Quantum phase transition in superconducting Au$_{0.7}$In$_{0.3}$ films of very low normal-state sheet resistance

M. M. Rosario, H. Wang, Yu. Zadorozhny, and Y. Liu

Department of Physics, The Pennsylvania State University, University Park, PA 16802

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We report the observation of a quantum phase transition (QPT), tuned by a parallel magnetic field, between a superconducting and metallic state in Au$_{0.7}$In$_{0.3}$ films of very low normal-state sheet resistance (< 90 Ω). These films can be modeled as a random array of superconductor-normal metal-superconductor (SNS) junctions. Electrical transport and tunneling measurements suggest that, in the metallic state, the film consists of superconducting In-rich grains not linked by Josephson coupling. Whether phase fluctuation, which is not expected to be strong in such an SNS junction system according to the phase–number uncertainty relation, or a different physical process drives the observed QPT is discussed.

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As an example of a quantum phase transition (QPT), the superconductor-insulator transition (SIT) in two dimensions (2D) has been an important subject of study in contemporary condensed matter physics [1]. Consideration of the 2D SIT observed in granular films [2,3] and superconductor-insulator-superconductor (SIS) Josephson junction arrays [4] usually starts from quantum phase fluctuations. The SIT observed in homogenous films [5,6,7] has been analyzed in a dirty Bose-Hubbard model [8] that builds on phase considerations as well. The physical origin of the phase fluctuation is captured by an uncertainty relation between the superconducting phase $\phi$ and the number of carriers $N$ with the form $\Delta \phi \Delta N \approx 1$. For SIS junction arrays or granular films, the large charging energy of the superconducting islands makes the transfer of Cooper pairs between neighboring islands difficult, suppressing the fluctuation in $N$. As a result, the fluctuation in the phase is enhanced, leading ultimately to an SIT. The introduction of shunt resistance or dissipation tends to suppress the phase fluctuation. In recent experiments, it was found that dissipation can restore the global phase coherence for a system sitting on the insulating side of the SIT [9,10,11], as anticipated [11]. For amorphous films, the localization of electrons by disorder can reduce $\Delta N$, and consequently lead to enhanced phase fluctuation and an SIT. With no exception, the normal-state sheet resistance $R_{\text{N}}$ of the films around the SIT is large, typically around $h/4e^2 = 6.45 \text{ k}\Omega$ [1].

Recently, the possible existence of a metallic state in 2D SIT systems has received renewed theoretical attention [12,13]. The apparent existence of such a state occurring between the insulating and the superconducting phase, marked by a flat resistance tail at the lowest temperatures, was seen in ultrathin granular films of Ga, Pb, and In measured down to 0.6 K [2], in thin Al films down to 0.3 K [14], and in SIS Josephson junction arrays down to 10 mK [1]. Based on experiments on amorphous MoGe films in perpendicular magnetic field [15], it was proposed that dissipation may open a parameter space for a metallic state to occur near a 2D QPT in general [13]. In all these previous studies, the metallic state was found in systems with large $R_{\text{N}}$ (> 1 kΩ). Here we report electrical transport and tunneling measurements in Au$_{0.7}$In$_{0.3}$ thin films, showing the existence of a QPT and a metallic state in films with very low $R_{\text{N}}$ (< 90 Ω).

Au$_{0.7}$In$_{0.3}$ films were prepared by sequential thermal evaporation of alternating Au and In layers, with the layer thicknesses determined by the desired atomic ratio of Au to In. The maximum solid solubility of In in Au is around 10% at room temperature. The interdiffusion of Au and In in a Au$_{0.7}$In$_{0.3}$ film results in a 2D system consisting of In-rich grains, with a maximum local temperature of 0.6–0.8K, embedded in a Au$_{0.9}$In$_{0.1}$ matrix, with $T_c$ around 77 mK [16]. Since the atomic composition and the size of the In-rich grains can vary randomly, strong spatial variation in the amplitude of the superconducting order parameter (the superconducting gap) is expected. It has been shown previously that this system can be modeled as a random array of superconductor-normal metal-superconductor (SNS) Josephson junctions [16].

Planar tunnel junctions of Au$_{0.7}$In$_{0.3}$/MgO$_x$/Mg were made in a standard cross geometry, with a junction size of 0.2×0.3 mm$^2$. The Mg bottom layer was thermally evaporated at ambient temperature, with subsequent growth of a native Mg oxide layer encouraged by the use of glow discharge in an O$_2$ environment. The deposition of the Au$_{0.7}$In$_{0.3}$ top layer was carried out with the substrate held at liquid nitrogen temperatures ($\approx$ 77 K) to help preserve the insulating barrier. The results presented here correspond to a junction with a normal state resistance of 115 Ω. Measurements on another junction of comparable resistance and two junctions of higher resistance ($\sim 10^4\Omega$) yielded qualitatively similar results.

Electrical transport measurements were carried out in a dilution refrigerator equipped with a superconducting magnet. The base temperature was < 20 mK. All electrical leads entering the cryostat were filtered with the
attenuation of 10 dB at 10 MHz and 50 dB at 300 MHz. Resistances and current-voltage \((I - V)\) characteristics were measured with a d.c. current source and a nanovoltmeter. Tunneling conductances \(G_j(V)\) were determined by taking the derivative of \(I - V\) curves numerically. The magnetic field was applied parallel to the film plane (estimated to be aligned within about 1°) and perpendicular to the tunneling direction.

Figure 1 shows the temperature dependence of the sheet resistance \(R_{\square}(T)\) in parallel magnetic field \(H_{||}\) for several Au_{0.7}In_{0.3} films. The normal-state sheet resistance of these films, ranging from 10–90 Ω, are very low compared with \(h/e^2\) where the 2D SIT typically occurs. The superconducting transition temperature \(T_c\) was found to decrease with increasing \(H_{||}\). For relatively small \(H_{||}\), a fully superconducting state was obtained. However, despite the presence of a substantial resistance drop slightly below the zero-field \(T_c\), zero resistance was not reached down to \(T = 20\) mK as \(H_{||}\) surpassed a critical value \(H_{c2}^\parallel\). Instead, a flat resistance tail was found, spanning nearly a decade in temperature. The limiting \((T \to 0)\) resistance increased exponentially with \(H_{||}\) (Fig. 2).

Figure 3 shows \(I - V\) characteristics for the 10 nm thick film (shown in Fig. 1b). Data presented in Fig. 1b were obtained at currents of 1µA or 100nA, with no qualitative differences in \(R(T)\) behavior. The \(I - V\) characteristic evolved from nonlinear to linear (ohmic) behavior with increasing \(H_{||}\) at the lowest temperatures (Fig. 3b). According to the Kosterlitz-Thouless (KT) theory \([13]\), the finite temperature superconducting transition in 2D is associated with the thermal unbinding of vortex-antivortex pairs, leading to a \(I - V\) characteristic of \(V \sim I^3\) at \(T = T_{KT}\). In these Au_{0.7}In_{0.3} films, the exponent was found to be less than 3 down to the lowest temperature \((V \sim I^{2.5})\), perhaps due to a vanishing \(T_{KT}\) < 25 mK. Despite this, the non-linear \(I - V\) characteristic at the lowest temperatures indicates that vortices and antivortices were present at \(H_{||} = 0\). Linear \(I - V\) characteristics were found even at 25 mK at \(H_{||} = 0.20\)T, above which the resistance tail emerged, suggesting the absence of vortex-antivortex unbinding in the metallic state. However, whether vortices and antivortices were absent, or were present but fully unbound, was not resolved.

The interesting question is whether In-rich grains remain superconducting in the metallic state. Tunneling measurements were carried out to address this. Single particle tunneling spectra, obtained in Au_{0.7}In_{0.3}/MgO_{2}/Mg junctions at various temperatures and applied parallel fields, are shown in Fig. 4. The tunneling spectra in zero field indicates that an energy gap opened below \(T = 0.28\) K. While a coherence peak is present in the tunneling spectra, the peak is smaller and the zero bias conductance \(G_j(V = 0)\) is larger than expected from BCS theory. A 35% suppression of the normal state DOS was observed at 20 mK for junction shown in Fig. 4a and 25–80% for others at the zero bias, resulting from either a substantial population of quasiparticles in Au_{0.7}In_{0.3} or junction leakage. The superconducting gap \(\Delta_0\) is estimated by the peak position to be 0.1 m€ at 25 mK, and is smaller if one uses \(G_j(V = \Delta/e) = G_j^{\parallel}\). With \(T_c = 0.82\) K, this leads to \(\Delta_0/k_B T_c = 1.41\), slightly smaller than the BCS result, \(\Delta_0/k_B T_c = 1.76\).

The evolution of the tunneling spectra with \(H_{||}\) is shown in Fig. 4b. With increasing field, more states were found within the gap until it closed at \(H_{||} \approx 0.55\) T. With increasing \(H_{||}\), the coherence peak decreased in height and broadened in width. The zero-bias conductance, which could be related to the total area of the normal region of the films, increased linearly with \(H_{||}\) (data not shown).

A natural picture concerning the observed superconducting-metallic state transition in Au_{0.7}In_{0.3} films emerges from these measurements. With the application of \(H_{||}\), the superconductivity in the In-rich grains is gradually reduced. Eventually, a sufficient number of grains become normal so that a percolating path of Josephson coupled superconducting grains can no longer form, leading to the disappearance of global superconductivity. The average separation between superconducting islands at \(H_{||}^c\) can be estimated. Finite Josephson coupling of an SNS junction is expected if the length of the N-layer is shorter than a few times of the normal coherence length \(\xi_N\). In the dirty limit, \(\xi_N = (hD/2\pi k_B T)^{1/2}\) where \(D = v_F \tau\) is the diffusion constant, \(v_F\) is the Fermi velocity, and \(\tau\) is the relaxation time in the Boltzmann formula for resistivity, \(\rho = m/ne^2\tau\). For the film shown in Fig. 1b, for example, we estimate \(\xi_N\) for the normal metal matrix (Au_{0.9}In_{0.1}) to be \(\approx 0.2\) µm at 20 mK. This suggests that the average separation between surviving superconducting islands is in the micron range, comparable to the average size of the largest In-rich grains.

Recently, Larkin and co-workers have proposed a theory for an SNS array of superconducting islands (of radius \(d\) and spaced \(b\) apart, such that \(b \gg d\)) proximity coupled to one another via a 2D normal film of dimensionless conductance \(g = \sigma/\left(\epsilon^2/\hbar\right)\). In their model, quantum phase fluctuations induced by disorder and Coulomb repulsion are responsible for the suppression of superconductivity. A QPT from a superconducting to a normal-metal state was shown to occur at \(g < g_c \approx (1/\pi)\ln(b/d)^2\). The corresponding critical sheet resistance \(R_{\square}^c\) can be substantially smaller than \(R_Q\). For the films shown in Fig. 1, we estimate \(R_{\square}^c \approx R_{\square}^{N}\). Then \(R_{\square}^{N} = 9.96\Omega\) would yield \(g_c = 404\). An unreasonably large distance between grains, given by \(b = d exp(63)\), would be obtained at the QPT, larger than the value inferred from the experiment, as described above. This appears to suggest that some important ingredients are missing from this model in its present form.

An alternative model has been proposed in which the
effects of fluctuations in the amplitude of the superconducting order parameter, primarily as a function of time, are taken into account \[ \xi_0. \] Presumably, the amplitude fluctuation will be present when the radius of the superconducting island \( d \) is smaller than the zero-temperature superconducting coherence length \( \xi_0 \), such that \( d < \xi_0 \). In the dirty limit, \( \xi_0 = (\hbar D/\Delta)^{1/2} \). This yields \( \xi_0 \approx 0.1 \mu m \) for the present study. The size of many superconducting grains in \( Au_{0.7}In_{0.3} \) films is expected to be smaller than this \[ 10 \] making substantial amplitude fluctuation plausible. The critical concentration of grains obtained in this model is substantially larger than that obtained in Ref. \[ 13 \], leading to a more reasonable value of critical conductance, qualitatively consistent with our experimental observation. Unfortunately, quantitative predictions are lacking, making a quantitative comparison between the experiment and the theory impossible.

It was previously emphasized that amplitude fluctuations played a role in the 2D SIT in ultrathin amorphous films \[ 7, 20 \]. The vanishing of the gap \[ 7 \], the broadening of the tunneling spectrum \[ 20 \] near the SIT were cited as the evidence for amplitude fluctuations. However, while the vanishing of the gap and condensation energy \[ 20 \], as well as the predictions are lacking, making a quantitative comparison between the experiment and the theory impossible.

The simplest consideration suggests that a deparing process is needed to induce amplitude fluctuation. The residual repulsive electron-electron interaction appears to be an obvious driving force for amplitude fluctuation. Theoretically, a magnetic field applied parallel to a homogeneously disordered 2D superconducting system was shown specifically to lead to strong amplitude fluctuations due to Zeeman splitting and strong spin-orbit coupling \[ 21 \]. In this context, physical phenomena which may result from the presence of negative superfluid density (i.e., \( \pi \) junctions with negative Josephson coupling), an extreme case of the amplitude fluctuation, have been observed in \( Au_{0.7}In_{0.3} \) cylindrical films \[ 22 \].

The observation of a flat resistance tail in films with such low \( R_{\parallel} \) is striking. Is it possible that the tail originates from electrons being at a temperature higher than the lattice temperature because of insufficient cooling? It has been emphasized that a flat resistance tail has never been observed in granular or amorphous films prepared in some laboratories \[ 6, 21, 22 \]. In the present work, noise from outside the system were eliminated by RF filters. However, microwave noise originating from room-temperature parts of the measuring leads inside the cryostat may still affect the film resistance. Effective elimination of these noises requires filtering at cryogenic temperatures, for which the current system is not equipped. On the other hand, a similar phenomenon in Josephson junction arrays was observed in studies carried out with such filters \[ 4 \]. In addition, the films in the present study were of very low \( R_{\perp} \), which should make heating due to the electromagnetic environment less significant.

Nevertheless, it is desirable to show directly that electrons still cool in the temperature range in which the flat resistance tail was seen. Figure 5a shows the tunneling spectra at 20 and 50mK in zero field, indicating that the electronic system still cooled down to the lowest temperatures. Above \( H_{\parallel} \), the spectra became less distinguishable. However, this could be due to deparing effects. For a cylindrical film above \( H_{\parallel} \), the resistance shows a negative \( dR/dT \) (Fig. 5c), indicating that the electrons were cooling in the metallic state for this sample. However, \( R(T) \) at 5 T for the 7.5nm thick film showed a change of slope or flattening-off at low temperatures (Fig. 5d). Whether this was because superconducting fluctuations were still present or electrons were not cooling was not resolved. Direct measurements of the electronic temperature are needed to clarify this issue.

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FIG. 1: Sheet resistance as a function of temperature, $R(\theta)(T)$, of superconducting Au$_{0.7}$In$_{0.3}$ films of several thicknesses, $t$, as indicated. Planar films with (a) $R^N = 89.2\Omega$, (b) $R^N = 55.7\Omega$, (c) $R^N = 9.90\Omega$, and (d) a cylindrical film with diameter $d = 550$ nm and $R^N = 9.96\Omega$ are shown.

FIG. 2: Semi-log plot of the limiting ($T \to 0$) resistance normalized to the zero field normal state resistance, $R/R^N$, as a function of applied parallel magnetic field normalized to the field required to suppress the resistance drop in $R(T)$, $H_\parallel/H_\parallel^N$, for the films shown in Fig. 1. The film given in Fig. 1c, not included in the plot as data above 0.2T is not available, showed similar behavior.

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FIG. 3: (a) $I-V$ characteristic of the 10 nm thick Au$_{0.7}$In$_{0.3}$ film at $H_\parallel = 0$. Curves are given for $T = 20$ mK, 90 mK, 0.1 K, 0.125 K, 0.175 K, and 0.20 K. The dashed line indicates linear (ohmic) behavior. The low current region of the 20 mK curve follows $V \sim I^\alpha$ where $\alpha \approx 2.5$. (b) $I-V$ characteristic at finite $H_\parallel$. Curves were taken at the fields indicated and at $T = 25$ mK, with the exception of the 0.1 T curve which was taken at 70 mK.
FIG. 4: Tunneling spectra of the 10 nm thick Au$_{0.7}$In$_{0.3}$ film. (a) Tunneling conductance as a function of voltage, $G_I(V)$, at several temperatures at $H_{||} = 0$. $T > 25$ mK curves are shifted up from the 25 mK curve for clarity. (b) $G_I(V)$ curves at finite $H_{||}$ and $T = 25$mK. $H_{||} > 0$ curves are shifted up from the $H_{||} = 0$ curve. The high energy features [e.g., at 0.2 mV for 0.28 K in (a)] appears to be related to a junction-dependent current redistribution process and is not intrinsic to Au$_{0.7}$In$_{0.3}$.

FIG. 5: Tunneling spectra of the 10 nm thick Au$_{0.7}$In$_{0.3}$ film for $T = 25$ and 50 mK at (a) $H_{||} = 0$ and (b) 0.23T. (c) $R_{G}(T)$ of a cylindrical film with $t = 30$nm and $d = 470$nm in $H_{||} = 0.26$T and 0.6T. (d) A close-up of the $R(T)$ of the 7.5nm film shown in Fig. 1a.
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