Zeeman Splitting of the Coulomb Anomaly: A Tunneling Study in Two Dimensions

Wenhao Wu,* J. Williams, and P. W. Adams
Department of Physics and Astronomy, Louisiana State University,
Baton Rouge, Louisiana 70803
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We report measurements of the tunneling density of states (DOS) of ultrathin Al films in which superconductivity is quenched by a parallel magnetic field, $H_{\|} > 48$ kG. The normal state DOS not only displays the usual logarithmic zero bias anomaly (ZBA) but also a new anomaly, superimposed on the ZBA, that appears at bias voltages $V \sim 2\mu_B H_{\|}/e$, the electron Zeeman splitting. Measurements in tilted fields reveal that the Zeeman feature collapses with increasing perpendicular field component, suggesting that it is associated with the Cooper electron-electron interaction channel. [S0031-9007(96)00791-0]

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It is now well known that disorder has a profound influence on the transport characteristics of one and two dimensional (2D) electronic systems [1]. In particular, the presence of disorder renormalizes the electron diffusivity down from its Boltzmann value [1,2]. This attenuation of the diffusivity with increasing disorder tends to reduce the capacity of the electrons to screen charge fluctuations resulting in an enhancement of the effective strength of electron-electron (*e*-*e*) interactions [3]. In spatially homogenous nondisordered metals, correlations resulting from screened Coulomb *e*-*e* interactions can be straightforwardly incorporated into Fermi-liquid theory by an overall renormalization of the electronic density of states (DOS) [1,3]. Though this renormalization can be significant, the resulting DOS remains a smooth function of energy near the Fermi surface. However, in strongly disordered systems, low electronic diffusivity limits the effectiveness of screening and states near the Fermi surface that were once accessible get "pushed" away by the repulsive Coulomb interaction thereby producing a singular depletion of states near the Fermi energy [3,4], E_F. These singularities have been observed in DOS measurements of disordered metals for many years and are now commonly known as Coulomb anomalies.

In moderately disordered systems, the *e*-*e* interaction correction to the DOS is dominated by two distinct propagation channels. The first is a diffusive particle-hole interaction channel that is known to produce a $\ln(E)$ singularity [3] in the 2D single electron DOS, where E is measured relative to E_F. This singularity is seen in tunneling measurements of the DOS as a logarithmic suppression of the tunneling conductance [5] at zero bias voltage which is often referred to as the zero bias anomaly (ZBA). In this Letter we will be primarily interested in the Zeeman splitting of the Coulomb anomalies in the presence of an external magnetic field. In the case of the particle-hole channel, the contributions to the DOS are different for particle holes with total spin $J = 1$ and $J = 0$. In particular, the presence of a magnetic field should produce only a Zeeman splitting of the $J = 1$ states, independent of field orientation [3].

The second interaction channel is a particle-particle interaction between spin singlet electrons with small total momentum and is commonly known as the Cooper channel. The Cooper channel can, in fact, lead to superconductivity if the effective particle-particle interaction is attractive as a result of electron-phonon coupling. The Cooper channel is expected to produce a $\ln[\ln(E)]$ singularity [3] in the 2D DOS. Since this singularity is significantly weaker than that of the diffusive channel, it has never been directly observed in tunneling experiments. Nevertheless, as with the particle-hole channel, the presence of a magnetic field is expected to produce a Zeeman splitting of this anomaly [3,4]. However, in contrast to the particle-hole interactions, the Cooper interaction anomaly is sensitive to time reversal symmetry and is therefore suppressed by a perpendicular magnetic field.

The Zeeman splitting of the $E = 0$ singularities should appear as satellite singularities in the tunneling conductance at bias voltages corresponding to the Zeeman splitting, $\pm g\mu_B H/e$, where g is the Landé g factor, e is the electron charge, and μ_B is the Bohr magneton. Such satellite singularities should arise from both the particle-hole channel and the Cooper channel if time reversal symmetry is not broken. However, though many tunneling experiments have been performed over the last decade on disordered 2D [5,6] and 3D [7] systems, the Zeeman splitting of the Coulomb anomalies has never been observed. For this reason it has been conjectured that the triplet $(J = 1)$ contribution to the particle-hole channel is small in most disordered metals [8]. Furthermore, since there have been virtually no normal state studies of superconducting films in supercritical parallel magnetic fields, the Zeeman splitting of the Cooper channel has never been addressed experimentally. Clearly, given the absence of a Zeeman splitting of the particle-hole channel, the search for a Zeeman splitting of the Cooper channel is a crucial test of the *e*-*e* interaction theory. Furthermore, the prospect of isolating and studying the Cooper channel singularities in disordered superconducting systems would offer a new and important microscopic window into the superconductor-insulator transition [5].

In the present Letter we present direct tunneling measurements of the normal state single particle DOS in ultrathin granular Al films. Though our films were superconducting below $T_c = 1.9$ K, we have been able to access the low temperature, $T \ll T_c$, normal state DOS spectrum by applying a parallel magnetic field to suppress superconductivity. Aluminum is an ideal system for these studies in that it has a low spin-orbit scattering rate and its parallel critical field, $H_{c\parallel} \sim 48$ kG, is easily accessible [9,10]. We chose a parallel magnetic field orientation for these studies in order to preserve time reversal symmetry in the 2D transport by virtue of the fact that the perpendicular component of the applied field, $H_\perp$, is near zero. In the data presented below not only did we observe the logarithmic ZBA previously reported in Al films [6] as well as other 2D systems [5] but we have for the first time observed the Zeeman splitting of the Cooper channel anomaly.

Electron tunneling measurements were made on ultrathin granular Al films that were fabricated by quench condensation of Al onto 77 K glass substrates. The sheet resistance R of the films was monitored during evaporation in order to produce films with $R > 1$ k$\Omega/\square$. Tunnel junction counterelectrodes were formed by first evaporating a 16 nm thick Al strip onto the glass slide over which the ultrathin film was later deposited. A tunnel barrier was formed by allowing the strip to oxidize in air for ~5 min. The junction area was 1.5 mm $\times$ 1.5 mm. Care was taken to ensure that tunnel junction resistance $R_J \gg R$ so that the voltage drop occurred predominantly across the junction barrier. The samples were mounted in a dilution refrigeration system equipped with a 90 kG superconducting magnet. Both dc and ac tunneling *I-V*'s were measured in a four-probe configuration. The film orientation relative to the applied field was controlled by an *in situ* mechanical rotator. The perpendicular critical field of the films [10] was ~15 kG whereas $H_{c\parallel} \sim 48$ kG. This enabled us to find the parallel field position to within $\pm 0.5°$ by maximizing H_c.

At low temperatures $T \ll eV/k_B$, where V is the bias voltage across the junction, the electron tunneling conductance $G(V) \propto N_f N_{ce}$ gives a direct measure of the microscopic single electron DOS of the film [11], N_f. The interpretation of the tunneling data is somewhat simplified if the tunneling counterelectrode DOS N_{ce} has a featureless spectrum. This was ensured by making the counterelectrode relatively thick, ~16 nm, as compared to the quenched condensed film which had a nominal metallic thickness of ~4 nm. The upper critical field of the counterelectrode was determined to be ~2.5 kG by noting the field at which the tunneling *I-V*'s crossed over from that of a superconductor-insulator-superconductor to that of a superconductor–insulator–normal conductor junction. As a systematic check, we studied 10 nm thick films with a sheet resistance $R \sim 5\Omega/\square$ and found a flat normal state DOS spectrum. Therefore N_{ce} appears only as a constant in the measured tunneling conductance.

Shown in Fig. 1 is the low temperature tunneling conductance as a function of bias voltage near $H_{c\parallel}$ for a film with a normal state sheet resistance $R = 4.2$ k$\Omega/\square$ and $R_J \sim 50$ kΩ. The left inset of Fig. 1 shows the hysteretic critical field that is associated with the first order spin-paramagnetic transition recently reported [10,12] in these films. The curves in Fig. 1 show the evolution of the DOS as the field is increased through the transition. Note that at $H_\parallel = 45$ kG the film was still superconducting and the Zeeman splitting of the BCS density of states is clearly seen. As first reported by Meservey *et al.* [9], the separation in the BCS conductance peaks corresponds to $V = 2\mu_B H_\parallel/e$ indicating that $g \approx 2$ for Al [13]. We have demonstrated this in the right inset of Fig. 1 where the peak positions are fit to the form $V = \Delta/e \pm \mu_B H_\parallel/e$. The y intercept of the fits is a measure of the gap, $\Delta/e = 0.39$ mV. The BCS conductance peaks, clearly evident in the 45 kG curve in Fig. 1, have completely collapsed at 48 kG, indicating that the film has gone into the normal state. Notwithstanding this, the 48 kG curve retains interesting features that we believe are properties of the normal state of the film. In the remainder of this Letter we will focus on the normal state tunneling DOS of the films.

There are two main features of the 48 kG curve of Fig. 1 that are notable. The first is a significant zero bias anomaly characterized by a 40% decrease in the DOS when the

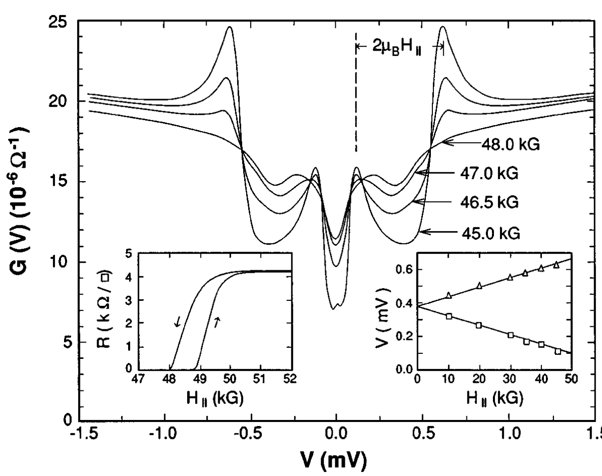

FIG. 1. The 30 mK tunneling conductance of a $R = 4.2$ k$\Omega/\square$ film as a function of bias voltage at several fields near the parallel critical field. At $H_\parallel = 45$ kG the Zeeman splitting of the BCS DOS is clearly observable as peaks in the conductance curve. The position of these peaks is plotted against $H_\parallel$ in the right inset for $H_\parallel < H_{c\parallel}$. The solid lines are fits to the form $V = \Delta/e \pm \mu_B H_\parallel/e$, where $\Delta/e = 0.39$ mV. The left inset shows the hysteretic critical field behavior in R vs $H_\parallel$ at 30 mK. The transition is first order below 270 mK.

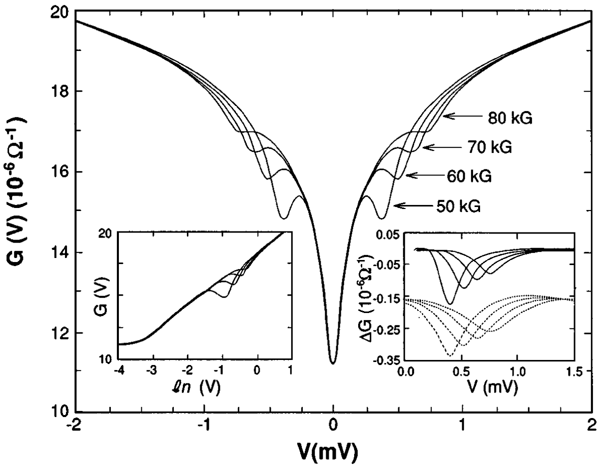

FIG. 2. The main figure is the normal state tunneling conductance of the film in Fig. 1 at several $H_{\|}$. The left inset shows the $\ln(V)$ dependence of the ZBA. The solid curves in the right inset show the Zeeman anomaly with the $\ln(V)$ background subtracted off. The dashed curves are the theoretical DOS of Ref. [3] and have been shifted down for clarity.

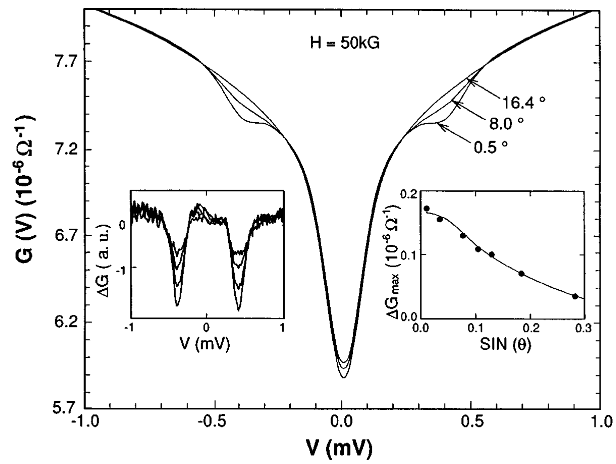

FIG. 3. Tunneling conductance of a $R = 2.0\ \mathrm{k}\Omega/\square$ sample at several field orientations. Parallel position corresponds to $\theta = 0°$. The left inset is a plot of the tunneling conductance curves at $\theta = 11.9°$, $8.0°$, $4.3°$, and $0.5°$ minus the $\theta = 30°$ curve. The dots in the right inset are the magnitude of the Zeeman anomaly as a function of $\sin(\theta)$. The solid line is the theoretical angular dependence from Ref. [3].

bias voltage is lowered from a few mV to zero. The second is a local minimum in the DOS near $V = 0.4$ mV. These satellite anomalies were observed only in samples with $R \geq 1\ \mathrm{k}\Omega/\square$. Furthermore, measurements at higher fields ruled out the possibility of remnant superconductivity as the source of the structure. Shown in Fig. 2 are tunneling conductance measurements at several fields well above $H_{c\|}$. Note that in this range of fields the ZBA is insensitive to $H_{\|}$. However, the satellite feature shifts out and broadens with increasing field. We have numerically integrated the curves in Fig. 2 and found that the total number of states remains constant to within a few percent. From the left inset of Fig. 2 it is evident that the satellite feature is superimposed on a $\ln(V)$ background. This $\ln(V)$ is, in fact, the expected particle-hole channel correction [3,4,6] to the DOS. Therefore, the satellites represent new singularities that arise in the presence of a parallel magnetic field.

We have made a qualitative comparison between our measurements and the theoretical Cooper channel DOS corrections given in Eqs. (6.5)–(6.9) of Ref. [3]. The theory predicts that the Zeeman splitting should be observable in the limit where $T \ll \mu_B H_{\|}/k_B$, $\hbar/k_B\tau_s$, where $\tau_s \sim 1.5e^{-11}$ s is the spin-relaxation time in thin Al films [6]. In fields of a few tesla these conditions are met for temperatures below 0.5 K. Shown as solid lines in the right inset of Fig. 2 are the Zeeman features with the logarithmic background subtracted off. The dotted lines are the predictions of Ref. [3]. Though we did not make a detailed quantitative fit to the theory, it is evident that the theory captures both the qualitative field dependence of the satellite anomalies and the overall relative magnitude of the anomalies. In Fig. 3 we show the dependence of the Zeeman anomalies on the orientation of the magnetic field for a $R = 2.0\ \mathrm{k}\Omega/\square$ sample. The dots in the right inset of Fig. 3 are the magnitude of the Zeeman anomalies as a function of $\sin(\theta)$, where $\theta = 0°$ corresponds to parallel position. The solid line is the predicted angular dependence from Eqs. (6.5)–(6.9) of Ref. [3]. The data in Fig. 3 leave little doubt that the anomalies originated from the Cooper *e*-*e* interaction channel.

In Fig. 4 we have plotted the position of the satellite anomalies as a function of $H_{\|}$ for two different samples. The theory outlined in Ref. [3] unambiguously predicts that the Zeeman anomalies should appear at a bias voltage $V_Z = 2\mu_B H_{\|}/e$. As shown by the solid line in Fig. 4, the position of the anomalies is linear in $H_{\|}$ with a slope

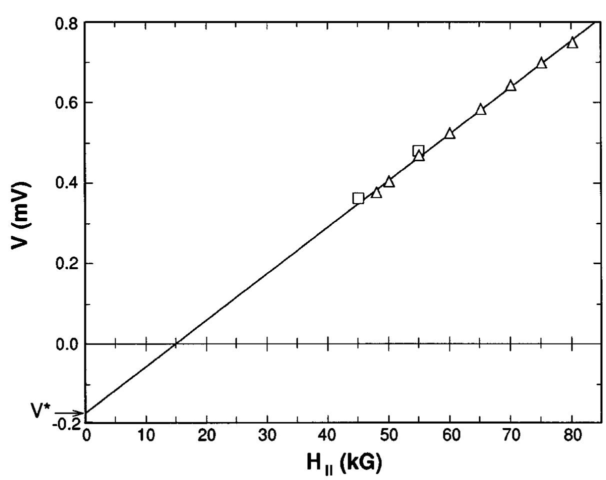

FIG. 4. The position of the satellite features in Fig. 2 as a function of $H_{\|}$ for $H_{\|} > H_{c\|}$. The solid line has a slope of $2\mu_B/e$ in appropriate units. Triangles: $R = 4.2\ \mathrm{k}\Omega/\square$. Squares: $R = 2.0\ \mathrm{k}\Omega/\square$.

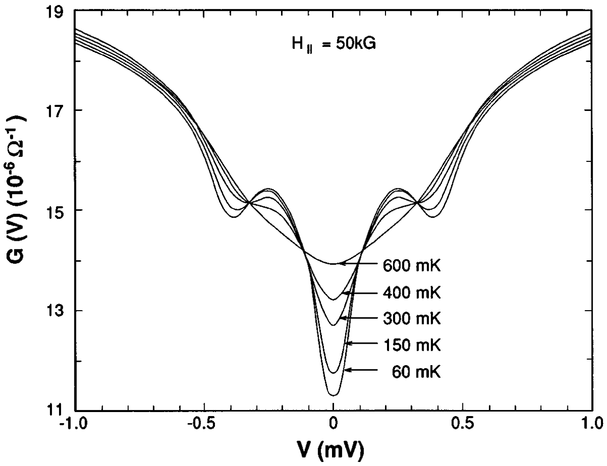

FIG. 5. The normal state tunneling conductance of the film from Fig. 2 at different temperatures. Note the temperature independent point at $V = 0.32$ mV.

of $2\mu_B/e$ as would be expected. However, the data have a finite zero field intercept $V^* \approx -0.17$ mV. This intercept was not due to an offset error, since the ZBA sets the zero of the bias voltage. The intercept in Fig. 4 is significant in that at $H_{\parallel} \sim 50$ kG, the anomaly positions are 33% below V_Z. In fact, V_Z had to be replaced by $V_Z - V^*$ in Eqs. (6.5)–(6.9) of Ref. [3] in order to produce the correct anomaly positions in the right inset of Fig. 3.

It is interesting that $V^* \sim -\Delta/2e$. This suggests that V^* may, in fact, be a measure of the effective attractive *e*-*e* interaction energy in the Cooper channel. We point out that because the experiments were carried out at low temperatures $T \ll T_c$, and in a parallel field, the phonon mediated BCS interaction that leads to superconductivity should have remained intact. In this sense it would be surprising if the effective energy of this interaction were *not* seen in the tunneling spectrum. Finally, in Fig. 5 we show the 50 kG tunneling spectrum of the 4.2 k$\Omega/\square$ film at several temperatures. Note that both the ZBA and the satellite anomalies are almost completely washed out at temperatures above 0.5 K. These data underscore the importance of studying the normal state properties of superconductors at temperatures well below T_c.

In conclusion, we have made the first direct measurements of the Cooper channel *e*-*e* interaction anomaly in a disordered 2D superconductor. Future studies of the Cooper channel anomaly in films with sheet resistances near and above the quantum pair resistance $R_Q = h/(2e)^2$ will provide a unique and fascinating microscopic probe of the role of attractive *e*-*e* interactions in the superconductor-insulator transition [5,10,14]. Extending these parallel field studies to nonsuperconducting films should also prove to be quite interesting.

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*Current address: Department of Physics and Astronomy, Michigan State University, East Lansing, MI 48824.

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