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Superconductivity on the localization threshold and magnetic-field-tuned superconductor-insulator transition in TiN films

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Temperature- and magnetic-field dependent measurements of the resistance of ultrathin superconducting TiN films are presented. The analysis of the temperature dependence of the zero field resistance indicates an underlying insulating behavior, when the contribution of Aslamasov-Larkin fluctuations is taken into account. This demonstrates the possibility of coexistence of the superconducting and insulating phases and of a direct transition from the one to the other. The scaling behavior of magnetic field data is in accordance with a superconductor-insulator transition (SIT) driven by quantum phase fluctuations in two-dimensional superconductor. The temperature dependence of the isomagnetic resistance data on the high-field side of the SIT has been analyzed and the presence of an insulating phase is confirmed. A transition from the insulating to a metallic phase is found at high magnetic fields, where the zero-temperature asymptotic value of the resistance being equal to h/e^2.

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The interplay between superconductivity and localization is a phenomenon of fundamental interest and the question of the nature of superconductivity and its evolution in two-dimensional disordered systems and a perpendicular magnetic field continues to receive a great deal of theoretical and experimental attention. Two-dimensional systems are of special interest as two is the lower critical dimensions for both localization and superconductivity. Two ground states are expected to exist for bosons at $T = 0$: a superconductor with long-range phase coherence and an insulator in which the quantum mechanical correlated phase is disjointed. The zero-temperature superconductor-insulator transition (SIT) is driven purely by quantum fluctuations and is an example of a quantum phase transition [1]. The superconducting phase is considered to be a condensate of Cooper pairs with localized vortices, and the insulating phase is a condensate of vortices with localized Cooper pairs. Between these two states there is the only metallic phase point, and this metal has a bosonic nature as well. The theoretical description based on this assumption was suggested in Ref. [2]. At finite temperatures, a quantum phase transition is influenced by the thermal fluctuations, and according to the theory: (i) the film resistance R near the magnetic-field-induced SIT at low temperature T in the vicinity of the critical field B_c is a function of one scaling variable $\delta = (B-B_c)/T^{1/\nu z}$, with the critical exponents ν and z being constants of order of unity; (ii) at the transition point the film resistance is of the order $h/(2e)^2 \approx 6.5$ kΩ (the quantum resistance for Cooper pairs). Although much work has been done, and in many systems the scaling relations hold [3 – 8], the magnetic-field-induced SIT in disordered films remains a controversial subject, especially concerning the insulating phase and the bosonic conduction at $B > B_c$. There is experimental evidence [7] that despite the magnetoresistance being non-monotonic, and in the magnetic fields above the critical one the derivative of resistance dR/dT is negative, the phase can be insulating as well as metallic. The behavior of the resistance in this region discussed in [5, 6] in terms of the magnetic-field-induced SIT (which is essentially *bosonic* in nature) can actually be explained on basis of a *fermionic* approach, namely, in the frames of the theory of the quantum corrections to the conductivity in disordered metals. The possibility of such interpretation is shown in the work [9] basing on the recent calculation of the quantum corrections due to superconducting fluctuations [10]. As a usual thermodynamic superconductor-normal metal transition, provided that the behavior of this metal is controlled, to a considerable degree, by the quantum corrections and a superconductor-insulator transition may have very sim-

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ilar experimental manifestations, some clear criteria are needed to enable one to tell which of the two underlies the behavior observed experimentally. Supposing the SIT to be the cause (e.g. the temperature dependence is found to be of an activated type) the question is then what effect the magnetic field may have on the bosonic insulator.

In this paper we present the results of measurements and detailed analysis of temperature- and magnetic field dependence of the resistance of TiN films, devoting attention to a careful examination of the presence of the insulating phase and its alteration on the high-field side of the SIT.

A TiN film with a thickness of 5 nm was formed on 100 nm of SiO_2 grown on top of <100> Si substrate by atomic layer chemical vapour deposition at 350°C [11]. Structural analysis shows that the formed TiN films are polycrystalline. The films exhibit low surface roughness and consists of a dense packing of the crystallities, with a rather narrow distribution of size and the average size is roughly $\sim$ 30 nm. The samples for the transport measurements were fabricated into Hall bridges using conventional UV lithography and subsequent plasma etching. Four terminal transport measurements were performed using standard low frequency techniques. The resistance data were taken at a measurement frequency of 10 Hz with an ac current $0.04 \div 1$ nA. The magnetic field was applied perpendicular to the film.

Four samples with the same thickness of 5 nm but different degrees of disorder were studied in the present work. We begin by showing the temperature dependence of the resistance $R(T,0)$ at zero magnetic field. $R(T,0)$ data are presented in Fig. 1 for sample labelled #1 and in Fig. 2 for all samples studied in this work. The resistance is non-monotonic function of the temperature as is seen more clearly in Fig. 2a. With decrease of T, the increase of the resistance, which is observed from $T = 300$ K, is followed by a drop to the superconducting state. The transitions are significantly broadened. To explore reasons of such behavior and to determine the main sample parameters we apply approach similar to the one used previously in Ref. [12]. As films under study are high resistive it should be expected that the dependence $R(T)$ is strongly affected by the contribution of superconducting fluctuations (the Aslamazov-Larkin correction [13]) even at the temperatures far from the transition temperature T_c:

$$\Delta G_{AL} = \frac{e^2}{16\hbar}\left[\ln\left(\frac{T}{T_c}\right)\right]^{-1}. \qquad (1)$$

After extraction this correction, with T_c being the only free parameter, we obtain the temperature dependence

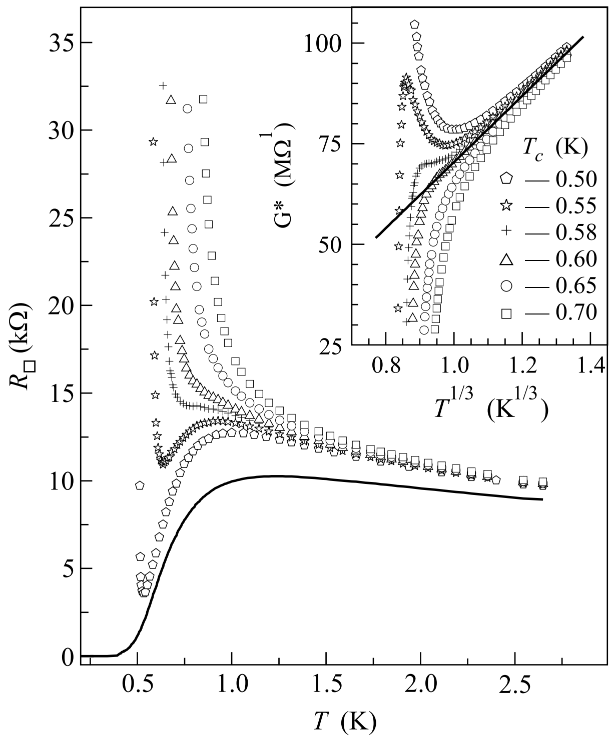

Fig.1. Temperature dependence of the resistance per square for sample #1 (solid line). Curves, depicted by symbols, correspond to $R^*(T_c)$ (see text) and are obtained after subtraction of the Aslamasov-Larkin correction (1) with different T_c, which are listed in the inset. These curves are presented as $G^* = 1/R^*$ vs. $T^{1/3}$ in the inset

$R^*(T_c) = (1/R_\square - \Delta G_{AL}(T_c))^{-1}$, which are depicted by symbols in Fig.1. The curves $R^*(T_c)$ obtained with $T_c < 0.6$ K are non-monotonic, whereas the ones corresponding to $T_c > 0.6$ K give too strong growth of the resistance. The choice of $T_c = 0.6$ K is confirmed by the further analysis of R^*, which is carried out for a start in terms of 3D "bad" metal in the vicinity of the metal-insulator transition (MIT) [14]. In the critical region of the MIT the behavior of the system is governed by electron-electron interaction and the temperature dependence of the conductivity is controled by the only temperature dependent scale $L_T = \sqrt{\hbar D/k_B T}$

$$\sigma = \frac{e^2}{\hbar}\frac{1}{L_T} \qquad (2)$$

Using Einstein's relation $\sigma = e^2 D(\partial N/\partial\mu)$, the conductivity can be rewritten as

$$\sigma = \frac{e^2}{\hbar}\left(T\frac{\partial N}{\partial \mu}\right)^{1/3}. \qquad (3)$$

The representation of (3) in the form of

$$G_{AA}(T) = a + bT^{1/3} \qquad (4)$$

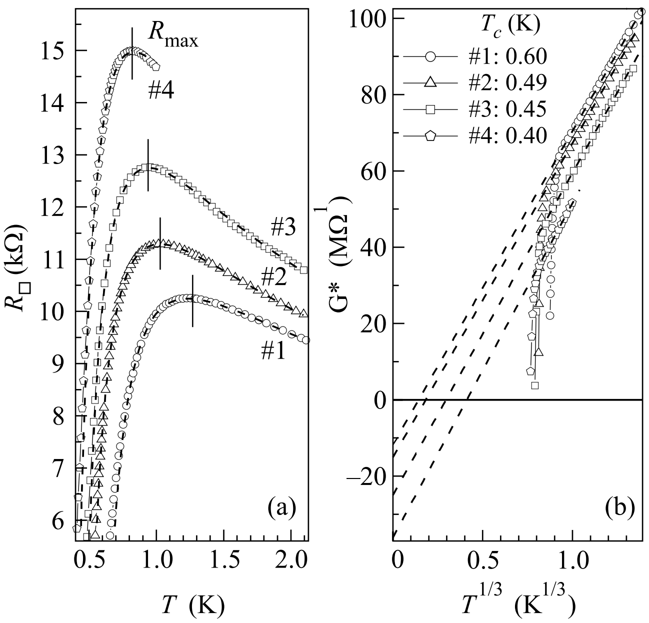

Fig.2. (a) Temperature dependence of the resistance per square for four samples (symbols) and corresponding calculated curves R_{AA+AL} (dashed lines) at optimally fitted values of a and b (4), and T_c. (b) $G^* = 1/R^*$ vs $T^{1/3}$ for the same samples at optimally fitted values of the critical temperature T_c listed in the figure

is usually used to discriminate between a metal and an insulator by means of determination of the sign of the parameter a at extrapolation to $T = 0$ (see as an example [15]). A positive value of a indicates the metallic state, whereas a negative value of a points to activated conductance at lower temperatures.

The inset of Fig.1 shows the conductance $G^* = 1/R^*$ versus $T^{1/3}$. The dependence $G^*(T^{1/3})$ at $T_c = 0.6$ K is monotonic and the temperature, to which the linear character of G^* is endured, is the lowest one. Determining in this way the values of a and b in (4) and T_c, we can fully describe the temperature dependence of the resistance film studied ($R_{AA+AL} = [G_{AA}(a,b) + \Delta G_{AL}(T_c)]^{-1}$). The outcome of the above procedure applied to all samples under study is illustrated in Fig.2. At $T > T_c$, R_{AA+AL} closely follows the experimental data, justifying the validity of the above procedure and indicating that, with the deduction of the direct contribution of the superconducting fluctuations, the conductance of the film is reasonably described by Eq. (4). It is interesting to note that the slope of the dependences $G^*(T^{1/3})$ plotted in Fig.2b (or the parameter b in Eq. (4)) is approximately the same for all samples studied, whereas the values $a \equiv G^*(0)$ differ significantly.

Additional information, which can been extracted from the analysis, is the estimation of the parameters like the compressibility

$$\frac{\partial N}{\partial \mu} = \frac{1}{k_B}\left(\frac{b\hbar}{e^2 d}\right)^3, \qquad (5)$$

the diffusion coefficient

$$D = \frac{k_B}{\hbar}\left(\frac{e^2 d}{b\hbar}\right)^2 T^{1/3}, \qquad (6)$$

and $k_F l = 3Dm/\hbar$. Here the value of b is determined from the linear approximation of G^* vs $T^{1/3}$ (Eq. 4), d – the thickness of the film. The following numbers are obtained: $\partial N/\partial\mu = 3.6\cdot 10^{21}$ eV^{-1}cm^{-3}, $D = 0.29$ cm^2/s $(T/\mathrm{K})^{1/3}$ and $k_F l = 0.74(T/\mathrm{K})^{1/3}$, that is $k_F l < 1$ at $T < 2.4$ K.

It should be noted that in order to take into account the superconducting fluctuations we have used the Eq. (1), which is valid for two dimensional case. The condition under which a film may be considered two dimensional with respect to superconducting fluctuations is

$$\ln\left(\frac{T}{T_c}\right) \ll \frac{\pi\hbar D}{8k_B T_c d^2}. \qquad (7)$$

Physically, this inequality denotes that the time of diffusive motion across the film is less than the fluctuation Cooper pair life time (or Ginzburg-Landau time)

$$\tau_{GL}^{-1} = \frac{8k_B T}{\pi\hbar D}\ln\left(\frac{T}{T_c}\right). \qquad (8)$$

The condition (7) can be rewritten as $d^2 \ll D\tau_{GL} = l_{GL}^2$. Using the estimation of diffusion coefficient we find that l_{GL} is larger than the film thickness at all temperatures under study.

One more relevant information can been received from the above analysis. Let us add Eqs. (3) and (1) and rewrite total conductance as follows:

$$G = G_{AA} + \Delta G_{AL} = \frac{e^2}{\hbar}\frac{d}{L_T} + \frac{e^2}{2\pi\hbar}\frac{l_{GL}^2}{L_T^2}. \qquad (9)$$

Then the temperature corresponding to minimal value of G is determined by the condition

$$L_T = l_{GL}\left(\frac{12}{\pi^2}\frac{l_{GL}}{d}\right)^{1/3}, \qquad (10)$$

and, accordingly, the maximum of $R(T)$ (R_{max} in Fig.2a) results from the condition $L_T \approx l_{GL}$. Thus, the non-monotonic temperature dependence of the resistance is the consequence of a competition of two length scales: l_{GL} being responsible for Cooper pairing and L_T defining the electron-electron interaction. It should be stressed, that the above estimations are rough, since the application of Eq.(3) implies that $a = 0$ in (4). There is not like this in our case and moreover for the films under study we get $a < 0$. It says that the underlying state is insulating.

We now turn to the evolution of the resistance with temperature for various magnetic fields. Fig.3a shows the isomagnetic temperature dependences of the resistance of sample #4. Not too high magnetic field ($B <$ < 2 T) destroys the long-phase coherence and reveals the underlying insulating state. The high-field data appear to be more metallic in character: the magnetic field results in a significant suppression of the insulating phase above 2 T. This is seen more clearly on a typical set of $R_\square(B)$ traces measured on sample #1 (see Fig.3b). The main feature of this graph is the presence of an intersection point at B_c, R_c. Using the B_c, we plot the same data against the scaling variable $|B - B_c|/T^{1/\nu z}$, and adjust the product of the critical exponents νz to obtain the best scaling of the data (inset of Fig.3b). However such behavior, previously regarded as the main evidence of the existence of SIT, is actually not incontestable proof of the presence of the insulating phase at $B > B_c$ [9]. In order to ascertain the type of phase we have analyzed the temperature dependence of the high-field conductance at different magnetic fields. In this regime $G(T)$ is well described by Eq. (4) as indicated in Fig.4a. As $G(0)$ is negative in fields higher than the critical field we can conclude that this phase is insulating. With further increase of B, the sign of $G(0)$ changes, which points towards the transition into the metallic state. The extrapolation to $T = 0$ allows us not only to determine the field of the insulator-metal transition (B_{IM}), but also the magnetic field dependence of the zero temperature

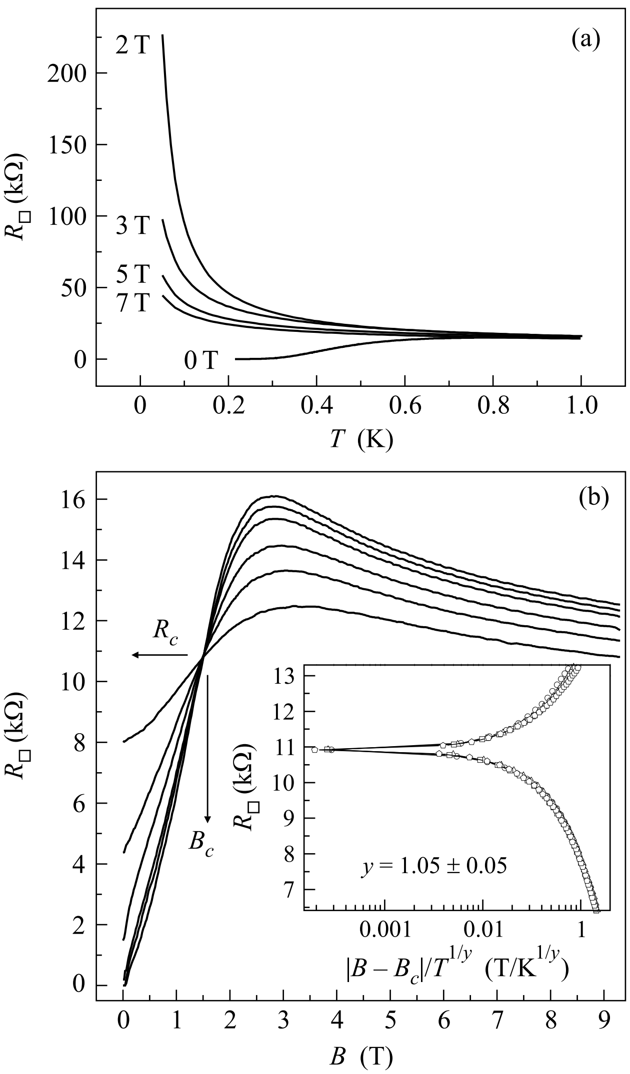

Fig.3. (a) Dependences $R(T)$ at different B of the sample #4. (b) Low-temperature isotherms of the sample #1 in the (B, R) plane. Different curves represent different temperatures: 0.35, 0.38, 0.42, 0.51, 0.61, and 0.76 K. The point of intersection: $B_c = 1.52$ T is the critical magnetic field, and $R_c = 10.9$ kΩ is the critical resistance. The inset shows a scaled plot of the same data with $y = \nu z = 1.05 \pm 0.05$

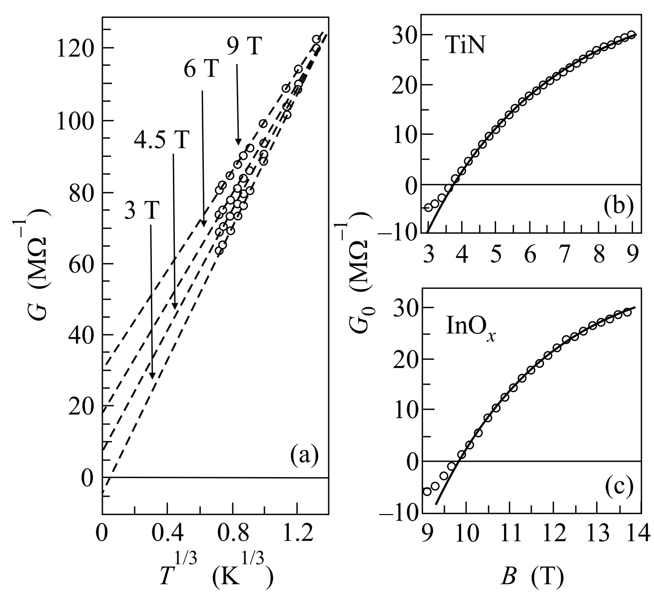

Fig.4. (a) Conductance $G = 1/R$ vs $T^{1/3}$ at different magnetic fields on the high-field side of the SIT for the sample #1. The magnetic field dependence of the zero temperature conductance determined from extrapolations in accordance with Eq. (4) are shown by symbols along with the dependences calculated from an empirical expression 11: (b) sample #1 (this work) and (c) InO_x [8]

conductance. The result of this procedure is presented in Fig.4b. Analysis of the zero temperature conductance at $B > B_{IM}$ reveals that it is well described by the empirical expression

$$G(T=0,B) = \frac{e^2}{h}\left(1 - \exp\left[\frac{B_{IM}-B}{B^*}\right]\right) \qquad (11)$$

shown by the solid line in Fig.4b. The magnetic-field-induced insulator-metal transition on the high field side of SIT was earlier observed in works of Gantmakher *et al.* on InO_x [7, 8]. Applying the same procedure to their data (see left panel of Fig.1 of Ref. [8]), we find that the dependence $G(T=0,B)$ is also well described by Eq. (11) (see Fig.4c). The exponential dependence of $G(T=0,B)$ may result from a broad dispersion of the binding energies of localized Cooper pairs. The most important result is the zero-temperature asymptotic value of the resistance, which is equal to the quantum resistance $R_Q = h/e^2$. The saturation of the low-temperature magnetoresistance to the quantum resistance was demonstrated on the beryllium films [16]. Although authors of the Ref. [16] consider that their films to be deeply within the insulating phase, it is not improbable that they are superconducting at lower temperature, i.e., that the correlated insulating phase consists of localized Cooper pairs. We believe that observed behavior, namely, the gradual approach of the low-temperature magnetoresistance towards the quantum resistance, is a very general feature of a bosonic insulator on the high-field side of the magnetic-field-tuned SIT.

In conclusion, we have studied the temperature- and magnetic-field dependence of the resistance of TiN films. We have demonstrated that the non-monotonic temperature dependence of the resistance at zero magnetic field results from the concurrence of superconducting correlations and localization, with presumably underlying insulating state having the bosonic nature. The destruction of the long-range phase coherence by a magnetic field highlights this insulating state. The further evolution of the system towards very high magnetic fields is in accordance with break-up of such localized Cooper pairs and drives the system eventually into a metallic regime. The saturation of the low-temperature magnetoresistance near the quantum resistance seems to be very general feature, occuring in several different materials. The nature of the insulating state and high field metallic state requires further investigation.

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