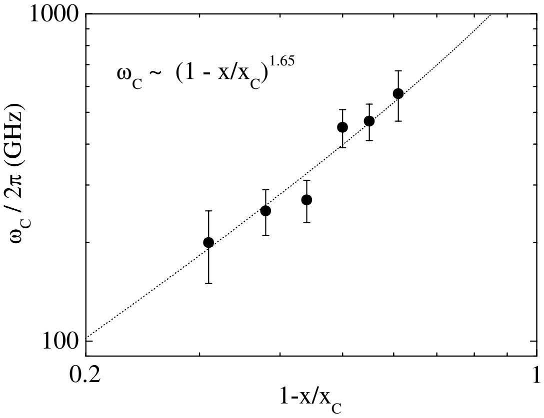

FIG. 4. The experimentally determined crossover energy scale is represented by the full circles with a fit to a power-law function of the relative concentration $(1 - x/x_c)$.

Recall that the Coulomb interaction energy between two sites forming a resonant pair is $U(r_\omega) = e^2/\varepsilon_1 r_\omega$, which is dependent on concentration via the dielectric constant and the localization length dependent most probable hop distance. The Coulomb gap width as determined by ES [4] is $\Delta = e^3 g_0^{1/2}/\varepsilon_1^{3/2}$. Although Lee *et al.* postulated that a smaller renormalized Coulomb gap governed the crossover, it is not unreasonable to expect that this gap scales with the ES single particle one as both are presumably caused by the same long range Coulomb interaction. The magnitude of the gap width is dependent on concentration through the dielectric constant and through the DOS term. Thus it is clear that a separate measurement of the full dielectric constant, ε_1 as a function of concentration, as we have done, needs to be performed in order to analyze the crossover energy scale. Although there is no expectation for ω_c or Δ to scale as a power law over the whole doping range, we may still parametrize these quantities over our doping range as power laws. We find that the crossover frequency ω_c is proportional to $(1 - x/x_c)^{1.65}$, and the full dielectric constant $\varepsilon_1 \propto (1 - x/x_c)^{-0.4}$. Using these values, it seems highly unlikely that $\hbar\omega_c = \Delta$, as the required concentration dependence of the DOS for this to be valid seems improbable.

In contrast, we do find strong support that the crossover energy is determined by the Coulomb interaction energy. By setting the measured crossover energy scale equal to the Coulomb interaction energy we are able to determine both the magnitude of the localization length and the exponent with which it diverges as a function of concentration. Using an appropriate prefactor for the overlap integral [17], $I_0 = 10^{13}\ \mathrm{s}^{-1}$ in the most probable hop distance term, r_ω, we find a localization length dependence as plotted in the bottom panel of Fig. 3. The localization length exponent is found to be close to unity, the value originally predicted by McMillan in his scaling theory of the MIT [18], and the magnitude of the localization length is reasonable. Both these results strongly point towards the Coulomb interaction energy as being the energy scale at which the observed frequency dependent crossover from ES to Mott-like hopping conduction occurs.

In summary, we have observed behavior consistent with a Coulomb glass across our entire range of doping concentrations in Si:P for the frequency and concentration dependence of the real and imaginary components of the conductivity. It is consistent with theoretical predictions except in predicting the relative magnitudes of σ_1 and σ_2 and the sharper than expected crossover from linear to quadratic frequency dependence. From the expected form of $U(x)$ and the reasonable behavior of the extracted localization length, it seems likely that the crossover is governed by the Coulomb interaction strength of a resonant pair and not the Coulomb gap.

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