Hall conductivity dominated by fluctuations near the superconducting transition in disordered films

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We have studied the Hall effect in superconducting tantalum nitride films. We find a large contribution to the Hall conductivity near the superconducting transition, which we can track to temperatures well above Tc and magnetic fields well above the upper critical field, Hc2(0). This contribution arises from Aslamazov-Larkin superconducting fluctuations, and we find quantitative agreement between our data and recent theoretical analysis based on time dependent Ginzburg-Landau theory.

I. INTRODUCTION

Thin superconducting films are characterized by reduced dimensionality and short coherence length, both of which contribute to the enhancement of fluctuations of the superconducting order parameter above the transition temperature. These fluctuations are expected to affect both thermodynamic and transport measurements. Properties such as the specific heat, magnetization and electrical conductivity in the vicinity of the superconducting transition have been studied both experimentally and theoretically. In particular, fluctuation effects on the electrical conductivity were first discovered by Glover and the diagonal elements of the conductivity tensor (or paraconductivity) are now well understood by Maki and Thompson. However, comparable experimental studies of the off diagonal (Hall) conductivity have been relatively limited in scope.

In this paper we investigate the longitudinal and Hall conductivities of ultrathin disordered tantalum nitride (TaNx) films as a function of perpendicular magnetic field close to and above the zero field critical temperature Tc(H = 0). Although both the longitudinal (Rxx) and Hall (Rxy) resistances vanish in the superconducting state, we find an enhanced Hall resistance above the superconducting transition temperature Tc(H). Such an enhanced resistance can be understood by considering dominant contributions of time-dependent fluctuation effects to the full conductivity tensor above Tc.

The Hall effect at temperatures near Tc has been studied in thin films of conventional superconductors such as MoSi, MoGe, NbGe, and amorphous InO as well as in strongly anisotropic cuprate superconductors. Nevertheless, it is not well understood and continues to be a topic of active research. For example, an unexpected sign reversal of the Hall voltage near Tc has sparked considerable debate (see e.g. Refs. [11] and [12] and references therein). However, all of these studies have been complicated by vortex physics below Tc or by contributions from the normal state Hall effect. A number of microscopic and phenomenological studies have considered contributions due to vortices, pinning effects, and superconducting fluctuations. Efforts to reconcile these studies have been hampered by the difficulty of probing fluctuation effects in the Hall conductivity in conventional superconducting films. Challenges include the combination of high carrier concentration and large longitudinal resistance typical for such systems. Thus, no conclusive picture for the effect of fluctuations of the superconducting order parameter on the Hall conductivity in the normal phase has been reached. In this paper we study field-dependent fluctuation effects in a regime where they may be unambiguously separated from vortex physics and from distinct normal state contributions. Thus, we provide a complete description of the Hall effect in a thin disordered film close to the phase transition into the superconducting state.

II. EXPERIMENTAL BACKGROUND

Tantalum nitride films were prepared using sputter deposition onto a Si substrate. Sample composition was analyzed using x-ray photoemission spectroscopy and determined to be 50±10 at. % N. Sample thicknesses were well controlled using the sputtering time and confirmed via x-ray reflectivity and TEM measurements; the sample thickness d is 4.9 nm. X-ray diffraction analysis showed no sign of crystalline order, and surface analyses showed no signs of granularity or inhomogeneity. Samples were patterned into Hall bar devices using standard optical photolithography techniques and Ar-ion etching, and Ti-Au electrical contact pads were deposited using electron beam evaporation. The active area of the devices is 400 µm × 100 µm. Linear longitudinal and Hall resistance were measured using standard four-point low frequency lock-in techniques in perpendicular magnetic fields; care
Rxx (Ω/□)

1000

750

500

250

0

Rxx (Ω/□) versus Temperature (K)

\[ \frac{\delta\sigma_{xx}}{\sigma_n} = \frac{e^2}{16\hbar} \ln \left( \frac{T}{T_c} \right). \] (1)

In amorphous films with moderate disorder, the interaction between quasiparticles and superconducting fluctuations leads to an additional singular contribution to the conductivity. Similar to the AL term, this contribution, known as the Maki-Thompson (MT) term \[ \delta\sigma_{xx}^{MT} = \frac{e^2}{8\hbar} \ln \left( \frac{\ln T/T_c}{\hbar/k_B T} \right) \ln^{-1} \left( \frac{T}{T_c} \right). \] (2)

This contribution is expected to be less significant in inhomogeneous systems. Note that the two expressions given above correspond to films in which the superconducting fluctuations are essentially two-dimensional (2D), while the quasiparticles are three dimensional.

In Fig. 1 we present the zero-field superconducting transition of sample 1; the normal state sheet resistance \( R_{xx}^n = 0.94 \text{kΩ/□} \). Also shown in Fig. 1 is the resistance measured in an applied field of 8 T, well above \( \mu_0 H_{c2}(0) \sim 5 \text{T} \). The TaNx film studied in this work can
be treated as two-dimensional with respect to superconducting fluctuations. Dense measurements of the resistive transition as a function of temperature and applied perpendicular magnetic field near $T_c$ on sample 2, shown in Fig. 2, were used to extract $\mu_0 d H c_2 / d T \approx 1.7 \ T/K$ near $T_c$. The superconducting coherence length $\xi(0) \approx 8.4 \ nm$ is larger than the film thickness, $d = 4.9 \ nm$. Hall measurements indicate a carrier density of $n \approx 9.1 \times 10^{22} \ cm^{-3}$ for both samples, from which we find that the bulk penetration depth is $\sim 20 \ nm$. In the presence of disorder the penetration depth increases to $\lambda \sim 140 \ nm$, while for a 2D film the relevant magnetic screening length becomes $\lambda_x = \lambda_x^2 / 2d \sim 1970 \ nm$. The mean free path is estimated to be $\ell \approx 0.2 \ nm$. Measured and calculated film parameters for both samples are summarized in Table I.

Adding the AL correction given in Eq. (1) to the normal state resistance (described in the Appendix), we fitted the zero-field resistance as a function of temperature. In the main panel of Fig. 1 we show that away from to $T_c$ the AL contribution dominates as expected for this class of dirty superconducting films. As the transition is approached, however, the AL expression no longer fits the data. The divergence of the conductivity is stronger than expected from Eq. (1), suggesting that the system is inhomogeneous. The inset of Fig. 1 shows that close to $T_c$ the conductivity diverges as $\sigma_{xx} \sim (\ln T / T_c)^{-1.33}$ which corresponds to pure AL contributions on a percolating cluster. (Note that we do not expect the presence of any such inhomogeneity effects to influence the Hall conductivity, as was previously shown by Landauer from geometrical considerations and by Shimshoni and Auerbach when quantum effects are included.) An additional explanation for the departure from the AL term close to $T_c$ can be attributed to the onset of critical fluctuations. This is because the reduced temperature $\ln T / T_c$ is comparable to the Ginzburg-Levanyuk number $G_i$ for this film:

$$G_i = \frac{2 \pi k_B T_c \kappa^2}{\mu_0 \xi(0) \delta d} \sim 0.06$$  \hspace{1cm} (3)

where $\kappa$ is the Ginzburg-Landau parameter.

Since in this film the superconducting transition is broadened, unambiguous determination of the transition temperature $T_c$ is difficult. This large uncertainty draws into question any quantitative analysis that relies on a precise value for $T_c$. For example, the inset of Fig. 1 shows the calculated fluctuation conductivity $\delta \sigma_{xx} = \sigma_{xx} - \sigma_n$ for three different choices of $T_c$. Although all three $T_c$ values fall in the range of temperatures in which the resistivity drops toward zero, the behavior of $\delta \sigma_{xx}$ as the temperature approaches $T_c$ are different in each case. Many studies use fits to AL theory to extract $T_c$, but the AL fits with $T_c = 2.7 - 2.8 \ K$ shown in Fig. 1 are inconsistent with the fluctuation conductivity in our films, and we can only roughly determine that $T_c$ is about $2.75 \ K$ for this sample. Our analysis of the field dependent Hall effect that follows is acutely sensitive to $T_c$, and this approach may be a particularly useful probe of this parameter.

**IV. HALL EFFECT**

We turn now to the transverse resistance measurements. Figure 3 shows the longitudinal and Hall resistance of sample 1 as a function of applied perpendicular magnetic field, at temperatures close to and above $T_c$. In the normal phase of a homogeneously disordered film like TaN$_x$, the Hall conductivity is determined by the fluctuations of the order parameter in addition to the quasiparticles, because vortex physics is not relevant. Thus, it is reasonable to expect that at temperatures above $T_c$ the deviation of the Hall conductivity from the normal state linear-magnetic-field dependence can be attributed to the fluctuations of the order parameter alone.

Fluctuation contributions to the Hall conductivity have been studied using a number of different formalisms. From phenomenological considerations, Lobb et al. parameterize the Hall conductivity at temperatures near $T_f$ with terms proportional to the magnetic field $H$ and $H^{-1}$

$$\sigma_{xy}(H) = \frac{c_1}{H} + c_2 H,$$  \hspace{1cm} (4)

which is intended to interpolate between the low-field region where $\sigma \sim H^{-1}$, and high fields where $\sigma \sim H$. We find that this simple form does not account for the Hall conductivity seen in TaN$_x$ above $T_c$.

Recent theoretical studies of superconducting fluctuation contributions to the Hall effect have extended...
previous calculations of the Hall conductivity\textsuperscript{15,20} to a broader range of temperatures and magnetic fields. Close to the critical temperature, Ref.\textsuperscript{27} shows that the fluctuation Hall conductivity $\delta\sigma_{xy}$ for a broad range of magnetic fields is

$$\delta\sigma_{xy} = \frac{2e^2k_BT\zeta}{\pi\hbar}\sum_{N=0}^{\infty} \frac{(N+1)(\mathcal{E}_N+1-\mathcal{E}_N)^3}{\mathcal{E}_N\mathcal{E}_N+1(\mathcal{E}_N+\mathcal{E}_N+1)^2}$$  

(5)

The function $\mathcal{E}_N$ describes the superconducting fluctuations in the diffusive regime:

$$\mathcal{E}_N(\omega, H, T) = \ln \left( \frac{T}{T_{c0}} \right) + \Psi \left( 1 + \frac{-i\omega + \Omega_c(N + 1/2)}{4\pi k_BT} \right)$$  

$$- \Psi \left( 1 + \frac{1}{2} \right) + \zeta \omega.$$  

(6)

The spectrum of these collective modes is determined by the equation $\mathcal{E}_N(\omega, H, T) = 0$. Here, $\Psi$ is the digamma function, and $\Omega_c = 4|e/\mu_0H|D$ is the energy of the cyclotron motion corresponding to the collective modes (where $D$ is the diffusion coefficient and $k_B$ is Boltzmann’s constant). Since the superconducting fluctuations carry charge, the magnetic field quantizes their spectrum. This is reflected in the sum over the index $N$ appearing in Eq. (5). The parameter $\zeta = -\frac{1}{2}\frac{\partial \ln T_c/\partial \mu}{\hbar} \times 1/\gamma\varepsilon_F$ (where $\varepsilon_F$ is the Fermi energy and $\gamma$ the dimensionless coupling constant of the attractive electron-electron interaction that induces superconductivity) describes the particle-hole asymmetry of the superconducting fluctuations.\textsuperscript{22,23} For a film with three-dimensional electrons and a simple electron spectrum $\zeta$ is negative. This parameter, which is essential for the Hall effect, is nonzero due to the energy dependence of the quasiparticle density of states.

The expression given in Eq. (5) corresponds to the AL contribution to the Hall conductivity in the region of classical fluctuations, meaning that it is valid as long as $\mathcal{E}_0(\omega = 0, H, T) \gg 1$. The contributions of the MT kin\textsuperscript{13} to the Hall conductivity are less singular and can be disregarded as the transition is approached. In the limit $H \to 0$, our result coincides with the one found in Ref.\textsuperscript{20}

$$\delta\sigma_{xy} = \frac{e^2\Omega_c \text{sign} H}{96\ln^2 T/T_{c0}}.$$  

(7)

To fit the experimental data collected at temperatures close to $T_c$ with Eq. (5), we calculated the Hall conductivity $\sigma_{xy} = -R_{xy}/(R_{xx}^2 + R_{xy}^2)$. Far from the transition, at high magnetic fields and/or temperatures, the measured Hall resistance is linear in the applied magnetic field and weakly temperature dependent. This behavior is expected at temperatures $T \gg T_c$ and/or high magnetic fields where the superconducting fluctuations are insignificant. Subtracting the normal (linear in magnetic field) component of the Hall conductivity, $\sigma_{xy}^n$, leaves only the fluctuation contribution $\delta\sigma_{xy}$ that is sensitive to the onset of superconductivity

$$\delta\sigma_{xy} = \sigma_{xy} - \sigma_{xy}^n.$$  

(8)
TABLE II: The best fit parameters, $T_{c0}^{fit}$, $D$, and $k_B\zeta$, for both samples.

| Sample | $T_{c0}^{fit}$ (K) | $D$ (cm$^2$/s) | $k_B\zeta$ (K$^{-1}$) |
|--------|-------------------|----------------|---------------------|
| 1      | 2.60±0.05         | 0.52           | 3.4×10$^{-4}$       |
| 2      | 2.53±0.05         | 0.59           | 4.6×10$^{-4}$       |

In the temperature range of interest the Hall resistance measured at $\mu_0H = 14$ T changes by $\sim 1\%$; the analysis that follows is insensitive to this slight temperature dependence. We determine the normal state $\sigma_{ny}(B) = \sigma_{xy}(B = 14$ T$)/14$ T$$. (The analysis that follows is not sensitive to the exact description of the longitudinal normal state resistance, described in the Appendix.) The fluctuation Hall conductivity $\delta\sigma_{xy}$ calculated for sample 1 is shown in Fig. 4 along with the fits to Eq. (7); the inset shows similar data and best-fit curves for sample 2. The theoretical calculations are in good agreement with the data over a wide range of temperatures and fields.

In fitting Eq. (5) for each sample, three parameters are used for the entire set of Hall conductivity curves: $T_{c0}^{fit}$, $D$ (the diffusion coefficient that enters $\Omega_c$), and $k_B\zeta$. For sample 1, the best-fit parameters are (i) $T_{c0}^{fit} = 2.60±0.05$ K, comparable to that determined from the $R_{xx}$ versus $T$ analysis ($T_{c0} \sim 2.75$ K), (ii) the diffusion coefficient $D = 0.52$ cm$^2$/sec, and (iii) the parameter $k_B\zeta = -3.4 \times 10^{-4}$ K$^{-1}$. Note that with a zero-temperature coherence length extracted from $H_{c2}$ we expect $D \sim \xi^2k_BT_{c0}/\hbar \approx 0.5$ cm$^2$/sec. Estimating $\zeta$ from the carrier density and taking $\gamma \approx 0.2$ which is suitable for this material, we obtain $k_B\zeta \sim -10^{-4}$ K$^{-1}$, in agreement with the values obtained from the fit. As is evident from Fig. 4 at $T = 2.6$ K $\approx T_{c0}$ the magnitude of $\delta\sigma_{xy}$ sharply increases as the magnetic field decreases and the transition into the superconducting state approaches. As shown in the inset to Fig. 4 the calculated fluctuation conductivity and fits to Eq. (5) for sample 2, are almost identical to sample 1. Best-fit parameter values for both samples are listed in Table II.

The above fitting procedure is acutely sensitive to $T_{c0}$ due to the stronger divergence of $\delta\sigma_{xy}$ as the transition is approached [compare Eqs. (1) and (7)], and provides a precise and clear route to extracting $T_{c0}$.

V. CONCLUSION

In summary, we have performed careful studies of the longitudinal and Hall conductivities at temperatures near and above the zero-field superconducting transition in disordered films of TaN$_x$. Studying fluctuation effects in the Hall conductivity is an experimental challenge in systems with high carrier concentration and large longitudinal resistance. These measurements appear to be consistent with theoretical analysis over a wide range of temperatures and magnetic fields. Observation and verification of this effect may facilitate more careful studies of superconducting contributions to the Hall effect and provide a direct route to extract information about the particle-hole asymmetry of the superconducting fluctuations through the parameter $\zeta$. Finally, such an analysis provides a more precise technique for estimation of the temperature where the gap closes.

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Appendix: Estimation of the normal state resistance

In many studies the normal state resistance is either weakly temperature dependent or determined experimentally by applying a large magnetic field to suppress...
superconductivity and assuming a negligible normal-state magnetoresistance (MR)\textsuperscript{23} In our samples, a large nonclassical MR prohibits such an approach, and so we need a well defined procedure to account for it. Figure 5 shows the resistance versus temperature of sample 1 up to 30 K for various values of applied magnetic field. The lower panel of this figure shows the MR, defined as

\[
\frac{\Delta R}{R}(T, H) = \frac{R_{xx}(T, H) - R_{xx}(T, 0)}{R_{xx}(T, 0)}, \tag{9}
\]

calculated using these same data. According to Kohler’s rule\textsuperscript{24}, the classical normal state MR should be a universal function of \(\omega_c \tau\), and in the low-field limit the MR \(\Delta R/R \sim (\omega_c \tau)^2\), where \(\omega_c = eB/m\) is the cyclotron frequency of the electrons, \(\tau\) is their elastic scattering time, \(B = \mu_0 H\), and \(\mu_0\) is the magnetic permeability. For our samples \((\omega_c \tau)^2 < 10^{-6}\), much smaller than the measured MR shown in Fig. 5 and the low temperature MR does not scale as a universal function of \(\omega_c \tau\). While we expect superconductivity effects to give a large MR close to the superconducting transition, this behavior should decay to zero as the temperature or magnetic field are increased. The measured MR at 30 K, well above \(T_c \sim 2.8\) K, is still three orders of magnitude larger than \((\omega_c \tau)^2\), and thus we must describe this large non-classical MR. Our first step in identifying the normal state resistance \(R^n\) is to fit the data at 10 K < \(T < 30\) K and various magnetic fields with the following function:

\[
\frac{\Delta R}{R}(T, H) = A(H)\exp\left[-\frac{T}{T_0(H)}\right]. \tag{10}
\]

By interpolation we can find the phenomenological parameters \(A(H)\) and \(T_0(H)\) for arbitrary fields, and hence, obtain an expression for the normal state MR at all \(T\) and \(H\). To now determine the normal state resistance at zero field, \(R^n(T, H=0)\), we use the expression for the normal state MR given in Eq. 10 and the resistance measured at 8 T:

\[
R^n_{xx}(T, H = 0) = \frac{R_{xx}(T, \mu_0 H = 8T)}{\frac{\Delta R}{R}(T, \mu_0 H = 8T) + 1} \tag{11}
\]

\[
R^n_{xx}(T, \mu_0 H = 8T) = \frac{R_{xx}(T, 0)}{A(H)\exp\left[-\frac{T}{T_0(H)}\right] + 1}. \tag{12}
\]

This approach necessarily recovers the measured zero-field resistance at high temperatures, assuming that deviations from the exponential temperature dependence of the MR at low temperature arise from superconducting fluctuations. Now we are fully equipped to estimate the normal-state resistance for any temperature and magnetic field \(R^n_{xx}(T, H)\) both close and far away from the transition

\[
R^n_{xx}(T, H) = \frac{R^n_{xx}(T, 0)}{A(H)\exp\left[-\frac{T}{T_0(H)}\right] + 1}. \tag{12}
\]

Recent studies\textsuperscript{22} of fluctuation phenomenon in high-\(T_c\) materials have used a similar approach to describe normal state behavior.

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