Hard Correlation Gap Observed in Quench-Condensed Ultrathin Beryllium

E. Bielejec, J. Ruan, and Wenhao Wu
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627

(Received 23 August 2000; published 26 June 2001)

We report on the tunneling density of states (DOS) in strongly disordered ultrathin Be films quench condensed at 20 K. Above 5 K, the DOS shows the well-known logarithmic anomaly at the Fermi level. Only in a narrow temperature range near 2 K is the DOS linearly dependent on energy, as predicted by Efros and Shklovskii. However, both the zero-bias conductance and the slope of the linear DOS are found to decrease drastically with decreasing temperature. Tunneling measurements at mK temperatures have revealed conclusively that a hard correlation gap opens up in the DOS.

DOI: 10.1103/PhysRevLett.87.036801 PACS numbers: 73.40.Gk, 71.30.+h, 72.15.Rn, 74.40.+k

It is known that electron-electron (*e-e*) Coulomb interactions can drastically alter the density of states (DOS) near the Fermi energy in disordered electronic systems. In the weakly disordered limit, Altshuler *et al.* [1] have predicted that interactions lead to a singular depletion of the DOS with a $|\epsilon|^{1/2}$ dependence in three dimensions (3D) and a $\ln|\epsilon|$ dependence [2] in two dimensions (2D), where ϵ is the energy measured from the Fermi level. These corrections have been observed in tunneling studies of the DOS in disordered metals in 3D [3] and 2D [4,5]. In the strongly insulating regime, Efros and Shklovskii (ES) have predicted [6,7] that Coulomb interactions lead to a soft Coulomb gap in the single-particle DOS, with a vanishing DOS at the Fermi level. This soft gap is quadratic in energy in 3D and linear in 2D. In both cases, the Coulomb gap is predicted [7] to lead to a variable-range hopping resistance of $R_\square(T) = R_0 \exp[(T_0/T)^\nu]$, where $\nu = 1/2$ and T is the temperature.

Although it was predicted over two decades ago, the Coulomb gap is by no means an understood subject. The existence of the ES Coulomb gap had mainly been inferred from transport studies such as glassy relaxation [8] and hopping conduction [9,10]. The ES Coulomb gap in 3D was directly observed a few years ago by tunneling in Si:B [11]. Direct evidence for the ES Coulomb gap in 2D has been reported only during the past year by Butko *et al.* [12], but no temperature dependence and magnetic field dependence have been reported. The ES Coulomb gap [6,7] describes the DOS for adding an extra electron to the ground state without allowing relaxation. Later theories [13] of the Coulomb gap, taking into consideration multielectron processes, have found a further reduction of the DOS near the Fermi energy, leading to a much harder gap with effectively no states within a narrow but finite range of energy. In fact, a change in the hopping exponent with decreasing temperature from $\nu = 1/2$ to $\nu = 1$ was observed in Si:B [10], suggesting that a hard gap might exist at low temperatures. Most recently, the Coulomb gap in 2D has become a subject of renewed interest [14] with the unexpected discovery of a metal-insulator transition in the 2D electron gas in semiconductor devices [15].

In this Letter, we report tunneling studies of the DOS in ultrathin Be films quench condensed near 20 K in a dilution refrigerator. This setup makes it possible to vary film thickness in fine steps to tune the films from the highly insulating limit to the weakly insulating limit, which can be done *in situ* at low temperatures and without exposure to air. It allows the use of a *single* junction to measure, thus to compare *in real units,* the densities of states from Be films of varying thickness following successive evaporation steps. Earlier studies have suggested [16–18] that quench-condensed Be films are nearly amorphous. It was found [17] that superconductivity could be fully established in films as thin as 12 Å. Although the superconducting transition temperature T_c of bulk Be is near 26 mK, quench-condensed Be films can have surprisingly higher T_c, reaching 10 K in thicker films [17]. Scanning force microscopy studies of our Be films, after warming up to room temperature, have found no observable granular structure down to 1 nm.

Our Be films were thermally evaporated onto bare glass substrates held near 20 K during evaporation. The substrates were mounted on a rotator, extended from the mixing chamber to the bore of a 12-T superconducting magnet. The orientation of the films with respect to that of the magnetic field could be calibrated *in situ* at low temperatures to better than 0.1° [19]. The films had a multilead pattern for resistance measurements, with an area of 3×3 mm^2 between the neighboring leads. Contact pads were preevaporated on the glass substrates. For tunneling measurements [20], one of the tunneling electrodes was an Al film of area 1×1 mm^2 and thickness 150 Å, evaporated at room temperature and oxidized in air for a period of 2 hours to 2 days. After the Be films were quench condensed, the resistance of the resulting Be/Al$_2$O$_3$/Al junctions were found to vary from 5 kΩ to 1 MΩ at 20 K. All the tunneling data and nearly all the film resistance data shown below were obtained by calculating the numerical derivative of the 4-terminal dc *I-V* curves measured using two Keithley 617 electrometers.

Figure 1 shows the temperature dependence of the film sheet resistance, $R_\square$, measured on one film section deposited on a bare glass substrate following successive

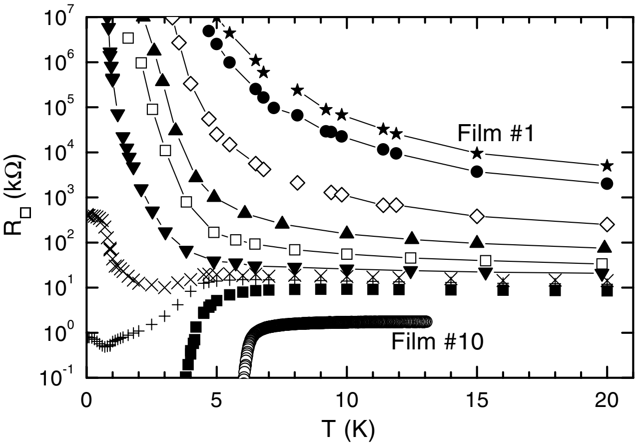

FIG. 1. Curves of film sheet resistance as a function of temperature measured on one film section following a series of deposition steps to increase film thickness. For curves from top to bottom, we label them as Film #1 to Film #10, respectively. The thickness for these films changed from 4.6 to 15.5 Å.

deposition steps to increase film thickness. The film changed its behavior from insulating to superconducting when $R_\square$ at 20 K was reduced to below 10 $\mathrm{k}\Omega/\square$ as the thickness was increased. Film #10 in Fig. 1, which was superconducting with $T_c \sim 6$ K, had a critical field H_c well above the 10-T field our magnet could reach at 4.2 K. Using the spin-paramagnetic limit [21], we estimate for this film that the upper bound of H_c is $\sqrt{2}\,\Delta/g\mu_B \approx 11.2$ T, where $g \approx 2$ is the Landé g-factor, μ_B is the Bohr magneton, and $\Delta \approx 0.92$ mV is the superconducting gap (see below). Early studies [22] estimated that the critical field was 18–20 T in quench-condensed Be films of $T_c = 8$–10 K, suggesting that these films were highly disordered with a very short penetration depth.

We measure the dc tunneling I-V from which the tunneling conductance, $G = dI/dV$, is calculated. When thermal broadening is unimportant, G is simply the product of the tunneling probability and the densities of states of the two electrodes. The quality of our junctions could be tested when the Be films became thick enough so that they were superconducting, such as Film #10 in Fig. 1. Our tunneling studies at 100 mK on Film #10 found that the combined superconducting gaps of Be and Al were 1.20 mV. In a 1.5-T perpendicular magnetic field, $H_\perp$, which was strong enough to suppress superconductivity in the Al electrode and yet was too weak to produce any measurable effect on the gap value of Film #10, we measured an energy gap of 0.92 mV for Film #10. Given that the T_c of Film #10 was near 6 K, this gap of 0.92 mV led to $2\Delta \approx 3.7 k_B T_c$. We note that, for the zero-field data presented below, we do not attempt to extract the DOS profile of the superconducting Al electrode from the measured tunneling conductance, since the energy scale associated with the DOS structures of the highly insulating Be films is far larger than the gap energy ($\sim$0.28 mV) of the superconducting Al electrode. Finally, we can rule out the possibility of the existence of a high resistance edge in the Be films at the boundary of the Al tunneling electrode which might have invalidated the tunneling data. For junctions in which the Be films cross the Al tunneling electrodes, we found that the resistance of the Be films crossing the Al electrodes was at least 3 orders of magnitude smaller than the resistance of the junctions, in the temperature range and Be film resistance range in which the tunneling data are presented below. This ensures that the measured voltage was the true bias voltage across the junctions. The observed low contact resistance of the Be films at the edge of the Al tunneling electrodes was surprising. Equally surprising was our observation that ultrathin Be films of sheet resistance of up to 600 $\mathrm{M}\Omega/\square$ showed virutally no contact resistance on typical contact pads such as Au(80 Å)/Cr(20 Å). Although we do not know the origin of the low contact resistance, we think it is related to the fact that the Be films wet most of the substrates much better than most other materials do and that the Be films are very smooth and nearly amorphous. We point out that the sheet resistance of the portions of the Be film deposited on top of the $\mathrm{Al_2O_3/Al}$ layers, which formed the junctions, could be different from the rest of the film deposited on the bare glass substrates. Thus one should be cautious when comparing the tunneling data presented below with the film sheet resistance shown in Fig. 1. The goal of this Letter is to report the tunneling DOS in quench-condensed ultrathin Be films, showing the evolution from a logarithmic anomaly to the ES linear Coulomb gap, and the eventual opening up of a hard gap. Simultaneous tunneling and resistance measurements of films deposited on exactly the same undercoating or substrates, such as $\mathrm{Al_2O_3}$, will be carried out in the future.

In Fig. 2, we plot the tunneling conductance measured at three temperatures from one of the junctions on Film #6 in Fig. 1. The data near 3 and 2 K in Fig. 2 appear to show the ES linear Coulomb gap [6,7] in 2D. However, with decreasing temperature, the slope of the linear DOS decreased sharply and the tunneling conductance at zero bias dropped toward zero. Thus the DOS cannot be described simply by the temperature-independent ES linear Coulomb gap. We point out that the resistance data measured from insulating films (Films #1 to #6) in Fig. 1 cannot be fitted to a simple hopping law with $\nu = 1/4$, $1/3$, $1/2$, or 1. This is probably not surprising, given that the measured tunneling DOS depends strongly on temperature. In contrast, the ES hopping law with $\nu = 1/2$ is derived for a temperature-independent soft Coulomb gap. We note that a significant temperature dependence of the tunneling DOS has been reported before in both 2D [5] and 3D [11]. The temperature dependence seen in Fig. 2 is likely due to the following reason. At low temperatures, electrons are strongly localized with a very low diffusivity, resulting in a very poor screening capability. This is the regime where the ES Coulomb gap is expected. As the temperature is raised, the diffusivity increases and the screening

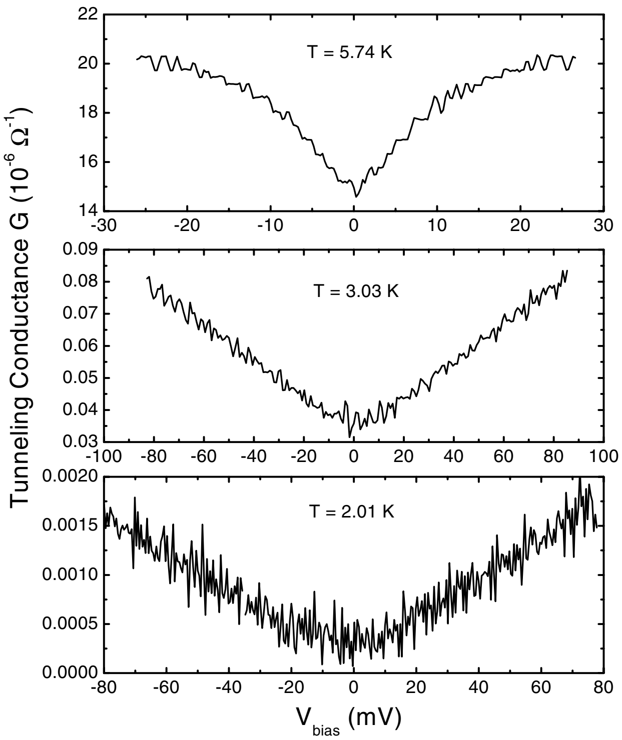

FIG. 2. Tunneling conductance G obtained from one junction on Film #6 in Fig. 1, as a function of V_{bias} at three temperatures, showing the depletion of the DOS by orders of magnitude as temperature was lowered. The noise seen in the data resulted from calculating the numerical derivative of the dc I-V curves.

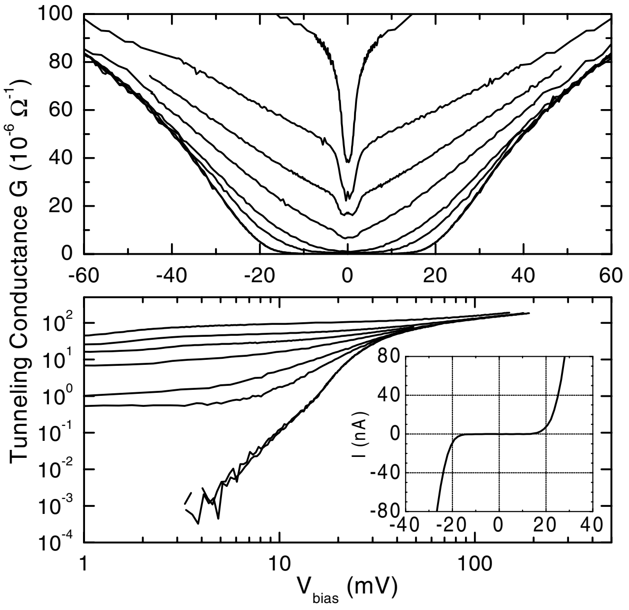

FIG. 3. Main figures show the tunneling conductance, G, as a function of bias voltage at various temperatures measured on Film #8 in Fig. 1, on both linear (top plot) and log-log (bottom plot) scales. In both plots, curves from top to bottom were measured at 3.30 K, 2.80 K, 2.50 K, 2.10 K, 1.40 K, 1.00 K, 500 mK, and 30 mK, respectively. The 500 and 30 mK curves fall on top of each other and are nearly indistinguishable. Inset: A dc tunneling I-V measured at 30 mK on Film #8, showing a hard gap of about 30 mV. In comparison, the superconducting gap in Film #10, $\Delta \approx 0.92$ mV, is much smaller.

capability is improved. In fact, the tunneling conductance at higher temperatures, such as 5.74 K, is logarithmic in energy, as predicted in the weakly disordered limit [1].

Film #6 from Fig. 1 was so insulating that tunneling measurements could not be made on this film much below 2 K, because at low temperatures such films formed highly resistive leads for the junctions. Earlier reports [11,12] have also discussed technical difficulties in tunneling experiments associated with high lead resistance. Tunneling measurements at much lower temperatures were possible only on slightly thicker films which were much more conducting, such as Films #7 and #8 in Fig. 1. The low resistance of such films ensured that the measured voltage was across the junctions, as we discussed earlier. In the inset to Fig. 3, we plot one dc tunneling I-V measured at 30 mK on Film #8. We see clearly a wide hard gap of about 30 mV in the DOS. In the main part of Fig. 3, we plot on both linear and log-log scales the tunneling conductance curves measured at a number of temperatures for $T \lesssim 3.3$ K. Above 5 K, the tunneling conductance also had a logarithmic anomaly. Apart from a sharp dip near zero bias, the tunneling conductance near 2 K showed a V-shaped linear energy dependence. The DOS depleted further with decreasing temperature and developed a temperature-independent hard gap below 500 mK.

Films #7 and #8 in Fig. 1 were quite conducting due to the existence of strong superconducting fluctuations. Nevertheless, we now argue that this hard gap is not a result of these superconducting fluctuations; rather, we believe that the hard gap is a manifestation of localization-enhanced interaction effects in strongly disordered films, and would be found in all our insulating samples if only we were able to measure tunneling in extremely insulating thinner films such as Film #6. First, it is unlikely that the hard gap originates from superconductivity since the width of the hard gap, 30–40 mV, is so much larger than the superconducting gap, $\Delta \approx 0.92$ mV, measured on Film #10. Next, we found that the width of the hard gap was reduced from 40 mV in Film #7 to 30 mV in Film #8. Although our crystal thickness monitor was not able to resolve the difference in thickness between these two films, Film #8 was produced by an additional evaporation step upon Film #7, and so is slightly thicker. Hence, we have found a correlation between a thinner film and a wider hard gap. Finally, for a fixed film thickness, we found that the hard gap broadened significantly with increasing perpendicular magnetic fields, $H_{\perp}$, in the low-field regime, as shown in Fig. 4. The hard gap was insensitive to a parallel magnetic field in the same field range. Such highly anisotropic behavior is most likely an orbital effect rather than a spin effect. Since a perpendicular magnetic field effectively localizes the electrons in 2D [23], we believe that the field broadening of the hard gap in Fig. 4 indicates a correlation

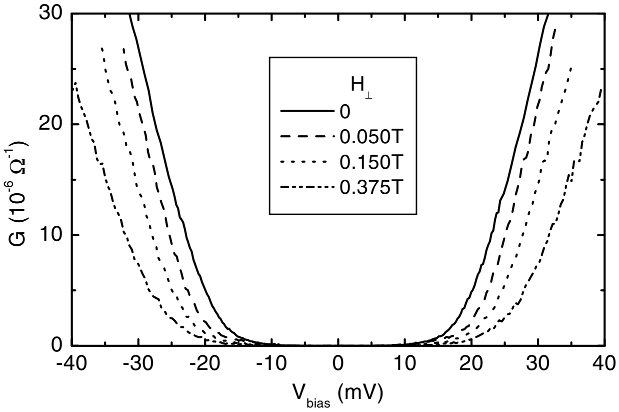

FIG. 4. Tunneling conductance, G, versus the bias voltage measured at 30 mK on Film #8 in Fig. 1, showing the broadening of the hard gap with increasing $H_\perp$.

between the hard gap and localization. In fact, correlation gaps induced by the localization effect of a strong perpendicular magnetic field have been observed before [23] in tunneling studies of the 2D electron gas in semiconductor quantum wells even in the limit of insignificant disorder.

In conclusion, we have observed in quench-condensed ultrathin Be films a strong dependence of the DOS on film thickness, temperature, and magnetic field. The DOS evolves from a logarithmic anomaly to the ES linear Coulomb gap near 5 K. At mK temperatures, a hard correlation gap as wide as 30–40 mV emerges at the Fermi level. As the films become thicker and less insulating, the hard gap narrows. Such behavior has never been observed before. We argue that this hard correlation gap results from the combination of localization and *e-e* Coulomb interactions, and that the hard gap should also exist in the strongly insulating Be films on which we were not able to perform tunneling experiments.

We gratefully acknowledge numerous invaluable discussions with S. Teitel, Y. Shapir, Y. Gao, and P. Adams. We thank S. Zorba and Y. Gao who performed scanning force microscopy studies of our quench-condensed Be films.

[1] B. L. Altshuler and A. G. Aronov, in *Electron-Electron Interactions in Disordered Systems,* edited by A. L. Efros and M. Pollak (North-Holland, Amsterdam, 1985), p. 1.

[2] B. L. Altshuler, A. G. Aronov, and P. A. Lee, Phys. Rev. Lett. **44**, 1288 (1980).

[3] R. C. Dynes and J. P. Garno, Phys. Rev. Lett. **46**, 137 (1981); J. Lesueur, L. Dumoulin, and P. Nedellec, *ibid.* **55**, 2355 (1985); T. R. Lemberger, *ibid.* **63**, 2132 (1989).

[4] A. E. White, R. C. Dynes, and J. P. Garno, Phys. Rev. B **31**, 1174 (1985); Shih-Ying Hsu and J. M. Valles, Jr., *ibid.* **49**, 16 600 (1994); J. M. Valles, Jr., R. C. Dynes, and J. P. Garno, *ibid.* **40**, 7590 (1989); Y. Imry and Z. Ovadyahu, Phys. Rev. Lett. **49**, 841 (1982).

[5] Wenhao Wu, J. Williams, and P. W. Adams, Phys. Rev. Lett. **77**, 1139 (1996).

[6] A. L. Efros and B. I. Shklovskii, J. Phys. C **8**, L49 (1975).

[7] B. I. Shklovskii and A. L. Efros, *Electronic Properties of Doped Semiconductors* (Springer, New York, 1984).

[8] D. Monroe, A. C. Gossard, J. H. English, B. Golding, W. H. Haemmerle, and M. A. Kastner, Phys. Rev. Lett. **59**, 1148 (1987); A. Vaknin, Z. Ovadyahu, and M. Pollak, *ibid.* **81**, 669 (1998).

[9] Y. Shapir and Z. Ovadyahu, Phys. Rev. B **40**, 12 441 (1989); Y. Zhang and M. P. Sarachik, *ibid.* **43**, 7212 (1991); Shih-Ying Hsu and J. M. Valles, Jr., Phys. Rev. Lett. **74**, 2331 (1995).

[10] P. Dai, Y. Zhang, and M. P. Sarachik, Phys. Rev. Lett. **69**, 1804 (1992).

[11] J. G. Massey and M. Lee, Phys. Rev. Lett. **75**, 4266 (1995); **77**, 3399 (1996).

[12] V. Yu. Butko, J. F. DiTusa, and P. W. Adams, Phys. Rev. Lett. **84**, 1543 (2000). The films used in their study were annealed and oxidized at room temperature. Those films must be thicker than ours because the resistance of our films would increase to infinity during warming up.

[13] A. L. Efros, J. Phys. C **9**, 2021 (1976); J. H. Davies, P. A. Lee, and T. M. Rice, Phys. Rev. B **29**, 4260 (1984); J. H. Davies, Philos. Mag. B **52**, 511 (1985); R. Chicón, M. Ortuño, B. Hadley, and M. Pollak, *ibid.* **58**, 69 (1988).

[14] A. A. Pastor and V. Dobrosavljević, Phys. Rev. Lett. **83**, 4642 (1999); E. Orignac, T. Giamarchi, and P. Le Doussal, *ibid.* **83**, 2378 (1999); P. Kopietz, *ibid.* **81**, 2120 (1999).

[15] D. Simonian, S. V. Kravchenko, M. P. Sarachik, and V. M. Pudalov, Phys. Rev. Lett. **79**, 2304 (1997).

[16] L. A. Yatsuk, Fiz. Nizk. Temp. **8**, 765 (1982) [Sov. J. Low Temp. Phys. **8**, 384 (1982)].

[17] E. E. Semenenko and V. I. Tutov, Fiz. Nizk. Temp. **22**, 666 (1996) [Sov. J. Low Temp. Phys. **22**, 511 (1996)].

[18] P. W. Adams, P. Herron, and E. I. Meletis, Phys. Rev. B **58**, R2952 (1998).

[19] Wenhao Wu, R. G. Goodrich, and P. W. Adams, Phys. Rev. B **51**, 1378 (1995).

[20] E. L. Wolf, *Principles of Electron Tunneling Spectroscopy* (Oxford University Press, Oxford, 1985).

[21] P. Fulde, Adv. Phys. **22**, 667 (1973); P. M. Tedrow and R. Meservey, Phys. Lett. **58A**, 237 (1976).

[22] B. G. Lazarev, L. S. Lazareva, E. E. Semenenko, V. I. Tutov, and S. I. Goridov, Akad. Nauk SSSR **196**, 1063 (1971) [Sov. Phys. Doklady **16**, 147 (1971)].

[23] J. P. Eisenstein, L. N. Pfeiffer, and K. W. West, Phys. Rev. Lett. **69**, 3804 (1992); H. B. Chan, P. I. Glicofridis, R. C. Ashoori, and M. R. Melloch, *ibid.* **79**, 2867 (1997).