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Quasiparticle Screening near a Bosonic Superconductor-Insulator Transition Revealed by Magnetic Impurity Doping

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Experiments show that the Cooper pair transport in the insulator phase that forms at thin film superconductor to insulator transitions (SIT) is simply activated. The activation energy \(T_0\) depends on the microscopic factors that drive Cooper pair localization. To test proposed models, we investigated how a perturbation that weakens Cooper pair binding, magnetic impurity doping, and phase frustration affects \(T_0\). The data show that \(T_0\) decreases monotonically with doping in films tuned farther from the SIT and increases and peaks in films that are closer to the SIT critical point. The observations provide strong evidence that the bosonic SIT in thin films is a Mott transition driven by Coulomb interactions that are screened by virtual quasi-particle excitations. This dependence on underlying fermionic degrees of freedom distinguishes these SITs from those in micro-fabricated Josephson Junction Arrays, cold atom systems, and likely in high temperature superconductors with nodes in their quasiparticle density of states.

What drives Cooper pair localization in films undergoing a superconductor-insulator quantum phase transition (SIT) has not been resolved\(^1\). In some models, the localization arises mainly from disorder induced Anderson localization effects\(^5\) and in others from repulsive Coulomb interaction or Mott transition effects\(^6\)–\(^8\). Experiments have been unable to discern the primary driver despite having established myriad signature characteristics of the Cooper pair insulator state like its giant positive magnetoresistance\(^9\)–\(^12\), islanded structure\(^13,14\) and Cooper pair dominated transport\(^15,16\). Here, we present magnetic impurity doping studies, which reveal that Coulomb interaction effects dominate the Cooper pair insulator transition in a-Bi thin films.

These investigations focus on the activation energy, \(T_0\), determined from the temperature dependence of the sheet resistance of the Cooper pair insulator

\[
R(T) = R_0 \exp(T_0/T) \tag{1}
\]

where \(R_0\) is a constant and \(T\) is the temperature\(^9,17,18\). Motivation for this focus is that the activation energy in a condensed matter system offers a window into its quasiparticle quasihole pairs out of the Laughlin ground state\(^19\)–\(^21\). Similarly, the low temperature heat capacity of conventional superconductors is characterized by an activation energy corresponding to half the binding energy, \(2\Delta\), of electrons in Cooper pairs in the BCS ground state\(^22\). At this point it is known that \(T_0\) for the Cooper pair insulator controls the rate of Cooper pair tunneling between localized states\(^15\) but differs from the Cooper pair binding energy since it grows from zero at the SIT critical point where \(\Delta \neq 0\)\(^15\).

In models of the Cooper pair insulator, \(T_0\) results from a competition between pair tunneling, characterized by a hopping rate \(t\) or a Josephson coupling energy, \(E_J\), that delocalizes pairs and either potential disorder or Coulomb interactions that localize pairs. Potential disorder drives Anderson localization\(^2,3,23\) of pair states with energies below a mobility edge in the density of states. \(T_0\) corresponds to the gap between localized and mobile pair states\(^3,24,25\) and increases with disorder or decreasing \(t\). Coulomb interactions, on the other hand, drive a Mott transition by creating a blockade to pair motion between localized states\(^26\). The blockade is characterized by a charging energy, \(E_c = 2e^2/C\), that depends on the capacitance between a localized state and its environment\(^7,8,26\)–\(^29\). In the limit, \(E_c \gg E_J\), a Mott gap appears in the transport,

\[
T_0 \approx E_c \left(1 - \frac{z E_J}{2 E_c}\right) \tag{2}
\]

with a second term that depends on coordination number \(z\) and \(E_J\) to account for Cooper pair screening\(^7,30\). Measuring how \(T_0\) responds to changes in parameters like \(E_J\) is necessary to test these models of Cooper pair localization. While previous experiments showed that \(T_0\) depends on many factors including magnetic field\(^9,12,18,31\), magnetic frustration\(^32\) and normal state resistance\(^10,33\), the relations between the factors and model parameters have not been defined well enough to compare directly with models.

We have employed a thin film platform\(^15\) that enables unique methods for probing the origins of \(T_0\). The films can be systematically doped with magnetic impurities, which reduces \(2\Delta\) and can be subjected to magnetic frustration, which reduces the average Josephson coupling between localized regions\(^12\) (see Figs. 1 a,b). Since \(E_J \propto \Delta\), the doping also reduces \(E_J\). For both the Anderson and Mott classes of models, reducing \(E_J\) is expected to enhance \(T_0\) and thus, Cooper pair localization. Surprisingly, we found that while magnetic frustration always enhances \(T_0\), magnetic impurity doping can reduce \(T_0\). We discuss how this result intimates that the superconductor to Cooper pair insulator transition is a Mott transition with a Coulomb blockade energy that depends...
on the pair binding energy.

Sub-nanometer thick amorphous Bi films were fabricated and measured in situ in the UHV environment of a dilution refrigerator based evaporator. Bi vapor was quench condensed onto an Sb wetting layer on the surface of two substrates simultaneously: an Anodized Alumina Oxide substrate, which has regular height variations and an array of pores, and a flat, fire polished glass substrate. Both substrates were held at a temperature, $t$, and an array of pores, and a flat, fire polished glass substrate. The 6 peaks thus give rise to 12 dots of thicker film in the peaks and valleys. Since $T_c$ increases with film thickness these dots form an array of sites that localize Cooper pairs in insulating films. Insulating films on flat substrates, by contrast, have only weakly localized, unpaired electrons$^{35,36,37}$. The film on the flat substrate served as a reference for monitoring 1) the maximum thickness and pairing amplitude that could appear in the films deposited on AAO and 2) the pairing impurity depositions.

Film sheet resistances were measured as a function of temperature, $R(T)$ in situ using standard four-point ac and dc techniques with sufficiently low current bias (0.2 nA) to ensure that the measurements were performed in the linear portion of the current-voltage characteristics. A superconducting solenoid applied magnetic fields perpendicular to the films.

The array of pores in the films enable us to explore magnetic field induced frustration effects on the Cooper pair insulator phase. The appearance of oscillations in the magnetoresistance was an early direct sign of localized Cooper pairs in a thin film system$^{15}$. The activation energy and location of the SIT critical point (see Fig. 1c) is periodic in the frustration $f = H/H_M$, where $H_M$ is the magnetic field that produces one superconducting flux quantum per plaquette. The average appears as $< E_J > = \sum_{<i,j>} \cos(\phi_i - \phi_j - A_{ij}) >$ where $\phi_i$ and $\phi_j$ are the phases on neighboring islands and $A_{ij}$ is the line integral of the vector potential between islands. Some array of islands, the energy barrier for Cooper pair transport is highest for $f = 1/2$. Phenomenologically, $< E_J(f) > \propto E_J f(2\pi f)$, where $F$ is a periodic function with maxima of 1 at integer $f$.

Magnetic impurity doping involved depositing Gd atop the Cooper pair insulator film$^{40}$. The impurities produce time reversal symmetry breaking spin flip scattering, which reduces the pair binding energy $2\Delta$. Their effect extends uniformly through the entire thickness of the films since the films are much thinner ($d \leq 1$ nm) than the superconducting coherence length ($\xi \geq 10$ nm)$^{41}$. The Gd deposition amounts, $x_{Gd}$, were below the micro-balance resolution, were monitored using a calibrated timing method and by measuring their effects on the $T_c$ of the reference film. The two methods agreed well. In the following, the relative $T_c$ shift on the reference film

$$\alpha_{Gd} = 1 - T_c(x_{Gd})/T_c(0)$$

(4)

to represent the pair breaking strength. The estimated maximum Gd doping in these experiments corresponded to $< 0.03$ monolayers.

We studied the effects of magnetic impurity doping and magnetic frustration on two films, I and II, that had different activation energies to explore how proximity to the SIT critical point influences the response. Points for films I and II are indicated on the schematic phase diagram in Fig. 1c, according to their relative activation
energies obtained from fits to the data shown in Fig. 1d. Other film I and II parameters are in the Table. The phase diagram shows two distinct critical points for the two frustrations investigated, \( f = 0 \) and \( f = 1/2 \). \( \delta T \) work where \( R_N \) is sheet resistance measured at 8K. Previous work\(^{15} \) indicated that the critical values of the tuning parameters for the SIT followed \( \delta T_0 > \delta T_1/2 \).

**TABLE I. Film I and II parameters.**

<table>
<thead>
<tr>
<th></th>
<th>( R_N )</th>
<th>( d_{Bi} )</th>
<th>( T_0(0) )</th>
<th>( T_0(1/2) )</th>
<th>( T_c(0) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>18.6 kΩ</td>
<td>0.99 nm</td>
<td>0.86 K</td>
<td>0.98 K</td>
<td>2.59 K</td>
</tr>
<tr>
<td>II</td>
<td>16.7 kΩ</td>
<td>1.2 nm</td>
<td>0.40 K</td>
<td>0.75 K</td>
<td>2.92 K</td>
</tr>
</tbody>
</table>

The arrows indicate the pairbreaking strengths corresponding to \( \delta T_0 = T_0(1/2) - T_0(0) \) larger for film II, which is closer to the SIT, shows that the frustration effect grows with the interisland tunneling rate.

By contrast, \( T_0 \)'s doping dependence does not align with simple expectations for three cases (film I at \( f=0 \) and \( f=1/2 \), and film II at \( f=1/2 \)). Pairbreaking reduces \( 2\Delta \), which should reduce \( E_f \) or \( t \) to make \( T_0 \) rise. Similarly, if the impurities were to randomly transform links into \( \pi \) junctions\(^{43} \), their effect would be to reduce \( E_f \) or \( t \) to make \( T_0 \) rise at large doping\(^{44} \). Thus, these three cases rule out disorder induced localization models in which \( t \) is the only \( \Delta \) dependent parameter\(^3 \). They also rule out Coulomb interaction models in which \( E_c \) depends only on the geometry of the localized states and the local dielectric constants\(^8 \). Magnetic impurity doping is not expected to influence dielectric properties. It might influence the geometry by causing the islands to shrink. That effect, however, would increase charging energies and thus, \( T_0 \).

A possible explanation for \( T_0 \) decreasing with pair-breaking is that \( E_c \) depends directly on \( \Delta \). This dependence emerges when the inter island charging energy greatly exceeds the pair binding energy \( \Delta \), i.e. \( E_c \gg \Delta \). In this limit, virtual quasiparticle tunneling processes, which depend on \( \Delta \), renormalize the capacitance of single junctions\(^{46,47} \). An estimate of \( E_c \) for the dots in the a-Bi films indicates they are in this limit. For 12 equivalent dots to fit around each pore, the dot radii must be \( r_{dot} = 13nm \). Using this length scale and the dielectric constant of aluminum oxide \( \epsilon = 10 \) gives \( E_c \approx \epsilon \pi r_{dot}/k_B \approx 3000K \gg \Delta \). Beloborodov and coworkers\(^7 \) included this effect in a model of granular films to derive a renormalized charging energy:

\[
\tilde{E}_c = \frac{2\Delta}{3\pi^2 g} \ln \left( gE_{c0}/\Delta \right) \quad (5)
\]

where \( g = G/(2e^2/h) \) is the dimensionless normal state conductance between grains and \( E_{c0} \) is the self charging energy of a dot. Thus, the charging energy becomes \( \Delta \) dependent.

Using \( E_c \) in Eq.(2), yields:

\[
T_0 = \frac{2\Delta}{3\pi^2 g} \ln \left( gE_{c0}/\Delta \right) - \frac{zg\Delta}{2} F(2\pi f) \quad (6)
\]

which can be compared with the experimental results (see Fig. (3)) using parameters, \( \Delta \), \( z \), \( E_{c0} \), and \( g \) fixed by measurements. Taking the temperature at which the
reference film resistance drops to 10% of its normal state resistance as \( T_{0} \), gives \( \Delta(\alpha_{Gd} = 0) = 1.7k_{B}T_{0} \) presuming the weakly coupled BCS relation between \( \Delta \) and \( T_{0} \) as appropriate for a-Bi films near the SIT\(^{36} \). The result is insensitive to the specific choice of the 10% criterion because of the 5% width of the resistance transitions and relatively weak dependence of \( T_{0} \) on variations in \( T_{c} \). With pairbreaking, the minimum energy for excitations becomes the spectral gap \( \Omega_{G} \), rather than \( \Delta \), the pairing potential. Thus, the calculated evolution of the spectral gap \( \Omega_{G}(\alpha_{Gd}) \) with doping\(^{49} \) is used instead of \( \Delta(\alpha_{Gd}) \). \( z = 2.5 \) is the average coordination for the dot arrays since half the islands have \( z = 2 \) and half have \( z = 3 \). \( E_{c0} \) is determined presuming the Josephson Junction array model\(^{7} \) employed to get Eq. (2). The dots are treated as disks on the surface of aluminum oxide in vacuum so that \( E_{c0} = 4e^{2}/(8\epsilon_{0}r_{dot}) \) with \( \epsilon = 10 \) and \( r_{dot} = 13nm \) as estimated above. The interisland conductances, \( g \) are set by the normal state sheet resistance as \( g = \frac{3e}{2\pi\epsilon}R_{N}^{-33} \). The expression for \( T_{0} \), however, is sensitive to variations in \( g \) that are smaller than the \( \approx 10\% \) systematic uncertainties in measuring \( R_{N} \). Consequently, the \( g \)’s were set within the window of uncertainty using \( R_{N} = 18.9k \Omega \) and 17.6k\( \Omega \) to make the calculated \( T_{0} \)’s at zero doping coincide with the data, for films I and II, respectively. Finally, \( F(0) = 1 \) and the \( F(1/2) \) values were set in accord with predictions of a theory of the magneto resistance oscillations\(^{49} \). That theory indicates that \( F(1/2) \) grows from 0.9 and 1.0 with increasing distance from the SIT. Accordingly, \( F(1/2) \) was set to 0.96 and 0.905 for films I and II, respectively, to match the zero doping data points in Fig. (3).

The predictions of Eq. (6) compare well with the data (see Fig. 3). Qualitatively, the calculated and measured \( T_{0} \) decrease monotonically with doping for films with lower \( g \) that are farther from the SIT and develop a maximum at higher \( g \). Quantitatively, the predicted and measured variations in \( T_{0} \) are similar in size. The \( \alpha_{Gd} \) scales for the data and the calculation differ by about a factor of two. This difference could indicate that the spectral gap to \( T_{c0} \) ratio decreases more rapidly in nanodots than predicted for bulk materials. Altogether, the agreement implies that 1) these Cooper pair insulators are Mott insulators with screened Coulomb interactions and 2) the non-monotonic behavior of \( T_{0}^{\prime}(\alpha_{Gd}) \) reflects the different \( \Delta \) dependencies of the first term (\( \propto \Delta \log \Delta \)) and the second term (\( \propto \Delta \)) in Eq. 6.

This Mott phase is distinct. The screening effect differentiates it from the unscreened Mott transition observed in micro-fabricated Josephson Junction Arrays for which \( \Delta > E_{c} \).\(^{50} \) Similarly, it differs from cold atom system Mott transitions, which have short range interactions and bosons that cannot decompose into constituent parts\(^{51} \). It is interesting also to consider implications for bosonic SITs in high temperature superconducting cuprates\(^{52} \). The nodes in their d-wave density of states could make virtual quasiparticle screening more effective than in fully gapped s-wave systems. Smaller \( T_{0} \)’s and/or deviations from simply activated transport could arise.

Finally, the disappearance of \( T_{0} \)’s frustration dependence at higher doping levels likely signals a crossover from transport that involves Cooper pairs to quasiparticle dominated transport. The crossover is smooth: the \( R(T) \) (Figs. 2a,b) maintain an activated form and \( T_{0} \) evolves without any clear discontinuities in its value or slope. The continued decrease of \( T_{0} \) with Gd doping suggests that the quasi-particle transport depends directly on the Cooper pair binding energy. This dependence arises for quasi-particle tunneling between superconducting dots as proposed to explain negative magneto-resistance in granular Pb\(^{53} \) and Indium Oxide films\(^{34} \). Within this model, the inferred values of \( 2\Delta \) at the crossover, presuming \( T_{0} = 2\Delta \), are 0.83 K and 0.6 K for films I and II, respectively. Both of these values fall below the transition temperatures of their associated reference films, which makes them reasonable.

To summarize, we investigated the influence of magnetic impurity doping, magnetic frustration, and sheet resistance on the transport activation energy, \( T_{0} \), of the Cooper pair insulator phase in amorphous Bi films on AAO substrates. \( T_{0} \)’s response implies that it depends directly on the energy binding the Cooper pairs and agrees well with a model\(^{7} \) of the Cooper Pair Insulator as a Mott Insulator in which virtual quasi-particle tunneling processes screen the Coulomb interactions that impede boson tunneling transport. The results rule out a number of other models\(^{2,4,6,8} \) and distinguish this Cooper pair insulator phase from that in micro-fabricated Josephson Junction arrays\(^{50} \) and the Bose insulator phase in cold superconductors.

FIG. 3. Comparison with Mott Insulator Transport Model with Virtual Quasiparticle Screening. Activation energy as a function of magnetic impurity doping calculated using Eq. 6 as described in the text. The red and blue lines correspond to films I and II, respectively and the solid and dashed lines correspond to \( f = 0 \) and \( f = 1/2 \), respectively. The black dots give the measured \( T_{0} \) at zero doping. Inset: Representation of the experimental results in Fig. 2 for films I and II with lines to guide the eye.
atom systems\textsuperscript{31} in which virtual quasiparticle processes exert negligible influence.

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\textsuperscript{1} V. F. Gantmakher and V. T. Dolgopolov, Physics-Uspekhi 53, 1 (2010).
\textsuperscript{3} A. Gangopadhyay, V. Galitski, and M. Muller, Physical Review Letters 111, 026801 (2013).
\textsuperscript{10} M. Steiner and A. Kapitulnik, Physica C-Superconductivity and Its Applications 422, 16 (2005).
\textsuperscript{25} T. T. Nguyen and M. Müller, arXiv:1606.07747.
\textsuperscript{26} K. B. Efetov, Sov. Phys. JETP 51, 1015 (1980).
\textsuperscript{28} T. I. Baturina and V. M. Vinokur, Annals of Physics 331, 236 (2013).
\textsuperscript{29} M. Swanson, Y. L. Loh, M. Randeria, and N. Trivedi, Physical Review X 4, 021007 (2014).
\textsuperscript{38} M. Müller, EPL (Europhysics Letters) 102, 67008 (2013).
\textsuperscript{40} J. S. Parker, D. E. Read, A. Kumar, and P. Xiong, Europhysics Letters 75, 950 (2006).
\textsuperscript{43} L. N. Bulaevskii, V. V. Kuzii, and A. A. Sobyanin, JETP Letters 25, 290 (1977).
\textsuperscript{44} E. Granato, Physical Review B 96, 184510 (2017).