Survival of superconducting correlations across the 2D superconductor-insulator transition: A finite frequency study

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The complex AC conductivity of thin highly disordered InO\textsubscript{x} films was studied as a function of magnetic field through the nominal 2D superconductor-insulator transition. We have resolved a significant finite frequency superfluid stiffness well into the insulating regime, giving direct evidence for quantum superconducting fluctuations around an insulating ground state and a state of matter with localized Cooper pairs. A phase diagram is established that includes the superconducting state, a transition to a ‘Bose’ insulator and an eventual crossover to a ‘Fermi’ insulating state at high fields. We speculate on the consequences of these observations, their impact on our understanding of the insulating state, and its relevance as a prototype for other insulating states of matter that derive from superconductors.

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INTRODUCTION

A quantum phase transition (QPT) is a zero temperature change of state as a function of some non-thermal parameter (pressure, doping, magnetic field, etc.) \cite{1,2}. The 2D superconductor-insulator quantum phase transition (SIT) is a particularly beautiful and illustrative one, reflecting a transition between the two eigenstates at the extremes of a superconductor’s fundamental uncertainty relation between phase and particle number (\(\Delta \theta \Delta n > 1\)). One of the key questions here concerns the nature of the zero temperature destruction of the superconducting state. Does it proceed in a mean-field fashion by destruction of the amplitude \(\Delta\) of the superconducting order parameter \(\psi = \Delta e^{i \theta}\) or is it dominated by fluctuations of the superconducting phase \(\theta\) and by extension what is the nature of the insulating ground state? The manner in which the superconductivity is destroyed is an issue that has direct relevance to many other important problems including that of high-temperature superconductivity \cite{3,4}.

A related issue is the nature of the insulating state. There have been a number of proposals for an insulator with strong superconducting correlations. For instance, Fisher and co-workers \cite{6,7} postulated a dual description - the so-called ‘dirty boson’ model - of the SIT in which the superconducting state reflects the condensation of Cooper pairs and localization of vortices, while the insulating state is characterized by condensed vortices and localized Cooper pairs. More recently, it has been proposed that the transition is an inhomogeneous one \cite{8}, and where global superconductivity may obtain by percolation of locally superconducting clusters \cite{9}. To date, evidence for superconducting signatures in the insulating state may have been found in an interesting positive magnetoresistance in insulating films which has been interpreted as a sign of vortex activation \cite{10} as well as in a crossover of the Hall coefficient \(R_{xy}\) \cite{11} at a field higher than the superconductor-insulator critical field \(H_{SIT}\). Although these results have been suggestive there has been a notable lack of direct evidence for superconducting correlations deep into the insulator. Moreover a number of outlying issues remain. For instance, the Cooper pair gap appears to close on the approach to the SIT in amorphous films, evidence seemingly incompatible with the existence of localized Cooper pairs \cite{12,13}. Still others have challenged the existence of a direct transition between superconducting and insulating states altogether and instead postulate the existence of an intervening metal \cite{14}.

In this paper we present the results of a comprehensive study of the complex AC conductivity through the nominal 2D SIT in InO\textsubscript{x} thin films using microwave cavities. This is the first such AC study of this system; all previous studies have used DC probes. Here, we have resolved a significant finite frequency superfluid stiffness in the insulating state which persists well into the strongly insulating regime and at fields up to 3 times the critical field, giving direct evidence for a state with localized Cooper pairs. This establishes a phase diagram with distinct regions dominated by superconducting, ‘Bose’ insulating, and ‘Fermi’ insulating effects.

EXPERIMENTAL DETAILS

Samples were 200 Å-thick 3mm-diameter highly-disordered amorphous indium oxide (α-InO) thin films

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prepared by e-gun evaporating high purity (99.999%) \(\text{In}_2\text{O}_3\) on clean 19mm-diameter sapphire discs in high vacuum \[17, 18\]. The deposition methods are essentially the same as in the work of Ref. \[17\] where it was demonstrated that amorphous \(\text{InO_x}\) can be made reproducibly by e-beam evaporation of \(\text{In}_2\text{O}_3\) and then subsequent low-temperature annealing. In this way the end result is quite different than films prepared via other methods that create a granular or nanocrystallite morphology.

Essentially identical films have been used in a large number of recent studies of the 2D superconductor-insulator quantum phase transition \[16, 19–24\]. Post-deposition, samples were room-temperature annealed for approximately one week in ambient air and then, except for short periods, were kept well below LN\(_2\) temperatures for the duration of the measurements (approximately 2 months).

Thin machined aluminum masks were used to pattern the films creating a 200 Å-thick 3mm-wide circular amorphous film centered on the disc. Deposition was well controlled and samples with specific properties can be made reproducibly \[17, 20\]. For structural characterization, we deposit two more films along with the sample: (1) onto a TEM grid for electron diffraction. (2) for AFM scans. We believe the films to be morphologically homogeneous with no crystalline inclusions or large scale morphological disorder for a number of reasons. The TEM-diffraction patterns are found to be diffuse rings with no diffraction spots, suggesting a completely amorphous layer with no crystalline inclusions. Moreover, the AFM images show continuous films with no voids or cracks and are in fact entirely featureless down to the limiting resolution of the AFM (a few nanometers). We also note that the \(R\) vs \(T\) curves when investigating the 2D superconductor-insulator transition \[16\] in these films are smooth with no re-entrant behavior that has been the hallmark of gross inhomogeneity.

Experiments were performed in a novel cryomagnetic resonant microwave cavity system. The cavity diameter was chosen to optimize performance in the 22 GHz (\(\hbar \omega / k_B = 1.06\) K) TE011 mode, but measurements were possible at a number of discrete frequencies from 9 to 106 GHz. Cavities were operated in a low power regime where the cavity response was independent of the input intensity to ensure no sample heating.

Various well-known relations were used to relate the resonances’ frequency shift \(\Delta \omega\) and change in inverse quality factor \(\Delta 1/Q\) upon sample introduction to the complex conductivity \[25–28\]. Extensive details of the data analysis scheme can be found elsewhere \[23\]. We only mention here that since the films are grown in the center of a sapphire disc and are hence centered along the cavity’s axis, we can make use of both in-plane electric fields or perpendicular magnetic fields. The symmetry of the sample’s placement means that, for all TE modes if there is an in-plane electric field at the sample position, the out-of-plane field is zero and vice versa. Different analysis schemes are used for the two possibilities. Again, see Ref. \[23\] for a very thorough discussion of our precise experimental arrangement and analysis scheme. A consequence of the sample’s extreme thinness is that the real part of the conductivity is essentially proportional to the change in inverse quality factor \(\Delta 1/Q\) and the imaginary part of the conductivity is proportional to frequency shift and there is only a weak mixing between
real and imaginary components. We of course used the complete analysis, which can be found in Ref. 23. The conversions from $\Delta$ and $\Delta\omega$ to conductivity were made by normalizing the AC resistance of the cavity data to DC data at temperatures well above the occurrence of superconductivity and insisting that the superfluid stiffness (defined below), was frequency independent at zero field as $T \to 0$.

DC resistance was measured on co-deposited samples in a two-probe configuration by low frequency AC lock-in techniques using excitation currents of 10$pA – 10nA$. The probe’s lead resistances, which have a negligible temperature dependence in the displayed temperature range, were well characterized and have been subtracted from the displayed data.

RESULTS

The zero field DC resistance curve shown in Fig. 1a is fairly typical for a highly disordered superconducting thin film showing an approximately log increase of $R$ with decreasing temperature, before the occurrence of a broad superconducting transition. As shown in Fig. 1b, when the field is increased the resistance curves cross over from a superconducting positive $dR/dT$ to an insulating negative $dR/dT$ behavior similar to previous field-tuned studies 23 to 31. We emphasize the very strong temperature dependence of the insulating state. At low temperatures and 8 $T$ the InO$_x$ films are very strongly insulating with a resistance that is approaching $10^8 \Omega$/□. When plotted as $R$ vs $H$ (Fig. 1c) the experimental data shows a low-temperature iso-resistance crossing point of 3.68 Tesla, which can be identified as the critical field of the SIT, $H_{SIT}$. The DC data can be shown to be consistent with previous studies 23 31 that found scaling as a function of the reduced variable $|H – H_{SIT}|/T^{1/z\nu}$ where $z\nu$ is consistent with the exponent for 2D classical percolation 4.

As the temperature is lowered, distinct regions typified by amplitude and then phase fluctuations of the superconducting order parameter $\psi = \Delta \exp{i\theta}$ are expected. Using the procedure of Gantmakher 23 24, a lower bound on the temperature where the amplitude becomes well defined $T_{c\alpha}$ can be estimated as the lowest temperature (Fig. 1a) that does not cause an inflection point in the extracted effective normal state resistance $R_N(T)$ as defined by the full expression for the Aslamazov-Larkin fluctuation resistivity 22. In two dimensions, this extracted temperature scale $T_{c\alpha} = 2.28\text{K}$ does not signify the occurrence of a phase transition, but instead represents the temperature scale below which the Cooper pair amplitude becomes well defined.

We now turn our attention to the AC conductivity. At low temperatures, the imaginary conductivity for a long-range ordered superconductor is expected to have the form $\sigma_i = N e^2 \omega m$ where $N$ is the superfluid density and $e$ and $m$ are the electronic charge and mass respectively. For a fluctuating superconductor one can define $\sigma_i = N e^2 \omega m^2$ where an additional frequency dependence is captured by a generalized frequency dependent superfluid density. The superfluid density is directly proportional to the superfluid stiffness which is the energy scale for inducing slips in the superconducting phase. The use of a frequency dependent superfluid stiffness or stiffness has been the usual treatment within, for instance, the finite frequency treatment of the Kosterlitz-Thouless-Berezinskii (KTB) transition 32. In Fig. 2a we display the $H = 0$ generalized frequency dependent superfluid stiffness, $T_{\theta}$ (in degrees Kelvin) extracted via the relation $\sigma_i = \sigma_\theta |KW_k/\omega_\theta|^2$, where $\sigma_\theta = \frac{4e^2}{h\omega_k}$ is the quantum of conductance for Cooper pairs divided by the film thickness. We emphasize that the superfluid stiffness $T_{\theta}$ is not a temperature, but is an energy scale expressed in temperature units. The superfluid stiffness curves all cross the predictive line ($T_{\theta} = 4T_{KTB}$) for the KTB transition in a range near 1.6 K, however a frequency dependence is acquired by the superfluid stiffness at a lower temperature of approximately 1.1 K. We associate this lower temperature as the KTB scale below which the superconductor becomes robust against vortex phase fluctuations. A somewhat similar phenomenology has been seen in quasi 2D high temperature superconductors at THz frequencies 5. The $T_{KTB}$ of $\approx 1.1\text{K}$ and $T_{c\alpha}$ of 2.28 K give the temperatures at zero field for phase coherence and a well defined superconducting amplitude, respectively. A detailed study of these fluctuation regimes at $H=0$ can be found elsewhere 23.

Using the BCS relation $2\Delta/k_B T_{c\alpha} = 3.53$ and the measured $T_{c\alpha} = 2.28\text{K}$, all of our displayed operating frequencies are well below the BCS expectation of 168 GHz for above threshold gap excitation at low temperature and zero field. In principle however, there may be a normal electron contribution to the response from both thermally excited electrons as well as from above gap excitations when $\hbar\omega > 2\Delta$ on the approach to $T_c$ or at high fields where the gap’s size may decrease. In practice however, these give a negligible contribution to the $\sigma_\theta$ response in a highly disordered material like ours as the motion of these essentially normal electronic excitations are highly damped. We can give a rough estimate of this contribution by a very approximate calculation as follows.

Mangetoresistance measurements on very similar samples have given a coherence length of approximately 6 nm 20, which is set by the normal state electron mean free path in highly disordered materials. This gives the very large effective scattering rate ($1/\tau$) of approximately 200 - 300 THz (using a rough estimate for the Fermi velocity $(c/200)$), which is reasonable based on the high disorder and large normal state resistivity of our sample. An estimate based on the normal state resistivity, using a free
from "normal" electrons as described in the text. (a) Temperature dependence at H=0.

Using this scattering rate, our definition of $T_\theta$ and its measured low temperature value (7.66 K), and the Drude relations we can estimate the maximum possible contamination contribution to the measured stiffness from “normal" electrons as $T_{\theta,\text{cont}} = T_\theta[T \to 0] \frac{(\omega \tau)^2}{1 + (\omega \tau)^2}$. This is the contribution if all the spectral weight in the superconducting delta function was scattered in a normal state electron-like fashion. At even our highest frequency, this contribution of $2 \times 10^{-6}$ K is far smaller than the experimental sensitivity. Even if the effective scattering rate was overestimated by even a factor of 100 the contamination contribution would only rise to approximately 0.02 K which is still well less than our sensitivity. Hence, the contribution from “normal" electrons is a completely insignificant contribution to $\sigma_2$ and hence $T_\theta$ and we can safely assume that all of our $\sigma_2$ signal is due to a superconducting contribution of some variety.

Displayed in Fig. 3 is the finite-frequency complex response at $H \neq 0$ for the 22 GHz data. Similar plots can be made at other frequencies. Fig. 3a shows the real part of the AC resistance measured over a range of temperatures and magnetic fields. The bold line is a contour defined by the AC resistance at the critical field (defined by the DC measurements) extrapolated to finite temperatures. We identify the region below this critical contour of $R = 2295 \Omega$ (in bold) as the domain over which the underlying $T = 0$ superconducting phase has influence.

In Fig. 3b is shown the superfluid stiffness $T_\theta$ at 22 GHz, using the same conversion as above, and plotted as a function of temperature and magnetic field. We observe that the superfluid stiffness is finite well into the insulating regime and even at fields and temperatures where the DC resistance can be inferred to be over $10^6 \Omega$. The use of relatively high probing frequencies allows us to resolve superconducting fluctuations into the insulating part of the phase diagram. An additional advantage of our high frequencies is that they are at least two orders of magnitude higher than a generous estimate for the vortex depinning frequency found in thin films of conventional superconductors. The vortex contribution to the optical response is therefore expected to be purely dissipative, leaving the superfluid as the only principal contributor to $\sigma_2$. With a low frequency probe one would have the additional complication of being sensitive to the vortex polarization as they are displaced from the pinning sites which would give an additional contribution to $\sigma_2$.

The finite-frequency superfluid stiffness falls quickly with increasing field, but remains finite above $H_{SIT}$, well into the insulating regime to fields almost 3 times the critical field $H_{SIT}$. This is the first direct measure of superconducting correlations well into the insulating side of the 2D superconductor-insulator transition in an amorphous film.

**DISCUSSION**

Our observation of a finite frequency superfluid stiffness at $H > H_{SIT}$ is not inconsistent with an insulating $T=0$ ground state. As alluded to above, our experiments are sensitive to superfluid fluctuations because we probe the system on short time scales via an experimental frequency $\omega_{\text{exp}}$ that is presumably high compared to an intrinsic order parameter fluctuation rate $\omega_{QC}$ close to the transition. Above $H_{SIT}$, an experimental probe in the limit $\omega_{\text{exp}} \ll \omega_{QC}$ will not detect superfluid fluctuations as a system with $T_\theta(\omega \to 0) \neq 0$ can support superfluid flow, behavior obviously incompatible with the notion of an insulator. On general grounds an insulator with $\sigma_1 = 0$ at zero frequency, must have a $\sigma_2$ that is negative at $\omega = 0^+$ by Kramers-Kronig considerations, meaning that such a system can not appear superconducting at low frequencies. It is only by using a relatively high $\omega$. 

**FIG. 2:** (color) Superfluid stiffness, $T_\theta \propto \omega \sigma_2$, extracted as described in the text. (a) Temperature dependence at H=0. Also shown is the theoretical $T_{KTB}$ which intersects $T_\theta$ around $T = 1.1K$. (b) $T= 0.5$ K magnetic field dependence at select low frequencies.

**FIG. 3:** (color) Superfluid stiffness, $T_\theta \propto \omega \sigma_2$, extracted as described in the text. (a) Temperature dependence at H=0. Also shown is the theoretical $T_{KTB}$ which intersects $T_\theta$ around $T = 1.1K$. (b) $T= 0.5$ K magnetic field dependence at select low frequencies.
FIG. 3: (color) (a) AC Sheet resistance ($\propto$ line is the critical resistance extrapolated to finite temperature. (b) Superfluid stiffness ($\propto T$ increasing field and the strong frequency dependence of $2b$ we see the dramatic drop in superfluid stiffness with indicates maximum.

We are able to extract a phase diagram that establishes the existence of superconducting correlations well into the insulating state. In Fig. 4 contours are plotted which denote the region above which our superfluid stiffness becomes almost indistinguishable from the normal state noise level (set at 1% of the $T \rightarrow 0, H = 0$ superfluid stiffness), thereby giving a measure of the extent of superconducting correlations into the insulating regime. The noise is greatest for frequency contours away from our cavity’s optimal operating frequency of 22 GHz, but it is evident that the higher frequency probe at 22 GHz allows one to examine the fluctuations of the order parameter persisting at fields higher than $H_{SIT}$ and temperatures higher than $T_{c0}$, at shorter length and time scales than the 9 or 11 GHz probes. In the high-frequency limit one would expect that the detection limit would eventually extrapolate to a field where the Cooper pairs are completely depaired. In the low frequency limit we would expect that the superfluid density would disappear at $H_{SIT}$. For 22 GHz we observe that the data extrapolates to a value of $H_{c2}$, which is close to the value of the pair breaking field, $H_{c2}$, found in similar films of InO$_x$,

FIG. 4: (color) 2D Field Tuned Superconductor-Insulator “Phase Diagram”. Contours with markers are the detection limit for $T_0$ at specified frequencies. Black dots on the temperature axis denote $T_{amp} (= T_{c0})$ and $T_{phase} (= T_{KB})$, signifying temperatures where the amplitude and phase respectively become well-defined. $H_{SIT}$ appears as a black dot and the contour of critical resistance from Fig. 3a appears as a solid black line. Open symbols represent data obtained from a small linear extrapolation beyond our maximum field of 8 Tesla.
at the extrapolated pair breaking field scale. Additionally it would be important to measure the Nernst effect in InO$_x$ films. Such measurements have recently revealed a finite Nernst signal at fields above $H_{SIT}$ in NbSi that has been interpreted in terms of superconducting fluctuations \[9\].

CONCLUSIONS

We have performed the first AC conductivity study across the 2D superconductor-insulator quantum phase transition. We find evidence for a finite superfluid stiffness at fields well into the insulating phase giving direct evidence for superconducting fluctuations around an insulating ground state. We emphasize that we believe the only true phase transition in the system (neglecting for a moment the possibility of an intervening metal phase) is at $H_{SIT}$ and that the contours in Fig. 4 give a frequency dependent crossover. We observe superconducting correlations not just asymptotically close to the SIT as one might for localized electrons, but an extended region above $H_{SIT}$. This establishes for the first time a model-free, unambiguous picture of an insulating state that is dominated by superconducting correlations - a \textit{‘Bose’} insulator. We conclude that at some level this \textit{‘Bose’} insulator is best described by localized Cooper pairs in contrast to localized electrons. Our phase diagram is characterized by a superconducting ground state, a phase transition to a \textit{‘Bose’} insulating state at $H_{SIT}$ and a crossover to \textit{‘Fermi’} insulator near a depairing field $H_{c2}$. This work raises certain questions about the expected electrodynamic response of the various proposed phases of matter. It might be expected, for instance, that the electrodynamic response of the various proposed phases

Finally, it has not escaped our notice that this work has a direct connection to many current theoretical proposals within the context of high-temperature superconductivity. Our results show in principle that such insulating ground states characterized by substantial quantum superconducting fluctuations can exist and serve as a prototype for other insulating states of matter that derive from superconductors \[11\–14\].

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In contrast to the usual convention in vortex physics, we parameterize out-of-phase current response as $\sigma_2$, and not $\frac{1}{\omega L}$, as the later quantity will diverge in a system with residual dissipation and vanishing superfluid density. Because vortex conductivities add in parallel with the charge response, a weak mixing into $\sigma_2$ of the in- and out-of-phase vortex response is possible which means that our quantity $T_\theta$ needs to be rigorously interpreted as a lower bound on the superfluid energy. However, a simple estimate based on our extracted vortex viscosity shows that this contribution can not be more than a few percent at our highest fields.