Coulomb Gap: How a Metal Film Becomes an Insulator

V. Yu. Butko,* J. F. DiTusa, and P. W. Adams

Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70806

(Received 10 September 1999)

Electron tunneling measurements of the density of states (DOS) in ultrathin Be films reveal that a correlation gap mediates their insulating behavior. In films with sheet resistance $R < 5000\ \Omega$ the correlation singularity appears as the usual perturbative $\ln(V)$ zero bias anomaly (ZBA) in the DOS. As R is increased further, however, the ZBA grows and begins to dominate the DOS spectrum. This evolution continues until a nonperturbative $|V|$ Efros-Shklovskii Coulomb gap spectrum finally emerges in the highest R films. Transport measurements of films which display this gap are well described by a universal variable range hopping law $R(T) = (h/2e^2)\exp(T_0/T)^{1/2}$.

PACS numbers: 71.30.+h, 72.15.Rn, 73.40.Gk

It has been known for some time now that 2D is the lower critical dimension for disordered transport and that even a noninteracting 2D electron gas will be localized in the presence of arbitrarily small disorder in the thermodynamic limit [1,2]. When these systems are probed at a finite length scale, by a magnetic field for instance, logarithmic corrections to the Drude conductivity are seen, but one does not expect a true metal-insulator transition in 2D or even a metallic phase in the conventional sense [2]. What is somewhat less well understood are the ramifications of Coulomb interactions and their attendant correlations, particularly in 2D systems with moderate to strong disorder. This issue in particular has now come under considerable scrutiny with the recent discovery of an apparent metal-insulator transition in the dilute 2D electron gas of Si metal-oxide-semiconductor field effect transistors (MOSFET's) [3,4]. This somewhat surprising discovery runs so counter to conventional wisdom that there has been speculation that perhaps e-e interaction effects are stabilizing an anomalous 2D metallic phase [5]. Clearly, a systematic study of the density of states (DOS) spectrum and corresponding transport characteristics of an increasingly disordered 2D electron system is needed to help illuminate the crucial interplay between disorder and correlations as the system is brought from the weakly to the strongly localized regime. In the present Letter we present such a study using ultrathin Be films with low temperature sheet resistances ranging from $R = 500\ \Omega$ to $2.6\ \mathrm{M}\Omega$.

The theory of interaction effects in disordered electronic systems has for the most part been developed in two extreme limits. In the weak disorder/interaction 2D limit it is known that the primary effect of e-e interactions is to produce a logarithmic suppression of the DOS at the Fermi energy [6], $\delta N \sim -\ln(V)$. This is commonly known as the zero bias anomaly (ZBA) and has been well established in a number of different systems via tunneling measurements of the DOS [7–9]. This depletion of the DOS is perturbative and results in a weakly metallic $\ln(T)$ transport conductivity [2]. In the opposite limit, i.e., strongly insulating regime, Efros and Shklovskii [10,11] have shown that the Coulombic interactions can produce a *nonperturbative* gap in the DOS, which is commonly known as the Coulomb gap. The 3D Efros-Shklovskii Coulomb gap, which has a quadratic energy dependence, has only recently been observed [12,13]. Interestingly, the 2D Coulomb gap is expected to be linear in energy [11],

$$N(eV) = \frac{\alpha(4\pi\epsilon_0\kappa)^2|eV|}{e^4}, \tag{1}$$

where κ is the relative dielectric constant, ϵ_0 is the permittivity of free space, and α is a constant of order unity. Despite Eq. (1) having been in the literature for more than 20 years now, there has been no direct spectroscopic verification of its peculiar linear energy dependence due to the intrinsic technical difficulties associated with measuring the DOS in thin insulating films [14]. Nevertheless, a direct measurement of this gap is important in that it is believed to play a crucial role in the electron transport [15] and thermoelectric [16] properties of highly disordered 2D metals and semiconductors. For instance, it is well known that insulating films typically obey a modified variable range hopping law of the form

$$R(T) = R_0\exp(T_0/T)^{\nu}, \tag{2}$$

where R is the film sheet resistance and R_0 is a constant. In the case of a flat DOS near the Fermi energy, the film simply obeys Mott's variable range hopping law with $\nu = 1/3$ [17]. If there is a simple gap in the DOS then T_0 is the gap energy for fixed range hopping and $\nu = 1$. Finally, if the DOS spectrum is given by Eq. (1), then one expects $\nu = 1/2$, R_0 to be of the order of the quantum resistance $R_Q = h/e^2$, [18] and

$$T_0 = 2.8e^2/(k_B 4\pi\epsilon_0\kappa\xi), \tag{3}$$

where k_B is the Boltzmann constant and ξ is the localization length [11]. Thus, when the $\nu = 1/2$ hopping form is observed, the DOS spectrum is simply assumed to be that of Eq. (1). In this Letter, we examine this assumption by presenting the first systematic electron tunneling study of the DOS in uniformly disordered metal films whose transport properties range from that of weakly metallic to strongly insulating. We show that with increasing film

resistance the DOS evolves from an essentially flat spectrum with a perturbative $\ln(V)$ ZBA to that of a Coulomb gap given by Eq. (1). We also show that the emergence of the Coulomb gap coincides with the emergence of the $\nu = 1/2$ hopping behavior of Eqs. (2) and (3).

The Be films used in the present study ranged in thickness from 1.5–2.0 nm with corresponding sheet resistances $R = 500\ \Omega - 3\ \mathrm{M}\Omega$ at $T = 50$ mK. They were deposited by thermally evaporating 99.5% pure beryllium powder onto fire polished glass substrates held at 84 K. The evaporations were made in a 4×10^{-7} Torr vacuum at a rate $\sim$0.30 nm/s. The film area was 1.5 mm $\times$ 4.5 mm. Scanning force micrographs of the films' exposed oxide surface did not reveal any salient morphological features down to the 0.7 nm resolution of the instrument. In fact, the films seemed to be as "smooth" as the fire-polished glass on which they were deposited. In addition, a transmission electron microstructural analysis of 15 nm thick Be films deposited on cleaved NaCl crystals at 84 K revealed that the films were composed of an ultrafine base structure that was interspersed with 5–15 nm Be nanocrystallites. Electron diffraction measurements showed no diffraction from the metallic base structure suggesting that it was amorphous. Similarly the oxide (BeO) produced a broad, continuous diffraction ring indicating its grain size was $<$1 nm [19].

The tunnel junctions were formed by exposing the films to atmosphere for 0.1–3 h in order to form a native oxide, then a 20 nm thick Ag counterelectrode was deposited directly on top of the film with the oxide serving as the tunnel barrier. The junction area was 0.7 mm $\times$ 0.7 mm. This technique produced tunnel junction resistances of order $R_{\mathrm{TJ}} \sim 1\ \mathrm{k}\Omega$ to $1000\ \mathrm{k}\Omega$ depending upon the exposure time and other factors. We were always careful to ensure that $R_{\mathrm{TJ}} \gg R_{\mathrm{film}}$ at low temperatures. Our lowest resistance Be films superconducted [19] which allowed us to test the integrity of the junctions by measuring the dc *I-V* characteristics at temperatures well below the superconducting transition temperature T_c. The subgap impedance of a "good" junction was always greater than $10^8\ \Omega$ [20].

Shown in the inset of Fig. 1 is the normal state conductance of the 2600 Ω film as a function of $\ln(T)$. This sample had a superconducting transition temperature $T_c = 0.33$ K which was suppressed by the application of a magnetic field oriented along the film surface. (T_c was a monotonically decreasing function of R.) This sample had the expected $\ln(T)$ weakly insulating behavior with some rounding below 100 mK. In stark contrast, the main body of Fig. 1 shows the activatedlike behavior of the 2.6 MΩ film. The solid line is a least squares fit to the data. The linearity of the data in Fig. 1 shows unequivocally that the hopping exponent $\nu = 1/2$. Furthermore the slope and intercept of the fit determine the parameters $T_0 \simeq 1.6$ K and $R_0 \simeq R_Q/2$ in Eq. (2). It appears that R_0 is universal in that it is of order the quantum resistance.

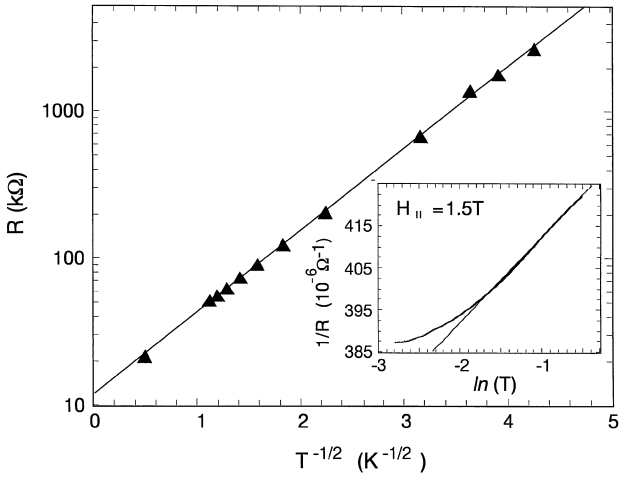

FIG. 1. Semilog plot of the resistance of the 2.6 MΩ Be film as a function of $T^{-1/2}$. The solid line is a linear fit to the data for which we get $T_0 = 1.6$ K and $R_0 \approx h/(2e^2)$; see Eq. (2). Inset: Conductance of the 2600 Ω film as a function of $\ln(T)$. The solid line is a guide to the eye.

In principle we can also use Eq. (3) along with the measured value of T_0 to calculate a localization length ξ. However, the relative dielectric constant κ in Eq. (3) is unknown for a highly disordered metal film. Alternatively, if one instead assumes a reasonable value of $\xi \sim 1$ nm and takes $T_0 \sim 1$ K, then Eq. (3) predicts $\kappa \sim 10^4$. This value seems reasonable in that it lies between the metallic and insulating limits of $\kappa \sim \infty$ and $\kappa \sim 10$, respectively. This is not an issue in 3D semiconducting systems where the dielectric constant is well defined in the insulating phase. In any case, it is evident that Fig. 1 is consistent with the existence of the Coulomb gap described by Eq. (1). Accordingly, if the Coulomb gap is indeed measurable in 2D, then it will be seen in a sample such as that of Fig. 1.

Electron tunneling provides one of the most direct experimental measures of the DOS spectrum of an electronic system. The usual tunnel junction geometry consists of an electronically "inert" metallic counterelectrode which is separated from the film by a highly insulating barrier, usually a few angstroms of oxide. When a potential is placed across the barrier, electrons tunnel from the counterelectrode and into the film. At low temperatures the tunnel current is proportional to both the counterelectrode's and the film's DOS [21]. For the technique to be useful in highly resistive films, however, care must be taken to ensure that the impedance of the tunnel junction is much higher than that of the film. Otherwise, some of the measured voltage drop will actually occur in the film itself, since the tunnel current must be drained off through the film. Additionally, there is a legitimate concern that screening effects associated with the close proximity of the highly conducting counterelectrode might "wash out" any Coulombic features in the DOS. Fortunately, this has been shown not to be the case in recent measurements of the Coulomb gap in 3D Si:B [13].

In order to demonstrate the resolution of our tunnel junctions and the 2D nature of the Be films, we have plotted in Fig. 2 the tunneling conductance of a $R = 530\ \Omega$ superconducting film, $T_c = 0.55$ K, in a parallel magnetic field just below its parallel critical field $H_{c\|} = 1.2$ T. Note that we can resolve two peaks on either side of $V = 0$. These peaks result from the Zeeman splitting of the usual BCS DOS peak and indicate that the superconductivity was spin paramagnetically limited [22]. The inset of Fig. 2 shows the normal state tunneling conductance of the same film in a supercritical parallel field. The normal state displays the expected $\ln(V)$ DOS anomaly which is consistent with the $\ln(T)$ transport behavior shown in the inset of Fig. 1.

In Fig. 3 we show the evolution of the DOS with increasing disorder by plotting tunneling spectra of Be films with $T = 50$ mK sheet resistances of $R = 530\ \Omega$, $2600\ \Omega$, $16\,000\ \Omega$, and $2.6\ \mathrm{M}\Omega$. The $530\ \Omega$ and $2600\ \Omega$ samples superconducted with $T_c = 0.55$ K and $T_c = 0.33$ K respectively. For those samples a parallel magnetic field was applied in order to suppress the superconducting state and a standard ac lock-in technique was used to measure tunneling conductance G directly. In the higher resistance films, the conductance was obtained by numerically differentiating dc I-V curves. Note that in each data set there is a significant ZBA. In the $530\ \Omega$ and $2600\ \Omega$ films the ZBA was of the weak disorder form $\delta N/N \sim -\ln(V)$. However, as is clearly evident in Fig. 3, the ZBA was no longer perturbative in films with $R > 10^4\ \Omega$. An exponential growth in the ZBA is shown in Fig. 4 where we have plotted $G(0)/G(15\ \mathrm{mV})$ as a function of R. By making a linear fit to the data we find that $G(0) \sim \exp(-R/R_c)$ where $R_c = 6000\ \Omega \approx R_Q/4$.

The most interesting attribute of the data in Fig. 3 is that the conductance traces become more linear at the highest resistances studied. The energy dependence of the $16\,000\ \Omega$ curve represents the DOS spectrum of a film somewhere between the weak and strong disorder limits and is correspondingly neither $\ln(V)$ nor $|V|$ in form. In fact, this spectrum is best described by a $V^{1/2}$ dependence. In contrast, the $2.6\ \mathrm{M}\Omega$ spectrum is quite linear and symmetric. For a consistency check, we also measured the dc I-V characteristic of the films over the same current and temperature ranges that were used in the tunneling measurements. The nonlinearity in these transport I-V's was negligible in comparison to the tunneling I-V's, indicating that the traces in Fig. 3 are representative of the tunneling behavior. The solid line through the $2.6\ \mathrm{M}\Omega$ data is a least squares fit to the form $G(V) = \beta|V|$ where β is an adjustable parameter. We believe that the linear behavior of these data represents the Efros-Shklovskii gap as given by Eq. (1). Unfortunately, we were unable to determine an absolute normalization of the tunneling conductance. The spectrum remained linear up to our maximum biases of ~ 100 mV. Consequently, we cannot make a quantitative comparison between Eq. (1) and the tunneling data. Nevertheless, the observation of a linear spectrum in films that show the expected hopping transport behavior is compelling.

In conclusion, we have made the first direct spectroscopic measurements of the Efros-Shklovskii Coulomb gap in a 2D system. We find the gap emerges from an exponential growth in the ZBA as the film sheet resistance is increased. The details of the activated transport behavior of films showing the gap are in excellent agreement with theory and have a universal prefactor of the order of the quantum resistance. In principle, spectroscopic studies of the Coulomb gap in films deposited *in situ* would allow for

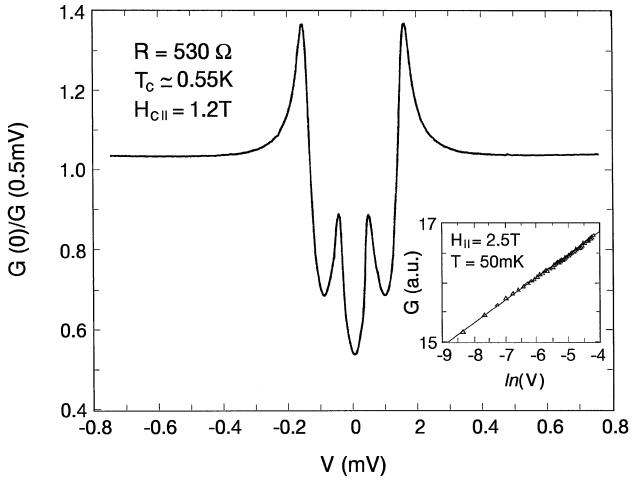

FIG. 2. Tunnel conductance in the superconducting state of the $530\ \Omega$ film in a parallel magnetic field $H_{\|} = 1.1$ T. The critical parallel field was $H_{c\|} = 1.2$ T. The tunneling spectrum demonstrates that we can resolve the Zeeman splitting of the usual BCS DOS. Inset: Normal state tunneling conductance of the same film as a function of $\ln(V)$ at $H_{\|} = 2.5$ T.

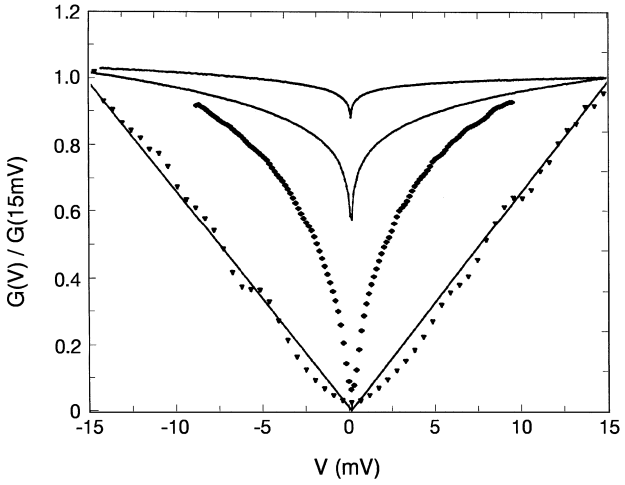

FIG. 3. Tunnel conductances normalized to G (15 mV) for Be films with $T = 50$ mK resistances of $R = 530\ \Omega$, $2600\ \Omega$, $16\,000\ \Omega$, and $2.6\ \mathrm{M}\Omega$ (top to bottom). The solid lines are a best fit to the form $G(V) = \beta|V|$, where β is an adjustable parameter. The $2.6\ \mathrm{M}\Omega$ film had a $R_{\mathrm{TJ}} = 0.6\ \mathrm{M}\Omega$ junction. To ensure that $R_{\mathrm{film}} \ll R_{\mathrm{TJ}}$ we took the $2.6\ \mathrm{M}\Omega$ data at 700 mK; see Fig. 1. The other curves were measured at 50 mK.

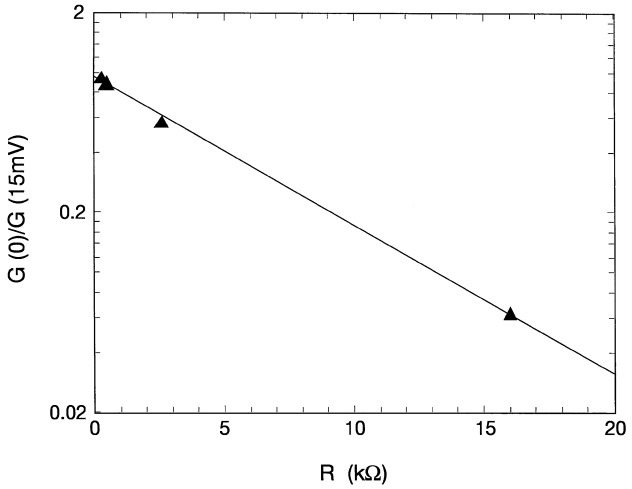

FIG. 4. Semilog plot of the normalized zero bias tunneling conductance as a function of R. The solid line is linear fit to the data.

an absolute calibration of the tunneling conductance since a variety of film thickness could be investigated using a single tunnel junction. Such measurements could then be used in conjunction with transport measurements to extract the two primary microscopic parameters of the theory, κ and ξ, via Eqs. (1) and (3).

We gratefully acknowledge discussions with Boris Shklovskii, Vladimir Dobrosavljevic, and Boris Altshuler. This work was supported by the NSF under Grants No. DMR 9972151 and No. DMR 97-02690.

*Permanent address: Ioffe Physical Technical Institute (PTI), Russian Academy of Sciences, Polytekhnicheskaya Street, 26, 194021, St. Petersburg, Russia.

[1] E. Abrahams, P. W. Anderson, D. C. Licciardello, and T. V. Ramakrishnan, Phys. Rev. Lett. **42**, 673 (1979).
[2] P. A. Lee and T. V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985).
[3] S. V. Kravchenko, W. E. Mason, G. E. Bowker, J. E. Furneaux, V. M. Pudalov, and M. D'Iorio, Phys. Rev. B **51**, 7038 (1995).
[4] D. Simonian, S. V. Kravchenko, M. P. Sarachik, and V. M. Pudalov, Phys. Rev. Lett. **79**, 2304 (1997).
[5] V. Dobrosavljevic, E. Abrahams, E. Miranda, and S. Chakravarty, Phys. Rev. Lett. **79**, 455 (1997).
[6] B. L. Altshuler, A. G. Aronov, M. E. Gershenson, and Yu. V. Sharvin, Sov. Sci. Rev. A **9**, 223 (1987).
[7] Yoseph Imry and Zvi Ovadyahu, Phys. Rev. Lett. **49**, 841 (1982).
[8] M. E. Gershenson, V. N. Gubankov, and M. I. Falei, Pis'ma Zh. Eksp. Teor. Fiz. **41**, 435 (1985) [JETP Lett. **41**, 535 (1985)].
[9] A. E. White, R. C. Dynes, and J. P. Garno, Phys. Rev. B **31**, 1174 (1985).
[10] A. L. Efros and B. I. Shklovskii, J. Phys. C **8**, L49 (1975).
[11] B. I. Shklovskii and A. L. Efros, *Electronic Properties of Doped Semiconductors* (Springer, New York, 1984).
[12] J. G. Massey and Mark Lee, Phys. Rev. Lett. **75**, 4266 (1995).
[13] J. G. Massey and Mark Lee, Phys. Rev. Lett. **77**, 3399 (1996).
[14] H. B. Chan, P. I. Glicofridis, R. C. Ashoori, and M. R. Melloch, Phys. Rev. Lett. **79**, 2867 (1997).
[15] Shih-Ying Hsu and J. M. Valles, Jr., Phys. Rev. Lett. **74**, 2331 (1995); Y. Shapir and Z. Ovadyahu, Phys. Rev. B **40**, 12 441 (1989); Youzhu Zhang and M. P. Sarachik, Phys. Rev. B **43**, 7212 (1991).
[16] J. J. Lin, J. L. Cohn, and C. Uher, in *Anderson Localization*, edited by T. Ando and H. Fukuyama (Springer-Verlag, New York, 1988), p. 186.
[17] N. F. Mott and G. A. Davis, *Electronic Processes in Non-Crystalline Materials* (Clarendon, Oxford, 1979), 2nd ed.
[18] R. Berkovits and B. I. Shklovskii, J. Phys. Condens. Matter **11**, 779 (1998).
[19] P. W. Adams, P. Herron, and E. I. Meletis, Phys. Rev. B **58**, 2952 (1998).
[20] M. Tinkam, *Introduction to Superconductivity* (McGraw-Hill, New York, 1996).
[21] E. L. Wolf, *Principles of Electron Tunneling Spectroscopy* (Oxford University Press, Oxford, England, 1985).
[22] P. Fulde, Adv. Phys. **22**, 667 (1973).