Electrodynamics of a Coulomb Glass in n-type Silicon

E. Helgren, N. P. Armitage and G. Grüner

Dept. of Physics and Astronomy, University of California Los Angeles, Los Angeles, CA 90095

(October 22, 2002)

Optical measurements of the real and imaginary frequency dependent conductivity of uncompensated n-type silicon are reported. The experiments are done in the quantum limit, $\hbar \omega > k_B T$, across a broad doping range on the insulating side of the Metal-Insulator transition (MIT). The observed low energy linear frequency dependence shows characteristics consistent with theories of a Coulomb glass, but discrepancies exist in the relative magnitudes of the real and imaginary components. At higher energies we observe a crossover to a quadratic frequency dependence that is sharper than expected over the entire dopant range. The concentration dependence gives evidence that the Coulomb interaction energy is the relevant energy scale that determines this crossover.

PACS numbers: 72.20.Ee, 71.30.+h, 71.45.Gm

Doped semiconductors close to the 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.

Anderton coined the term Fermi glass to describe a non-interacting 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 makes an insulator, did not address the issue of electron-electron interactions as pointed out by Pollak [3], and then Efros and Sklovskii (ES) [4]. A Fermi glass that includes interactions between localized electrons has been termed 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, $\Delta$ [5].

By taking into account the mean Coulomb interaction between two sites forming a resonant pair $U(r_w) = e^2/\varepsilon_1 r_w$, where $r_w = \xi|\ln(2I_0/\hbar \omega)|$ is the most probable hop distance between pairs, $I_0$ is the hopping attempt rate and $\varepsilon_1$ 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 $\beta$ is a constant of order one, $g_0$ is the non-interacting density of states and $\xi$ is the localization length. This formula takes on a different frequency dependence in two limits. When the photon energy, $\hbar \omega > U(r_w)$, one recovers the same quadratic frequency dependence that Mott derived for a non-interacting 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, $\hbar \omega < U(r_w)$, the conductivity of a Coulomb glass will show an approximately linear dependence on frequency, plus logarithmic corrections. The imaginary component of the complex conductivity, $\sigma_2$, as predicted by Efros [6], should be identical to $\sigma_1$ 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 non-interacting 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 $\hbar \omega > k_B T$, 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. M. 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 non-interacting 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 the components consistent with predictions of a Coulomb glass, but a discrepancy arises in the predicted 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 $\Delta$.

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 determined using the Thurber scale [9]. A number of samples were etched with a 4%HF + 96%HNO₃ 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 $x_c$, a percentage ratio of the sample’s dopant concentration to the critical concentration at the MIT.

![Graph showing frequency-dependent conductivity](image)

**FIG. 1.** $T = 2.8$ K real part of the frequency dependent conductivity versus frequency. The Si:P samples shown are at 50% and 69% dopant concentration relative the critical concentration, $x_c = 3.5 \times 10^{18} \text{ cm}^{-3}$. 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).

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 1.2 K and in the millimeter spectral range down to 2.8 K. With these base temperatures the quantum limit $\hbar \omega > k_B T$ ($1 K \approx 20 \text{GHz}$) of the system was being investigated.

The real part of the frequency dependent conductivity, $\sigma_1$, at $T = 2.8$ K for two samples is shown in Fig. 1. This data, representative of all the samples, shows 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 pre-factor 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. A complex conductivity obeying a power law can be expressed in a simple Kramers-Kronig compatible form, $\sigma_1(\omega) + i \sigma_2(\omega) = A \omega^\alpha$, in order to have an independent means for determining the exponent. To determine the power $\alpha$ one can utilize the ratio of $|\sigma_2|$ versus $\sigma_1$ (with the frequency as a variable). The power $\alpha$ is given by,

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

(2)

![Graph](image)

**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 is adapted from Ref. 8. The middle panel shows the calculated powers of $\alpha$ as determined from 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 have 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 $\xi$ becomes comparable to the characteristic frequency dependent length scale, e.g. the dephasing length, $\xi_{\omega}$ [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 $\omega \rightarrow \omega^{1/2}$ would show an averaged power of the frequency dependence similar to that measured for NbSi shown in the middle panel of Figure 3. The fact that we see an $\alpha \approx 1$ across our whole doping range in Si:P, but an $\alpha$ 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 non-interacting 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 as compared to NbSi.

Because our data spans 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 $\Delta$. Fig. 4 shows data of our ex-
Determined by the Coulomb interaction energy though. 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 pre-factor for the overlap integral [17], \( I_0 = 10^{13} s^{-1} \) in the most probable hop distance term, \( r_w \), we find a localization length dependence as plotted in the bottom panel of Fig. 3. We find that the localization length exponent is 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 \( \sigma_1 \) and \( \sigma_2 \) 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.

We would also like to thank Steve Kivelson as well as Boris Shklovskii for useful conversations. This research was supported by the National Science Foundation grant DMR-0102405.

FIG. 4. This work’s experimentally determined crossover energy scale is represented by the full circles with a fit to a function of the relative concentration \((1 - x/x_c)\) commonly used to normalize data from different materials.

Experimental determination of crossover energy scale plotted versus the commonly used functional form, \( 1 - x/x_c \). M. 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 extends the observed range of the sharp crossover deep into the insulating regime as M. Lee et al.’s original measurements were closer to the critical concentration.

Recall that the Coulomb interaction energy between two sites forming a resonant pair is \( U(r_w) = e^2/\varepsilon_1 r_w \), 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 M. Lee et al. postulated that a smaller 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 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, \( \varepsilon_1 \) as a function of concentration, as have been done, need be performed in order to analyze the crossover energy scale. Although there is no theoretical expectation for \( \omega_c \) or \( \Delta \) to scale as a power law over the whole doping range, we may still parameterize these quantities over our doping range as power laws. We find that the crossover frequency \( \omega_c \) 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 the \( \hbar \omega_c = \Delta \), as the required DOS’s concentration dependence for this to be valid seems improbable.

We do find strong support that the crossover energy is determined by the Coulomb interaction energy though.

[1] P. W. Anderson, Comments S. S. Phys. 2, 193 (1970).
[2] N. F. Mott and E. A. Davis, Electronic Processes in Non-Crystalline Materials, Second Edition, (Oxford University Press, Oxford, 1979).
[3] M. Pollak, Disc. Faraday Soc. 50, 13 (1970).
[4] A. L. Efros and B. I. Shklovskii, in Electron-electron Interactions in Disordered Systems, edited by A. L. Efros and M. Pollak (Elsevier New York, 1985), p. 409-482.
[5] A. L. Efros and B. I. Shklovskii, J. Physics C 8, L49 (1975).
[6] A. L. Efros, Sov. Phys. JETP 62, 1057 (1985). An analogous three-dimensional form of the two-dimensional theory described in this paper can be derived.
[7] M. Lee and M. L. Stutzmann, Phys. Rev. Lett. 30, 056402 (2001).
[8] E. Helgren et al., Phys. Rev. Lett., 87, 116602 (2001).
[9] W. R. Thurber et al., J. Electrochem. Soc. 127,1807 (1980).
[10] G. Grüner, *Millimeter and Submillimeter Wave Spectroscopy of Solids* (Springer Verlag, Berlin, 1998).
[11] A. Schwartz et al., Rev. Sci. Instrum. 66, 2943 (1995).
[12] H.-L. Lee, J. P. Carini and D. V. Baxter, Phys. Rev. Lett. 80, 4261 (1998).
[13] H.-L. Lee et al., Science 287, 633 (2000).
[14] S. L. Sondhi et al., Rev. Mod. Phys. 69, 315 (1997).
[15] S. A. Kivelson, private communication.
[16] B. Shapiro and E. Abrahams, Phys. Rev. B 24, 4889 (1981).
[17] Personal correspondence with B. I. Shklovskii. The \( I_0 \) used is determined by the Bohr energy of phosphorous.
[18] W. L. McMillan, Phys. Rev. B 24, 2739 (1981).