Exchange Fields and the Finite Bias Tunneling Anomaly in Paramagnetically Limited Superconducting Al Films

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We present an experimental investigation of the finite bias anomaly in the tunneling density of states of superconducting Al films above the paramagnetic limit. We show that the anomaly is a manifestation of a new fluctuation mechanism that forms a pseudogap near the Zeeman energy. The field dependence of the anomaly energy is in good agreement with the recent theory of Aleiner and Altshuler, provided that the proper normal-state Landé g factor is used. We argue that the normal-state g factor is reduced from the bare value of $g_n = 2$ to $g_n \sim 1.7$ by a negative exchange field in the paramagnetic phase. [S0031-9007(99)09200-5]

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A magnetic field oriented along the plane of a thin film superconductor affects superconductivity differently than does a perpendicular field. In a parallel magnetic field, the motion of the electrons in the transverse direction is restricted by the film thickness [1] and as a result the accumulation of the Aharnov-Bohm phase is diminished. If the film thickness is much smaller than the coherence length, then superconductivity is destroyed by virtue of the Zeeman breaking of the Cooper pairs. In metals with a small spin-orbit scattering rate, such as Al and Be, for instance [2,3], the critical field transition occurs when the Cooper pair Zeeman splitting is of the order of a superconducting gap [4]. This is the spin-paramagnetic transition.

Recently there has been a renewed interest in the dynamics and phase diagram of the spin-paramagnetic transition in low dimensional systems [5–7]. This interest stems from the fact that in lower dimensions (i.e., grains and films) the field preserves the time-reversal symmetry (TRS) of the superconductor, allowing one to probe quantum phenomena for which this symmetry is essential. In the case of films, a parallel field maintains TRS which produces a spin-paramagnetic transition that is first order [5,8] and fundamentally hysteretic [9–11] at low temperatures. In fact, recent tunneling experiments have shown that the electronic density of states (DOS) of the film itself is hysteretic at the transition [12]. Another unexpected consequence of TRS preservation is that the normal state (paramagnetic phase) of the film retains some coherent quantum features usually associated with the superconducting state. This latter effect is seen as the emergence of a virtual gap near the Zeeman energy in the normal-state DOS spectrum that appears only in the parallel field. When this effect was first reported [13] it was thought to be a ramification of the Zeeman splitting of the Cooper channel e-e interaction channel [14]. However, two important discrepancies arose when a detailed comparison to the e-e interaction theory was made. The first was that the overall size of the effect seemed too large to be associated with the Cooper interaction which has a $\ln[\ln(V)]$ energy dependence. The second problem was that the anomalies were positioned at voltages that were 20%–30% smaller than the Zeeman voltage, V_Z [13]. After the initial experimental report was published, one of the authors and Altshuler reanalyzed the DOS in the paramagnetic phase using nonperturbative techniques [15]. It was found that even though the mean-field BCS order parameter disappears in the paramagnetic phase, a well-pronounced superconducting fluctuation mode still exists. Electrons that tunnel with the proper energy can, in fact, produce a resonant excitation of the mode and thereby cause a strong tunneling DOS singularity [16]. The bias voltage corresponding to the resonant mode and the corresponding DOS singularity, V^*, was predicted to be universal,

$$V^* = \frac{1}{2}\left(V_Z + \sqrt{V_Z^2 - (\Delta/e)^2}\right), \qquad (1)$$

for 0D (grain), 1D (wires), and 2D systems, where $V_Z = g_n \mu_B H_\parallel / e$ is the Zeeman voltage, g_n is the normal-state Landé g factor, μ_B is the Bohr magneton, and Δ is the zero-temperature, zero-field gap energy. The fluctuation mechanism represented by Eq. (1) is new and quite unexpected. Contrary to conventional wisdom, it can and does produce observable pairing effects *in the paramagnetic phase*, even far from the transition. In the present Letter we present electron tunneling studies of the DOS of ultrathin Al films in supercritical parallel magnetic fields. We provide the first quantitative comparison between the above theory and experiment. Furthermore, we present new data which exhibit anomalies that are approximately 3 times larger than those first reported in Ref. [13],

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leaving little doubt that the anomalies are a real manifestation of paramagnetically limited superconductivity.

The Al films used in these experiments were made by thermally evaporating 2–2.5 nm of Al onto fire polished glass microscope slides that were cooled to 84 K. The films had a transition temperature $T_c \sim 2.7$ K, perpendicular critical field $H_{c2} \sim 2$ T, and a tricritical point $T_{\rm tri} \sim 600$ mK [5]. Tunnel junctions were formed by exposing the films to the atmosphere for 0.2–3 hours in order to form a native oxide. Then a 9 nm thick Al counterelectrode was deposited directly on top of the film with the oxide serving as the tunnel barrier. The junction area was 1mm × 1mm. This technique produced tunnel junction resistances $R_j \sim 10$ kΩ to 1000 kΩ depending upon the exposure time and other factors. We were always careful to ensure that $R_j \gg R_{\rm film}$. The integrity of the junctions was tested by measuring the dc I-V characteristics in zero magnetic field at $T = 30$ mK. Under these conditions both the film and the counterelectrode were superconducting and the subgap impedance of a "good" junction was always greater than 10^8 Ω. Because the counterelectrode was relatively thick, its parallel critical field was ~2.7 T, whereas the film's critical field was typically ~5.8 T. Therefore all of the tunneling data presented are either *normal-insulator-superconductor* or *normal-insulator-normal* tunneling. The films were aligned to within 0.1° of parallel by an *in situ* mechanical rotator.

At low temperatures the tunnel junction conductance is proportional to the DOS of the film [17]. In Fig. 1 we show the tunnel conductance of 1 kΩ/sq Al film near the parallel critical field. The curve with the two large peaks on either side of $V = 0$ is, in fact, representative of a superconductor in which the usual BCS DOS has been Zeeman split by the field. As first reported by Meservey *et al.* [2], the BCS conductance peaks are positioned at $V = \Delta/e \pm g_s \mu_B H_\parallel/e$, where g_s is the quasiparticle g factor. By measuring the peak separations as a function of $H_\parallel$ we determine $g_s \sim 2$. Similar measurements of the mean peak positions give an extrapolated zero-field gap of $\Delta/e = 0.45$ mV. The curve with the three dips is the normal-state tunneling spectrum. The origin of the unusual features of this spectrum is the primary focus of this Letter.

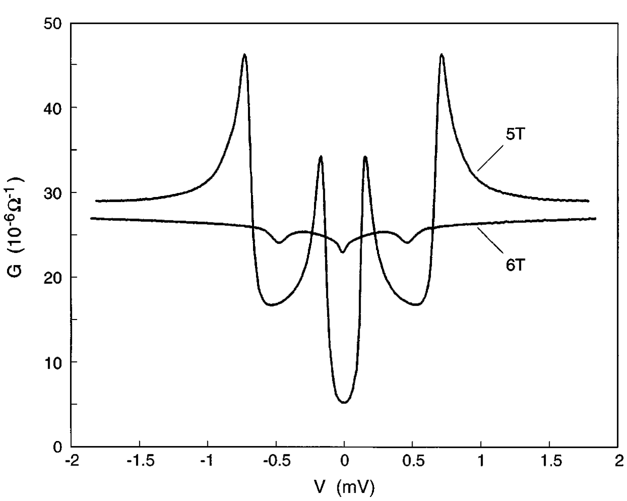

FIG. 1. Tunneling conductance of a 1 kΩ/sq Al film in the normal and superconducting states at 30 mK. The 5 T curve represents the Zeeman split BCS tunneling density of states.

In Fig. 2 we show the normal-state tunneling spectrum at several supercritical parallel fields. There are two main features of the curves of Fig. 2 that are interesting. The first is a significant field independent zero-bias anomaly characterized by a 15% decrease in the DOS when the bias voltage is lowered from ~2 mV to zero. The second is a sharp local minimum in the DOS at a finite field dependent voltage. The anomaly at zero bias is due to e-e interactions and has the expected $\ln(V)$ dependence [13,14]. However, the satellite anomalies must be of a different origin. Because these anomalies move out to higher voltages with increasing field, we can be sure that they are *not* due to remnant superconductivity. Also we see the anomalies at fields that are far above the spin-paramagnetic limit. Furthermore, in contrast to the zero-bias anomaly, the Zeeman anomalies are very sensitive to field orientation and disappear when the sample is tilted just a few degrees out of parallel alignment; see the inset of Fig. 2.

A crucial test of the fluctuation mechanism described in Refs. [15] and [16] is to verify that Eq. (1) correctly predicts the position of the Zeeman anomalies as a function of $H_\parallel$. However before applying Eq. (1) we must first determine g_n. In principle this can be done by comparing

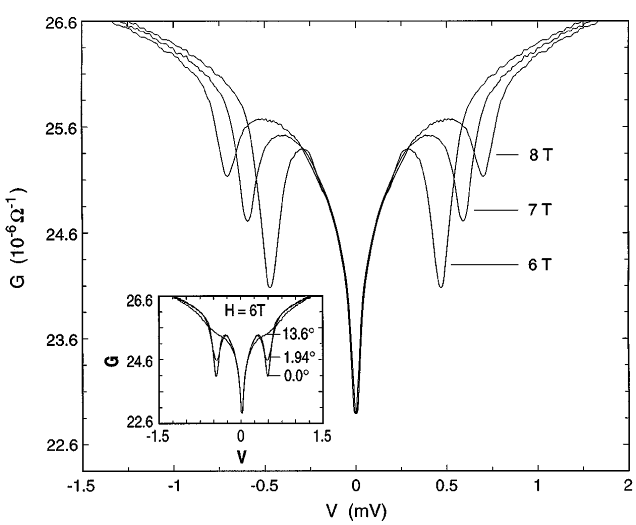

FIG. 2. Normal-state tunnel conductance spectrum at 30 mK. The suppression of the conductance at zero bias is due to the usual $\ln(V)$ e-e interaction anomaly. The satellite features are due to the superconducting fluctuation anomaly discussed in the text. The inset shows the attenuation of the satellite features when the film is rotated out of parallel alignment; $\theta = 0.0$ corresponds to parallel orientation.

the measured parallel critical field with the Clogston-Chandrasekhar [4] critical field $H_{c\|}^{CC} = \Delta/\sqrt{2}\mu_B$. Shown in Fig. 3 are the critical parallel fields as a function of temperature for the film used in Figs. 1 and 2. The critical field transition is hysteretic at low temperatures; thus the two symbols in the plot represent up-sweep and down-sweep transitions. The dashed line is the Clogston-Chandrasekhar critical field assuming $g_n = 2$. Note that the measured critical fields are significantly higher than $\Delta/\sqrt{2}\mu_B$. This discrepancy was first reported in Al films nearly 30 years ago [1]. It has been suggested that it is a consequence of spin-orbit scattering [1]. However, direct measurements of the spin-orbit scattering rate in Al give values that are too small to account for $H_{c\|}$ [1,18]. Furthermore, the data in Fig. 3 suggest a tricritical point at $T_{\rm tri} \sim 600$ mK which is inconsistent with a significant spin-orbit scattering rate [1]. We propose that the discrepancy is an indication that the effective normal-state g factor is significantly less than 2. This is in agreement with recent spectroscopic measurements of Zeeman splitting of discreet electronic states in nanoscale Al grains which also show $g_n < 2$ [6]. It is not obvious how $g_n < 2$ in the normal state when we find $g_s \sim 2$ in the superconducting state. One possible explanation is that there is a significant electron exchange field [19] in the Al films. If this were the case, then one would expect that the exchange field would be greatly diminished in the superconducting phase due to the fact that most of the relevant electronic density would be in paired spin singlet states. Of course, in the normal state the electrons are polarized near the Fermi surface, and exchange effects could be significant. In terms of a dimensionless exchange parameter, γ, the effective g factor can be written as $g_n = 2/(1 - \gamma)$. Now the Clogston-Chandrasekhar critical field becomes

$$H_{c\|}^{CC} = \sqrt{(1 - 8)}\,\Delta/\sqrt{2}\mu_B\,. \qquad (2)$$

If we take the measured critical field in Fig. 3 to be the mean of the up-sweep and down-sweep values at 30 mK, $H_{c\|} = 5.8$ T, then we get a negative exchange interaction $\gamma \sim -0.12$ and a corresponding g factor $g_n = 1.75 \pm 0.05$. For comparison, a similar analysis of earlier samples for which $\Delta/e \sim 0.38$ mV and $H_{c\|} \sim 4.9$ T gives $\gamma \sim -0.11$ and $g_n = 1.80 \pm 0.05$.

With these independent measurements of both Δ/e and g_n we can now test the accuracy of Eq. (1). In Fig. 4 we have plotted the position of the satellite anomalies, V^*, as a function of $H_\|(H_\| > H_{c\|})$ for Al films with two different gap values. The triangles in Fig. 4 correspond to a $\Delta/e = 0.38$ mV sample and the squares to the $\Delta/e = 0.45$ mV sample used in Figs. 1–3. The arrows in the figure indicate the measured gap voltage just below the transition. It is interesting that the anomaly voltage is equal to that of the gap at the transition. For reference the solid linear line in Fig. 4 is the Zeeman voltage V_Z using $g_n = 2.0$. The solid curves through the data points are least squares fits to Eq. (1) in which g_n was varied and the measured values of Δ/e were used in the formula. Best fits were obtained with $g_n = 1.75$ and $g_n = 1.65$ for the $\Delta/e = 0.38$ mV and the $\Delta/e = 0.45$ mV data sets, respectively. Clearly, there is quite good agreement between the theory and the data. Furthermore, the g factors obtained from these fits are consistent with the values of $g_n \sim 1.80$ and $g_n \sim 1.75$ obtained from Eq. (2).

In conclusion, we show that the Zeeman anomaly in the DOS of paramagnetically limited Al films is well described

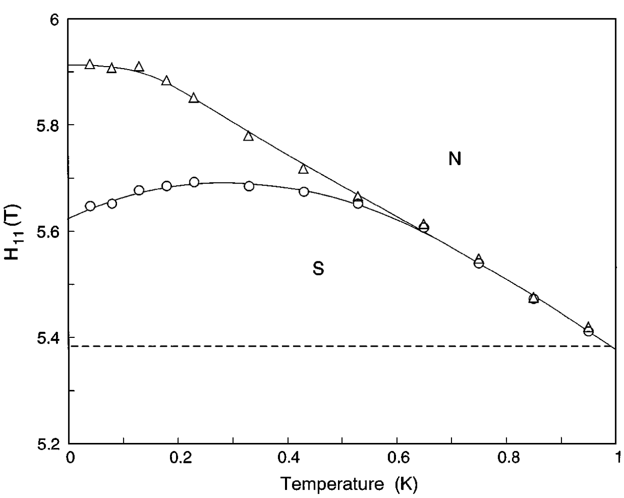

FIG. 3. Parallel critical field values as a function of temperature as measured by the onset of a gap in the zero-bias tunnel conductance. The critical field is hysteretic at low temperatures. Triangles: up-sweep critical field; circles: down-sweep critical fields. The solid lines are provided as a guide to the eye. The dashed line is the expected Clogston-Chandrasekhar critical field with the normal-state g factor equal to 2.

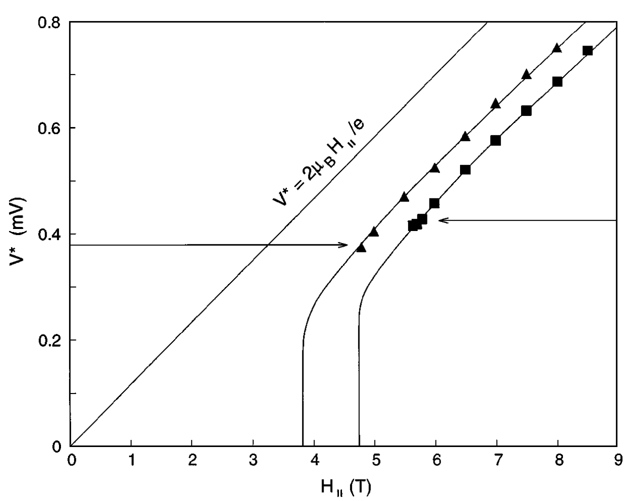

FIG. 4. Voltage position of the satellite anomalies in Fig. 2 as a function of the parallel field. Triangles: $\Delta/e = 0.38$ mV sample; squares: $\Delta/e = 0.45$ mV sample. The linear line is the Zeeman voltage with $g_n = 2$. The curved lines are least square fits of Eq. (1) to the data in which g_n was varied for the best fit. The arrows indicate the measured gap voltages at the transition.

by the quantum fluctuation mechanism of Refs. [15] and [16]. These fluctuations and their corresponding manifestations in the paramagnetic phase DOS are highly singular and, in fact, are much larger than one would expect from usual electron interactions via the Cooper channel. It would be interesting to search for the anomaly in other low spin-orbit scattering superconductors such as nongranular Be films. A systematic investigation of the effects of dimensionality should also prove interesting. This is especially true for superconducting grains where the fluctuation anomaly is expected to produce a hard gap at V^*.

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