Quantum Metallicity on the High-Field Side of the Superconductor-Insulator Transition

T. I. Baturina[1,2] and C. Strunk[2]
[1]Institute of Semiconductor Physics, 630090, Novosibirsk, Russia
[2]Institut für experimentelle und angewandte Physik, Universität Regensburg, Regensburg, Germany

M. R. Baklanov and A. Satta
IMEC, Kapeldreef 75, B-3001 Leuven, Belgium

(Received 18 May 2006; published 21 March 2007)

We investigate ultrathin superconducting TiN films, which are very close to the localization threshold. Perpendicular magnetic field drives the films from the superconducting to an insulating state, with very high resistance. Further increase of the magnetic field leads to an exponential decay of the resistance towards a finite value. In the limit of low temperatures, the saturation value can be very accurately extrapolated to the universal quantum resistance h/e^2. Our analysis suggests that at high magnetic fields a new ground state, distinct from the normal metallic state occurring above the superconducting transition temperature, is formed. A comparison with other studies on different materials indicates that the quantum metallic phase following the magnetic-field-induced insulating phase is a generic property of systems close to the disorder-driven superconductor-insulator transition.

DOI: 10.1103/PhysRevLett.98.127003 PACS numbers: 74.78.−w, 72.15.Rn, 73.50.−h, 74.40.+k

The investigation of disordered superconducting films is of fundamental importance to understand the impact of electron-electron interaction and disorder on the ground state of many-body systems [1]. Metal and insulator are two basic ground states of the electrons in solids. The Cooper pairing, a dramatic manifestation of the attractive part of the electron-electron interaction, results in an instability of the Fermi sea and the formation of a new ground state. This superconducting state is characterized by long-range phase coherence and the possibility of nondissipative charge transport. On the other hand, disorder acts in the opposite direction, as it favors the repulsive part of the electron-electron interaction and the localization of the electron wave function. The competition between localization and superconductivity can result in an insulating ground state—the so-called Bose insulator, which is formed by localized Cooper pairs [2–4].

At zero temperature the transition between these two phases, the superconductor-insulator transition (SIT), is driven purely by quantum fluctuations and is one of the prime examples of a quantum phase transition [5,6]. Experimentally, the SIT can be induced by decreasing the film thickness [7,8] and close to the critical thickness also by magnetic field [9]. These possibilities are commonly distinguished as disorder-driven SIT and magnetic-field driven SIT. In the latter case, the magnetic field is supposed to suppress first the macroscopic phase coherence, while the Cooper pairing may survive locally. At sufficiently low temperatures this results in a sharp increase of the resistance up to several orders of magnitude [10]—the Bose insulator. Upon further increase of the magnetic field, the localized Cooper pairs are gradually destroyed, leading to a strongly negative magnetoresistance [11]. So far, direct experimental evidence for the existence of localized Cooper pairs is still scarce.

In agreement with the scenario above, early investigations [11] of amorphous InO_x films revealed a strongly nonmonotonic magnetoresistance. The resistance value in the high-field limit roughly approached the normal state resistance R_N, as expected, if the normal metal phase reappears. However, a closer analysis of the data on amorphous InO_x [11–13] and on our polycrystalline TiN in the limit $T = 0$ and large B reveals that the resistance of the films does not return to R_N [14]. Hence, the nature of the Bose insulator and its behavior in a strong magnetic field remain an open issue.

In this Letter, we show that the magnetoresistance of ultrathin TiN films decays exponentially at high magnetic fields and then saturates at a value considerably *higher* than the normal state resistance. The saturation resistance can be extrapolated with high accuracy toward $T = 0$ and turns out to be h/e^2, independent of the degree of disorder. The application of our analysis to existing data on InO_x films [12] reveals the same behavior. This demonstrates the universal character of the theoretically so far unexplained quantum metallic phase in disordered superconducting films exposed to high magnetic fields.

The TiN films with a thickness of $\lesssim 5$ nm were formed by atomic layer chemical vapor deposition onto a $\mathrm{Si/SiO_2}$ substrate. Structural analysis shows that the films consist of a dense packing of crystallites, with a rather narrow distribution of sizes around ~30 nm. The samples for the transport measurements were patterned into Hall bridges using conventional UV lithography and subsequent plasma etching. The film resistance was measured in perpendicular magnetic field using a standard four-probe lock-in tech-

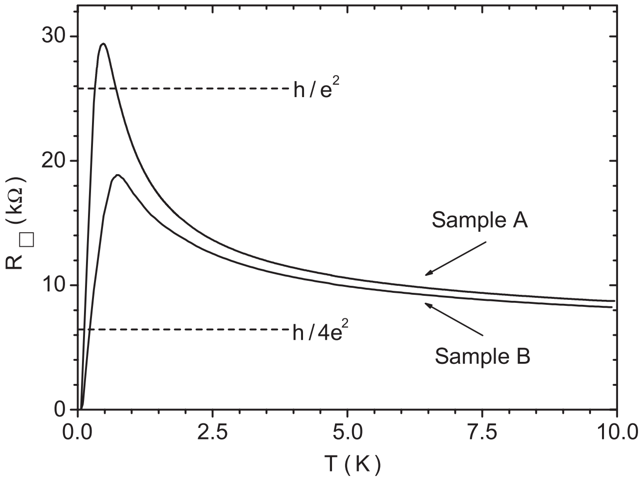

FIG. 1. Temperature dependence of $R_\Box$. The two TiN films differ slightly in their normal state resistance $R_\Box(10\ \mathrm{K}) = 8.24\ \mathrm{k}\Omega$ (sample A) and $8.74\ \mathrm{k}\Omega$ (sample B), respectively. The resistance reaches a maximum at $R_\Box(T_\mathrm{m} = 0.48\ \mathrm{K}) = 29.4\ \mathrm{k}\Omega$ (sample A) and $R_\Box(T_\mathrm{m} = 0.72\ \mathrm{K}) = 18.6\ \mathrm{k}\Omega$ (sample B). All data in this work were taken at a measurement frequency of 0.4–2 Hz with an ac current 0.01–1 nA.

nique. From earlier investigations, we estimated that the product of Fermi wave vector and elastic mean free path $k_F\ell < 1$ below 2 K [14].

In Fig. 1, we plot the temperature dependence of the resistance per square $R_\Box$ at zero magnetic field for two films, which are very close to disorder-tuned SIT. As the temperature decreases, $R_\Box$ of both samples increases, then reaches a maximum value at an intermediate temperature T_m, and finally drops again, showing the transition into the zero resistance state. Note that there is no drop in the resistance around 5.6 K, the bulk transition temperature of TiN. The latter would be a characteristic of a granular film with only weak tunnel coupling between the grains [15]. The absence of such a feature in our data indicates that our films are nominally homogeneous with strong metallic coupling between the crystallites [16].

From the bosonic model, the critical resistance of the zero-field superconductor-insulator transition is expected to be close to a universal value—the quantum resistance for Cooper pairs $h/(2e)^2$ [2–4]. However, this is still a controversial issue. Up to now, only for Bi films [7,8], a critical sheet resistance R_c close to the predicted value of $h/(2e)^2$ has been observed. In other materials the resistance at the transition was found to deviate significantly from the expected universal value, for instance, on Pb $- R_c \simeq 12\ \mathrm{k}\Omega$ [8], Al $- R_c \simeq 24\ \mathrm{k}\Omega$ [8], Be $- R_c \simeq 10\ \mathrm{k}\Omega$ [17]. In sample A the maximal $R_\Box$ at $B = 0$ even exceeds the value of $h/e^2 = 25.8\ \mathrm{k}\Omega$, implying that the usual perturbative theories must fail to describe the data, since the change of $R_\Box$ is much larger than $R_\Box$ itself.

In Fig. 2, we show the magnetoresistance measured at temperatures down to 60 mK and at magnetic fields up to 16 T. The resistance varies nonmonotonically with B and reaches a maximum value at a magnetic field B_m, followed by a rapid drop and gradual saturation at magnetic fields more than an order of magnitude larger than B_m. The value of B_m slightly shifts towards larger magnetic fields as the temperature increases. Yet the nonmonotonic shape of $R(B)$ still persists even when $T > T_m$. This is seen in the inset of Fig. 2 (top), where we plot magnetoresistance data in the high temperature region corresponding $dR/dT < 0$ at zero magnetic field.

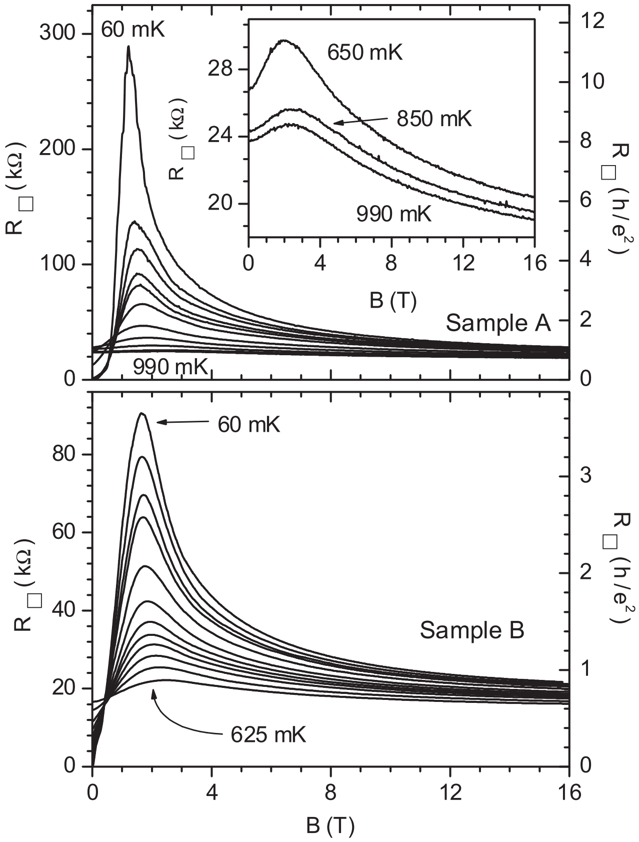

FIG. 2. Sheet resistance $R_\Box$ in perpendicular magnetic field. Top: (sample A) $T = 60$, 80, 95, 120, 140, 180, 300, 450, 650, 850, 990 mK. The inset shows a close-up view of the $R(B)$ curves measured at temperatures corresponding $dR/dT < 0$ at zero magnetic field. Bottom: (sample B) $T = 60$, 75, 90, 100, 130, 180, 220, 260, 300, 360, 480, 625 mK. $R_\Box(T = 60\ \mathrm{mK})$ reaches a maximum at $B_m \sim 1.2$ T (sample A) and at $B_m \sim 1.6$ T (sample B).

The saturation occurs at a resistance R_sat near the quantum resistance h/e^2 (see right axes of Fig. 2). Interestingly, R_sat only slightly increases as T approaches zero. This indicates metallic rather than the insulating behavior observed at B close to B_m.

We now turn to the main result of this work, which is the analysis of the negative magnetoresistance of our samples on the high-field side of the superconductor-insulator transition, where the saturation of $R(B)$ is observed (Fig. 2). First, we plotted the expression $\ln[1/R_\mathrm{sat} - G_\Box(B)]$ vs B, for each value of T. Here, $G_\square = 1/R_\square$ is the conductance per square. By varying the value of $R_{\rm sat}$ for each curve, we could linearize $\ln[1/R_{\rm sat} - G_\square(B)]$ vs B over a large range of B with a T-independent slope, as is seen in Fig. 3. This indicates that $G_\square \propto \exp(-B/B^*)$ exhibits a simple exponential decay at high magnetic fields with a characteristic magnetic field B^*. In addition, the curves in Fig. 3 show a slightly T-dependent offset.

These observations can be condensed into a simple phenomenological expression for the high-field magnetoconductance as a function of temperature and magnetic field:

$$G_\square(T, B) = 1/R_{\rm sat}(T) - \beta(T)\exp(-B/B^*), \qquad (1)$$

where B^* is a constant, which increases with the degree of disorder, and $\beta \simeq e^2/h$ is weakly T dependent and accounts for the slight offset of the curves for different T. The T dependence of $R_{\rm sat}$ and β for both samples under study is shown in Figs. 4(a) and 4(c), respectively.

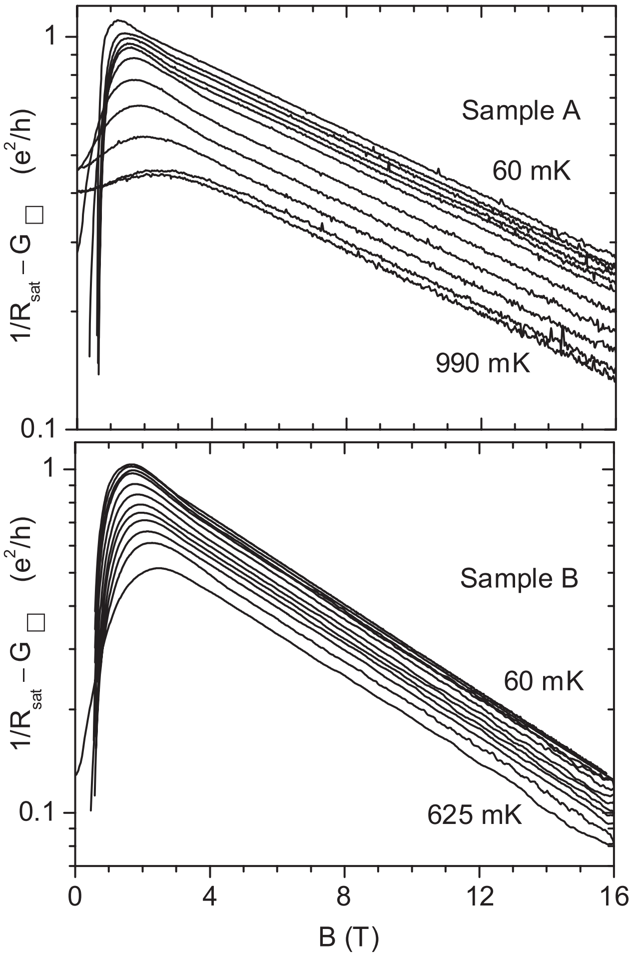

FIG. 3. Scaling plot of the data in Fig. 2. For certain values of $R_{\rm sat}$, $\ln[1/R_{\rm sat} - G_\square(B)]$ varies linearly versus B, with a T-independent slope. The linear slope corresponds to a characteristic field $B^* \simeq 10.7$ T (sample A) and $\simeq 6.8$ T (sample B), respectively.

One more important result is the temperature dependence of $R_{\rm sat}(T)$. For both samples, we have obtained a relatively weak quasimetallic T dependence of $R_{\rm sat}$. A plot of $1/R_{\rm sat}(T)$ vs $T^{1/3}$ [Fig. 4(b)] reveals that the T-dependent part of $1/R_{\rm sat}(T)$ closely follows a $T^{1/3}$ power law. In addition, for both samples $1/R_{\rm sat}(T)$ extrapolates in the limit $T \to 0$ very accurately towards e^2/h. This can already be seen in the high-field region of $R(B)$ in Fig. 2 and indicates a sample independent, possibly universal behavior. A nonmonotonic magnetoresistance similar to our results was reported also for disordered thin InO_x films [see Fig. 1(a) in [12] and Fig. 3(b) in Ref. [13]]. For the data of Ref. [12] we have performed the same analysis as for our data on TiN films and found the same scaling and the same extrapolation $R_{\rm sat}(T \to 0) = h/e^2$. The specific advantage of TiN over InO_x is its significantly lower value of B_m which allows the observation of $G_\square \propto \exp(-B/B^*)$ in a wide range of magnetic field [18].

From these observations the following scenario emerges: the superconducting state of our TiN films at $B = 0$ is rapidly destroyed by quantum phase fluctuations as the magnetic field is moderately increased, while the Cooper pairing may survive locally. It was recently suggested that strong mesoscopic fluctuations of the energy gap [19] can induce such phase fluctuations also in *homogeneous* thin films. If localized Cooper pairs and a Bose insulator exist, these are expected to be suppressed at higher magnetic fields. This corresponds to a strong decrease of $R(B)$. Phenomenologically, the exponential dependence of

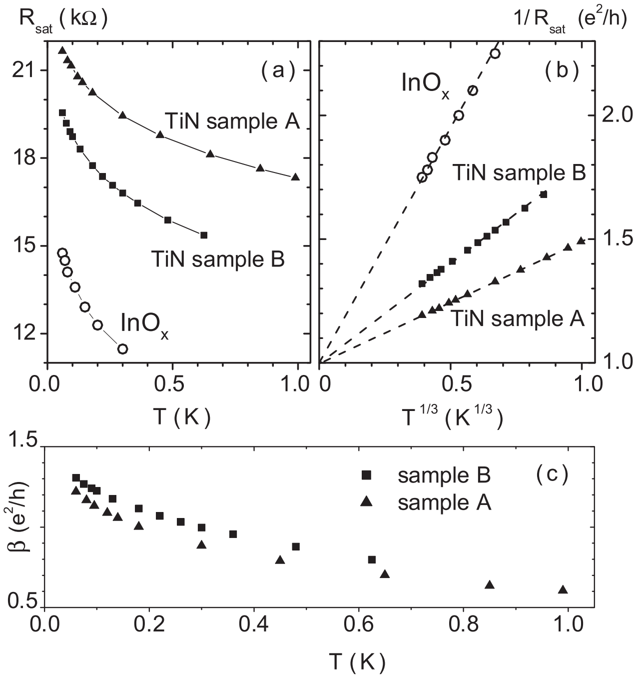

FIG. 4. Temperature dependence of $R_{\rm sat}$ and β. (a) $R_{\rm sat}(T)$, (b) $R_{\rm sat}^{-1}(T^{1/3})$, and (c) $\beta(T)$ for samples A and B and the data from Fig. 1(a) of Ref. [12]. For the latter, we obtained $B^* \simeq 4.5$ T and $\beta \simeq 2e^2/h$, nearly independent of T.

$G_\square(T, B)$ in Eq. (1) may result from a broad dispersion of binding energies of localized Cooper pairs [20]. As shown by Suzuki *et al.* [21] there is little spin-orbit scattering in TiN. Thus spin is a good quantum number in this system. In the absence of orbital pair breaking [22] and spin-orbit scattering, the Zeeman splitting of the localized Cooper pairs seems to be the likely mechanism behind the suppression of the Bose-insulator phase. At very high fields the behavior is again metallic and independent of B, however with a zero-temperature resistance, which is significantly different from R_N at high T and $B = 0$. The resistance $R_{\rm sat}(T \to 0) = h/e^2$ in this metallic phase turns out to be universal in the sense that it is independent of the material and the degree of disorder in the films.

A similar negative magnetoresistance with a saturation near h/e^2 in high magnetic fields has recently been reported for *insulating* Be films by Butko and Adams [23]. Since the saturation resistance appeared to be of purely quantum nature, these authors have introduced the term ''quantum metallicity'' for the peculiar metallic behavior at high magnetic fields. Their highly disordered films have resistances up to $\sim 4h/e^2$ in zero magnetic field and do not reveal a zero resistance state down to $T = 40$ mK. Because of the strong similarities between their insulating Be films and the superconducting InO_x and TiN films investigated here, we suggest to use the same term quantum metallicity for the high magnetic-field state, despite the evident differences in zero magnetic field. It is well possible that the insulating state in Ref. [23] is also formed by localized Cooper pairs, since there are experiments demonstrating the existence of the thickness- and the magnetic-field-tuned superconductor-insulator transition also in thin Be films [17,24,25].

These striking similarities of the high magnetic field behavior for several different materials (Be, InO_x, and our TiN films), showing the SIT, point toward a common microscopic mechanism underlying the suppression of the Bose insulator in the limit of high magnetic field. In the absence of any quantitative theory, we would like to draw attention to the fact that the observed strongly negative magnetoresistance with a tendency to saturation can be described by the remarkably simple empirical expression of Eq. (1) [26]. This expression holds in a very wide range of B and T and may prove useful for future theoretical considerations required for a deeper understanding of the nature of the Bose insulator and the high-field quantum metallic state.

We thank V. Gantmakher for providing the raw data from Ref. [12] and continuous support, A. Goldman for drawing our attention to Ref. [23], D. Weiss and W. Wegscheider for access to their high magnetic field system, and M. Feigel'man, A. Finkelstein, and V. Vinokur for useful discussions. This research has been supported by the Program ''Quantum macrophysics'' of the Russian Academy of Sciences, the Russian Foundation for Basic Research (Grant No. 06-02-16704), and the Deutsche Forschungsgemeinschaft within the GK 638.

[1] For a review, see, e.g., A. Goldman and N. Markovic, Phys. Today **51**, No. 11, 39 (1998).
[2] M. P. A. Fisher and D. H. Lee, Phys. Rev. B **39**, 2756 (1989).
[3] M. P. A. Fisher, G. Grinstein, and S. M. Girvin, Phys. Rev. Lett. **64**, 587 (1990).
[4] M. P. A. Fisher, Phys. Rev. Lett. **65**, 923 (1990).
[5] S. L. Sondhi, S. M. Girvin, J. P. Carini, and D. Shahar, Rev. Mod. Phys. **69**, 315 (1997).
[6] E. L. Shangina and V. T. Dolgopolov, Phys. Usp. **46**, 777 (2003).
[7] D. B. Haviland, Y. Liu, and A. M. Goldman, Phys. Rev. Lett. **62**, 2180 (1989).
[8] Y. Liu, D. B. Haviland, B. Nease, and A. M. Goldman, Phys. Rev. B **47**, 5931 (1993).
[9] A. F. Hebard and M. A. Paalanen, Phys. Rev. Lett. **65**, 927 (1990).
[10] G. Sambandamurthy, L. W. Engel, A. Johansson, and D. Shahar, Phys. Rev. Lett. **92**, 107005 (2004).
[11] V. F. Gantmakher, M. V. Golubkov, V. T. Dolgopolov, G. E. Tsydynzhapov, and A. A. Shashkin, Pis'ma Zh. Eksp. Teor. Fiz. **68**, 337 (1998) [JETP Lett. **68**, 363 (1998)].
[12] V. F. Gantmakher, M. V. Golubkov, V. T. Dolgopolov, A. A. Shashkin, and G. E. Tsydynzhapov, Pis'ma Zh. Eksp. Teor. Fiz. **71**, 693 (2000) [JETP Lett. **71**, 473 (2000)].
[13] M. A. Steiner, G. Boebinger, and A. Kapitulnik, Phys. Rev. Lett. **94**, 107008 (2005).
[14] T. I. Baturina, D. R. Islamov, J. Bentner, C. Strunk, M. R. Baklanov, and A. Satta, JETP Lett. **79**, 337 (2004).
[15] See, e.g., H. M. Jaeger, D. B. Haviland, B. G. Orr, and A. M. Goldman, Phys. Rev. B **40**, 182 (1989); A. Frydman, O. Naaman, and R. C. Dynes, Phys. Rev. B **66**, 052509 (2002).
[16] A. M. Finkelstein, Physica (Amsterdam) **197B**, 636 (1994).
[17] E. Bielejec and W. Wu, Phys. Rev. Lett. **88**, 206802 (2002).
[18] Typically one has $B_{\rm m} = 8$–10 T for InO_x, but only 4.6 T for the sample in Ref. [12].
[19] M. A. Skvortsov and M. V. Feigel'man, Phys. Rev. Lett. **95**, 057002 (2005).
[20] A. I. Larkin (private communication).
[21] T. Susuki, Y. Seguchi, and T. Tsuboi, J. Phys. Soc. Jpn. **69**, 1462 (2000).
[22] Preliminary measurements in parallel magnetic field result in a qualitatively similar behavior as in the perpendicular field orientation. This indicates that the orbital pairbreaking, which usually determines the upper critical field, is of less importance in our films.
[23] V. Yu. Butko and P. W. Adams, Nature (London) **409**, 161 (2001).
[24] E. Bielejec, J. Ruan, and W. Wu, Phys. Rev. Lett. **87**, 036801 (2001).
[25] E. Bielejec, J. Ruan, and W. Wu, Phys. Rev. B **63**, 100502(R) (2001).
[26] We have also tried to fit the experimentally observed decay of the resistance per square with a power law ($R - R_{\rm sat} \propto B^{-\alpha}$). In this case it was not possible to fit the data at different temperatures with the same exponent and reasonable values of $R_{\rm sat}$.