Double critical regimes at the superconductor-metal transition in ultrathin niobium films

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The upper critical field \( (H_{c2}) \) and the magnetic field induced superconductor-metal transition (SMT) are studied in ultrathin Nb films of thickness \( d \) ranging from 1.2 nm to 20 nm, which undergo a transition from polycrystalline to amorphous structure at the thickness \( d \approx 3.3 \) nm. The \( H_{c2} \) is found to be orbitally limited in polycrystalline films, and paramagnetically limited in the amorphous films. The SMT is observed in all films with \( d < 11.3 \) nm, with the critical field \( B_c \) approximately constant in polycrystalline films, and decreasing as a power-law with the film conductance \( G \) in the amorphous films. The scaling analysis identifies two different critical regimes in the amorphous films, with the critical exponents consistent with the \((2+1)D \) XY model for a 2D superconductor in the clean (disordered) limit at high (low) temperatures, respectively; in addition, at the lowest \( T \) quantum metal phase is observed. These results suggest inhomogeneous nature of the amorphous films, in the form of superconducting islands immersed in the metallic background. Some unusual features, not observed in other systems, include the suppression of SC correlations on cooling, and the exponent of the power law in the dependence of \( B_c \) on \( G \), which differs from theoretical predictions. These features may be caused by paramagnetic pair breaking, or by the proximity of quantum metal phase.

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I. INTRODUCTION

Superconducting (SC) state in homogeneously disordered, two-dimensional (2D) films may be destroyed by tuning of various parameters, such as, for example, external magnetic field \( (B) \), disorder, or doping. The increase of the field induces two, distinctly different transitions: superconductor-insulator transition (SIT) or superconductor-metal transition (SMT) in strongly or weakly disordered films, respectively. The nature of these transitions is not fully understood. Two early scenarios have been considered, fermionic and bosonic, predicting that the Cooper pairs are either broken or survive the transition, respectively. Over the last decade, the development of microscopic inhomogeneities in the form of intertwined SC and non-SC islands near the transition has been proposed. The SC islands are indeed observed by scanning tunneling spectroscopy near the SIT, while conflicting tunneling results are reported for the SMT.

The SIT/SMT is a quantum phase transition (QPT). While it occurs at \( T = 0 \), it affects the behavior of the observables at nonzero \( T \), leading to specific scaling laws, which may be tested experimentally. In case of \( B \)-induced SIT/SMT the scaling analysis has been performed for many 2D disordered films. The variety of the correlation-length critical exponents \( \nu \), extracted from these experiments, defies easy classification. However, several recent reports successfully ascribe unusual scaling behaviors to specific disorder present in the films. For example, two-stage SIT with different scaling exponents in copper oxide films is attributed to distinct vortex glass phases at different \( B \), while the SMT with different critical regimes in a 2D electron gas at SC oxide interfaces is explained by the development of proximity-coupled SC islands, in agreement with theoretical predictions. The other interesting results include the observation of diverging dynamical critical exponent \( z \) (Griffiths singularity) near SMT in several systems.

Here, we use scaling analysis to study the \( B \)-induced SMT in the thin niobium (Nb) films. This issue has not been investigated before, despite many studies devoted in the past to properties of Nb films. The films studied here, with thickness \( d \) between 1.2 nm and 20 nm, are sandwiched between two Si barrier layers of 10 nm to prevent oxidation, as described recently. With decreasing \( d \) the films undergo the change of structure from polycrystalline to amorphous at \( d \approx 3.3 \) nm, accompanied by the growing contribution of electron carriers to the conduction, which in the bulk Nb is dominated by holes. X-ray Photoelectron Spectroscopy evaluation suggests that this effect is due to strong surface scattering of holes, enhanced by a small admixture of Si ions (at the level of 5-10 at.%) into the Nb layer closest to the interface.

In the present study we find that the upper critical field \( (H_{c2}) \) is orbitally limited in polycrystalline films, but becomes paramagnetically limited in amorphous films. The scaling analysis reveals that in the amorphous films depending on temperature two different critical regimes, with different critical magnetic field, \( B_c \), and different product of scaling exponents \( \nu z \), are observed. In the low-\( T \) (high-\( T \)) regime \( \nu z \) is larger (smaller) than 1, consistent with the \((2+1)D \) XY model for 2D superconductor in the dirty (clean) limit, respectively (assuming that \( z = 1 \)).
TABLE I: Parameters of Nb films: thickness \(d\) (nm), sheet resistance \(R_N\) (\(\Omega\)), film transition temperature \(T_c\) (K), SMT critical field \(B_c\) (T), upper critical field \(\mu_0H_{c2}(0)\) (T), coherence length \(\xi(0)\) (nm).

| \(d\) (nm) | \(R_N\) (\(\Omega\)) | \(T_c\) (K) | \(T_{\text{on}}\) (K) | \(l\) (nm) | \(B_c\) (T) | \(\mu_0H_{c2}(0)\) (T) | \(\xi(0)\) (nm) |
|-----------|----------------|-----------|----------------|-----------|-----------|----------------|-----------|
| 1.2       | 2712           | 0         | 0.35           | 0.15      | 0.29      | 15.0           | 14.8      |
| 1.3b      | 1873           | 0.8       | 1.3            | 0.21      | 1.19      | 11.7           | 11.7      |
| 1.3a      | 1503           | 0.86      | 1.5            | 0.26      | 1.45      | 1.46           | 15.0      |
| 1.4       | 1653           | 1.19      | 1.87           | 0.22      | 1.46      | 1.5            | 14.8      |
| 2.2       | 886            | 2.28      | 2.28           | 0.26      | 2.52      | 2.41           | 11.7      |
| 3.2       | 578.8          | 2.38      | 3.13           | 0.27      | 3.48      | 3.28           | 10.0      |
| 3.3       | 447.9          | 2.29      | 2.99           | 0.34      | 3.5       |                |           |
| 3.9       | 359.3          | 2.67      | 3.39           | 0.36      | 3.4       |                |           |
| 5.3       | 137.2          | 4.14      | 4.62           | 0.69      | 3.43      | 3.13           | 10.3      |
| 6.7       | 120.9          | 4.75      | 5.22           | 0.62      | 3.5       |                |           |
| 7.6       | 78.5           | 5.43      | 5.75           | 0.84      |           |                |           |
| 9.5       | 38.0           | 6.31      | 6.48           | 1.33      | 3.32      | 3.15           | 10.2      |
| 11.3      | 30.8           | 6.16      | 6.29           | 1.37      | 3.3       |                |           |
| 16        | 6.97           | 7.49      | 7.6            | 3.22      |           |                |           |
| 20        | 7.83           | 7.6       | 7.7            | 2.40      | -         | 2.64           | 11.2      |

We discuss the possible link of these features to the spin paramagnetism of ultrathin Nb films, or to the approach to quantum metal state, which is observed at the lowest temperatures.

II. EXPERIMENTAL DETAILS

The resistance per square, \(R_{sq}\), as a function of temperature and magnetic field up to 9 T, perpendicular to the film plane, was measured on a lithographically patterned resistance bridge using a standard four-probe method, with dc current for \(T > 2\) K, and low frequency, ac current for the millikelvin-\(T\)-range down to 80-100 mK. Care was taken to remain in the Ohmic regime. In the \(300\) range the data were accumulated during the field sweep, while at higher \(T\) both the field, and the temperature sweeps were used. To cover various \(T\)-ranges in different samples several different cryostats were used. He-4 or PPMS for high-\(T\) range, LHe dilution refrigerator for \(T < 1\) K, and \(^3\)He cryostat for \(0.3\) K \(< T < 2\) K.

The details on the film preparation and calibration of \(d\) have been described previously. In Appendix A we include a brief summary of the most important previous findings on the film morphology, together with some additional details on the measurement methods. Since the dependence of the \(R_{sq}\) on \(d\) is mostly monotonic, we use \(d\) values to label different films. However, some fluctuations of film thickness (\(~10\%) are unavoidable for small \(d\), contributing to scattering of \(R_{sq}\) values; this is illustrated by two different films with \(d = 1.3\) nm, labeled “a” and “b”. Table lists all films in the set, together with parameters determined in this study. First four parameters are: the normal-state resistance measured at \(T = 10\) K (\(R_N\)), the midpoint of the SC transition temperature at \(B = 0\) (\(T_c\)), the onset of SC transition (\(T_{\text{on}}\)), defined at the point where \(R_{sq}/R_N = 0.95\), and the mean free path (\(l\)), estimated based on the \(R_N\) value. For polycrystalline films we use \(\rho_{N,T} = \text{const}\), where \(\rho_N = R_Nd\) is the resistivity, and \(\text{const} = 3.7 \times 10^{-12}\Omega\) cm\(^2\) for crystalline niobium. For amorphous films a modification relating \(\text{const}\) to residual resistivity has been proposed, and we use it for our estimate. Despite this modification the \(l\) values in the thinnest films fall below the average atomic distances, indicating that \(l\) is underestimated, probably due to differences between actual band structure parameters and those used in Ref.22.

III. RESULTS

A. Magnetoresistance

Figs. show the dependencies of \(R_{sq}(T)\) for various \(B\) for films with \(d\) equal to 9.5 nm (a), 3.9 nm (b), 1.4 nm (c) and 1.2 nm (d). The SC transition at \(B = 0\) is present in all films with \(d \geq 1.3\) nm, with the width increasing with decreasing \(d\), from 0.1 \(\pm\) 0.4 K for polycrystalline films, to about 2 K for amorphous films. Fig. (d) shows that even in the film with \(d = 1.2\) nm non-complete SC transition is seen below 1 K. With the increase of the magnetic field the SC transition broadens, and the broadening grows with the decrease of \(d\). In polycrystalline samples below the \(T_c\) the resistance shows activated behavior, with activation energies consistent with collective vortex pinning. On the other hand, on cooling of the amorphous films, after the initial activated region, the resistance saturates at the finite, \(B\)-dependent level, which is smaller than the normal-state resistance. An example of this behavior is shown in Fig. (c); the details will be discussed elsewhere. Similar saturation has been observed for many other films with SMT. It indicates the formation of quantum metallic phase, whose nature is still under debate. The increase of the magnetic field leads eventually to a negative magnetoresistance (MR) at high magnetic field has been reported for weakly.
disordered films α-Mo$_2$Si$_{1-x}$ and Nd$_2$-Ce$_2$CuO$_{4+x}$, and attributed to the presence of SC fluctuations on the nonsuperconducting side of the SMT.

The characteristic feature of SMT transition is that at some critical magnetic field, $B_c$, and critical resistance, $R_c$, the derivative $dR_{sq}/dT$ switches from being positive to negative. The $B_c$ is indicated by red arrows in Figs. (c) and (d). This type of behavior produces crossing of the isotherms on the $R_{sq}(B)$ graphs. The examples of such graphs are presented in Figs. (a-c) for films with $d$ equal to 9.5 nm (a), 1.4 nm (b), and 1.2 nm (c). We observe such crossings in all Nb films with $d \leq 11.3$ nm. The $B_c$ (in the limit of lowest measured $T$) is almost $d$-independent in all polycrystalline films, equal to about 3.4 T, while it is reduced in amorphous films with decreasing film thickness to about 0.29 T in the film with $d = 1.2$ nm. The $R_c$ increases monotonously with the decrease of $d$ in all films, reaching the value of about 2.85 kΩ in the film with $d = 1.2$ nm. This value is much smaller than the quantum resistance for Cooper pairs, $R_Q = \hbar/4e^2 \approx 6.45$ kΩ, at which the SIT is predicted to occur in the bosonic scenario.

Interestingly, in all amorphous films the $B_c$ exhibits peculiar behavior displayed in the right inset to Fig. (c). Namely, the crossing of the isotherms, which defines $B_c$, remains constant within experimental accuracy below some temperature $T_0$, which decreases with decreasing $d$, from about 0.5 K (for $d = 2.2$ nm), down to about 0.2 K (for $d = 1.2$ nm). However, at higher temperatures the crossings of the consecutive pair of isotherms shift to higher value for most of the films, except for the film with $d = 1.2$ nm. An example of such shift of isotherm crossing for film with $d = 1.4$ nm is presented in detail in Appendix B. A plot of the temperature $T_0$ versus $B_c(0)$ (which is the $B_c$ measured at the lowest $T$...
for each film), on a double logarithmic scale, is shown in left inset to Fig.2(c). It reveals that \( T_0 \) scales approximately as \( \sqrt{B_c(0)} \). The shift of the \( B_c \) has been reported previously for some systems.\(^{20,25,63,67}\) However, usually the \( B_c \) is seen to shift to higher values on cooling as a result of the strengthening of SC correlations as the system evolves deeper into SC state. Such a behavior is seen in the present experiment only in the case of the thinnest film (\( d = 1.2 \) nm) with non-complete SC transition. In all other amorphous films we observe shift of the \( B_c \) to lower values on cooling towards \( T_0 \), what suggests the suppression of SC correlations.

The relation between \( B_c(0) \) and \( R_c \) for all films may be summarized on a log-log plot of the \( B_c(0) \) versus critical conductance, \( G_c = 1/R_c \) (inset to Fig.2(b)). Qualitatively different behavior is seen in the polycrystalline and in the amorphous films. While in the polycrystalline films the \( B_c(0) \) is very weakly dependent on \( G_c \), in the amorphous region it is well described by the power law, shown by blue line in the inset to Fig.2(b), \( B_c(0) = A(G_c - G_{c0})^p \), where \( A \) is a constant and \( p = 0.6 \pm 0.03 \). The value of \( G_{c0} = 3.3 \times 10^{-2} \, \Omega^{-1} \), at which the \( B_c(0) \) reaches zero, is slightly smaller than the conductance of the \( d = 1.2 \) nm film (equal to \( 3.5 \times 10^{-4} \, \Omega^{-1} \)). Therefore, \( G_{c0} \) most likely has a meaning of the conductance of a metallic, non-SC background. Looking for the possible interpretation of such behavior we note that the power-law dependence of \( B_c \) on the film conductance has been predicted by a model of disordered array of SC puddles (or islands) coupled by a metallic background through a proximity effect.\(^{11}\) Since in our films with decreasing \( d \) a growing contribution of electron carriers to the conductance is detected at low temperatures,\(^{38}\) it is possible that these carriers form metallic, non-SC background. We note, however, that the exponent of power law \( p \), which we observe, differs from exponents calculated in Ref.\(^{12}\), which are predicted to be 1 or 1.4 for low and high film conductance, respectively. We will return to the discussion of these features later.

B. Upper critical field

It is important at this point to estimate the upper critical field, \( H_{c2} \). We define \( H_{c2} \) using the point of the SC onset (\( R_{so}/R_N = 0.95 \)). This eliminates the influence on the \( H_{c2} \) of the vortex-related broadening of the transition, which is very large here. Fig.3(a) shows \( H_{c2}(T) \) for representative films. We observe that the slope of the \( H_{c2}(T) \)-line at \( T_c \), \( (dH_{c2}/dT)_{T_c} \), increases with decreasing \( d \) in polycrystalline films, and slightly decreases in amorphous films. Since the slope is proportional to \( N_F\rho_N n, \) where \( N_F \) is the density of states,\(^{56}\) the initial increase is most likely caused by the increase of \( \rho_N \) due to enhanced surface scattering, while the slight decrease in amorphous films must be related, in addition, to the decrease of \( N_F \), which compensates the increase of \( \rho_N \). We also see that \( H_{c2}(0) \) is rapidly reduced in the amorphous films, and in the thinnest films at the lowest \( T \) a small region of double-valued \( H_{c2} \) appears, as shown in detail for film with \( d = 1.4 \) nm in Fig.3(b), where \( H_{c2} \) displays maximum at \( T_{max} \approx 0.42 \) K.

Solid lines show the fits to the data using the conventional one-band, dirty limit WHH theory,\(^{27}\) which takes into account both spin paramagnetism and spin-orbit scattering through the Maki parameters \( \alpha \) and \( \lambda_{so} \). In the absence of spin-paramagnetic effect and spin-orbit interaction (\( \alpha = 0 \) and \( \lambda_{so} = 0 \)) the orbital pair breaking limits the upper critical field, which is given by \( H_{orb}(0) = H_{c2}(0) = -A(dH_{c2}/dT)_{T_c}, \) with the prefactor \( A \) proportional to \( 2\Delta/kT_c \). In the presence of spin-paramagnetic effect the upper critical field is modified according to \( H_{c2}(0) = H_{orb}(0)/\sqrt{1 + \alpha^2} \), where \( \alpha = \sqrt{H_{orb}(0)/H_P(0)} \), and \( H_P(0) \) is the zero-temperature paramagnetically limited field.\(^{58}\) In our calculation \( A \) is adjusted to account for an increase of \( 2\Delta/kT_c \) with decreasing Nb film thickness (from 3.9 in polycrystalline films to 4.5 in ultrathin amorphous films),\(^{28}\) while \( \alpha \) and \( \lambda_{so} \) are treated as adjustable parameters. Thanks to wide \( T \)-range of the data in amorphous films, the Maki parameters may be estimated with reasonable accuracy;\(^{58}\) they are listed in Table III in Appendix C, in which we also quote the WHH formulas used in the fits.

Based on the fits, we estimate \( H_{c2}(0), H_{orb}(0), \) and \( H_P(0) \), which are plotted as a function of \( d \) in Fig.3(c), together with Maki parameters. The zero-temperature coherence length \( \xi(0) \), calculated using Ginsburg-Landau formula \( \rho_0H_{c2}(0) = \Phi_0/2\pi\xi^2(0) \) with \( \Phi_0 = 2.07 \times 10^{-15} \) Wb, is listed in Table III. The \( \xi(0) \) is larger than \( d \) for all films with \( d \leq 9.5 \) nm, confirming that the films are 2D. The plots indicate the initial increase of \( H_{c2}(0) \) with the decreasing \( d \) in polycrystalline films, followed
by a rapid decrease in amorphous films. This dependence is related to the interplay of $H_{\text{orb}}$ and $H_P$. While the $H_P(0)$ is decreasing monotonously with decreasing $T_c$, $H_{\text{orb}}(0)$ increases sharply on the approach to polycrystalline/amorphous boundary due to enhanced surface scattering. As a result, while in thicker polycrystalline films the orbital pair breaking dominates, on the approach to amorphous region the $H_{c2}$ becomes paramagnetically limited.

We note also that the dependence of $\lambda_{so}$ on $d$ follows essentially the dependence of $H_{\text{orb}}$ on $d$ (which is reasonable), that is, $\lambda_{so}$ is negligible for the thickest films, becomes the largest at the polycrystalline-amorphous boundary, and it is strongly suppressed in the thinnest films. According to WHH theory, the $H_{c2}$ should become double-valued when Maki parameter $\alpha$ exceeds the value $\alpha_c = (1 + 1.589 \lambda_{so}/\lambda^c)/(1 - \lambda_{so}/\lambda^c)$, where $\lambda^c = 0.539$. This condition is fulfilled in the present experiment for the thinnest films, in which we indeed observe double-valued $H_{c2}$.

C. Scaling analysis

We now turn attention to the scaling analysis. We will use conventional assumption that the magnetic field $B_c$, which is the crossing point of the isotherms, defines the critical field, at which QPT may occur in the $T = 0$ limit. We note that such an assumption has been challenged recently by the studies of the ac-conductivity data for films of InO$_x$, which show that the relation between the $B_c$ and SC correlations is complicated, and depends strongly on the disorder. However, since in case of thin Nb films none such data exists, we proceed here with conventional assumption, and treat the present results as a first step to more advanced studies in the future.

It is predicted that in the vicinity of the QPT at $T = 0$ the spatial correlation length $\xi$ and the dynamical correlation length $\xi_d$ diverge as a power law on the approach to the critical point $B_c$, $\xi \propto |\delta|^{-\nu}$ and $\xi_d \propto |\xi|^z$, where $\delta = B - B_c$, and $\nu$ ($z$) is spatial (dynamical) critical exponent. At $T \neq 0$ the time dimension is limited by temperature fluctuations, what introduces $T$-dependent dephasing length, $L_\phi \propto T^{-1/\nu}$, beyond which quantum fluctuations lose phase coherence. This leads to a prediction that in the critical region all relevant quantities are universal functions of the scaling variable $|\delta|T^{-1/\nu}$. Here, in order to verify scaling hypothesis, we use two methods:

1. We test if the resistance data, measured at various fixed temperatures in the vicinity of $B_c$ may be collapsed on a single curve given by $R_{sq}(B,T) = R_{sq}(0)|B - B_c|t$ by adjusting the parameter $t(T)$ at each temperature, where $t$ should follow the power law, $t(T) = T^{-1/\nu}$. Fig.4 shows examples of this analysis for films with $d = 9.5$ nm, 2.2 nm, and 1.4 nm.

2. We calculate the partial derivative, $(\partial R_{sq}/\partial B)_{B_c} \propto T^{1/\nu}$, and plot it versus $1/T$ to extract the slope, which gives inverse of the product of critical exponents $\nu z$. This is illustrated in Fig.5.

As shown in Fig.4(a), in the case of polycrystalline film with $d = 9.5$ nm a good collapse of the data is obtained, with a single critical field, and product of critical exponents $\nu z = 0.6 \pm 0.1$. However, this is not the case for amorphous films. Fig.4(c) shows that for film with $d = 1.4$ nm we can identify two different $T$-ranges, in which data may be collapsed. One is low-$T$ range, 0.1 K $< T < 0.4$ K, with a critical field $B_c = 1.46$ T, and $\nu z = 2.4 \pm 0.2$, and the other is high-$T$ range, 0.6 K $< T < 1$ K, for which we find $\nu z = 0.9 \pm 0.2$. Note that in the high-$T$ range on the increasing $T$ the $B_c$ shifts slightly, from 1.58 T to 1.69 T (as shown in the inset to Fig.4(c)), nevertheless good collapse may be achieved with average $B_c = 1.66$ T. The critical resistances in these two ranges are also slightly different. Qualitatively similar behavior, with two distinct $T$-ranges, is seen in the case of film with $d = 2.2$ nm (Fig.4(b)), for which we
which shifts with $B$ low- marked by small vertical arrows in Fig. 5(a). On the other slope is zero—this is the quantum metal region. When $d$ film with films vertically, so all of them overlap the data for the plot in Figs. 5(b) and 5(c) the data for low- $K$) with smaller slope. In addition, at the lowest $T$ temperature different investigated temperature range. Instead, we observe two several amorphous films. It is evident that the data do obtain $\nu_\tau = 1.6 \pm 0.2$ in low-$T$ range, and $\nu_\tau = 0.8 \pm 0.1$ in the high-$T$ range.

These observations are confirmed by second method. Fig. 5(a) shows the plot of $(\partial R_\tau / \partial B)_B$ versus $1/T$ for several amorphous films. It is evident that the data do not follow straight lines with the same slope across whole investigated temperature range. Instead, we observe two different $T$-ranges with distinctly different slopes, marked at the top of the figure by arrows: high-$T$ range ($T \gtrsim 0.5$ K) with larger slope, and low-$T$ range ($0.12 \, K \lesssim T \lesssim 0.35$ K) with smaller slope. In addition, at the lowest $T$ ($T \lesssim 0.12$ K) the $R_\tau$ is saturated (labeled “sat”), so that the slope is zero—this is the quantum metal region. When calculating the derivative, we took care to determine it at actual crossings of consecutive isotherms (actual $B_c$), which shifts with $T$ at $T > T_0$; this shift, however, does not change the slope, so it has no influence on the value of $\nu_\tau$. Note that the crossover between high-$T$ range and low-$T$ range is not the same for all films, but increases with increasing $d$; in fact, it occurs approximately at the temperature $T_0$, below which $B_c$ is constant. The $T_0$ is marked by small vertical arrows in Fig 5(a). On the other hand, the $B_c$ remains constant at the crossover between low-$T$ range and quantum metal region; there is no apparent signature of this crossover in the $B_c$ value.

Interestingly, in both high-$T$, and low-$T$ ranges the slopes for many different films are similar, suggesting similar critical exponents. To emphasize this point, we plot in Figs. 5(b) and 5(c) the data for low-$T$ and high-$T$ range, respectively, after shifting the data for different films vertically, so all of them overlap the data for the film with $d = 1.4$ nm (this amounts to multiplying data for each film by a different constant, $C_1$ or $C_2$ for high-$T$ or low-$T$ range, respectively). In the high-$T$ range we include, in addition, the data for polycrystalline films—it is seen that they show exactly the same slope as the data for amorphous films. From the slope of straight lines, fitted to all data for different films in each $T$-range, average critical exponents are determined: $\nu_\tau = 0.6 \pm 0.1$ in high-$T$ range, and $\nu_\tau = 2.2 \pm 0.2$ in low-$T$ range.

We summarize the scaling analysis results in Fig. 5(a), where points show the $d$-dependence of $\nu_\tau$ extracted separately for each film using method (1) and (2), while lines show the average values as fitted to data in Figs. 5(b) and 5(c). The high-$T$ values are all similar, independent of the sample or the method used. On the other hand, low-$T$ values of $\nu_\tau$ extracted separately are considerably scattered, between 1.5 and 2.4, possibly due to small $T$-range of the mK measurements in case of some films. Nevertheless there is no doubt that the low-$T$ exponent...
in the amorphous films exceeds 1.

D. Discussion

Before further discussion of this result, we construct a $T = 0$ phase diagram of Nb films in the $B - d$ plane (Fig. 6(b)), $d$ on a logarithmic scale). The left (linear) scale shows $H_{c2}(0)$ and the critical field $B_c(0)$. Note that in polycrystalline films the $B_c(0)$ is somewhat higher than the $H_{c2}(0)$, what most likely results from arbitrary definition of $H_{c2}(0)$. On the other hand, with decreasing $d$ in the amorphous films the $B_c(0)$ approaches $H_{c2}(0)$, or even becomes slightly smaller. This is caused by the shift of $B_c$ to lower values with the decrease of $T$ towards $T_0$ in the amorphous films, as shown in the insets to Fig. 2(c).

The fact that $B_c(0)$ and $H_{c2}$ are close is similar to results observed in other weakly disordered systems with the SMT; this is in contrast to the materials with the SIT, in which usually $B_c(0)$ is much smaller than $H_{c2}$.

On the right (logarithmic) scale we show film conductance $G_e - G_{e0}$. The continuous green line shows the fit to the data, which indicates that for $d \geq 1.3$ nm the conductance follows a power law dependence on the film thickness, $G_e - G_{e0} \sim d^{-2}$, consistent with the surface scattering in thin films, as has been previously discussed. $G_e - G_{e0}$ drops by an order of magnitude at $d = 1.2$ nm, in accordance with the fact that only non-complete SC transition survives in this film. As already mentioned, the $B_c(G_e)$ dependence suggests a possibility that in the amorphous films at low $T$ inhomogeneities develop, in the form of disordered arrays of SC islands coupled via metallic background by proximity effect. On the other hand, no sign of such inhomogeneities exist in polycrystalline films. We sketch this behavior schematically in the insets of Fig. 3(b), where blue are the SC regions, and pink depicts the metallic, non-SC background.

The high-$T$ value of $\nu z$ observed here is close to 2/3. Such $\nu z$ has been observed in conventional 2D superconducting films, for example, in a-Bi$_2$Sr$_2$CaCu$_2$O$_8$ or a-NbSe$_2$. This is consistent with $(2+1)$D $XY$ model for a 2D superconductor in the clean limit, provided that $z = 1$, which is the usual assumption in the case of the system with long-range Coulomb interactions between charges, confirmed experimentally in several materials. $\nu z$ increases with decreasing temperature, at high $T$ it may be smaller than the size of the SC island, while at low $T$ it may encompass many islands coupled by proximity effect, so that at low $T$ the disorder becomes evident in the scaling exponent.

It is possible that the observation of different critical regimes in the present experiment is also enabled by the $T$-dependence of the $L_\phi$. However, it is important to stress that in the Nb films we observe features, which are distinctly different from behavior of 2D electron gas. These include the exponent in the power-law dependence of $B_c$ on $G_e - G_{e0}$, $p = 0.6$, which differs from the theoretical prediction, and the peculiar suppression of the $B_c$ in the clean regime on the approach to $T_0$, which suggests the suppression of SC correlations. In addition, at the lowest $T$ quantum metal state develops. In the following we discuss the possible explanations of these features.

Considering first the value of the exponent $p$ we note that the theoretical calculations have been done for two limiting cases, with magnetic field coupling either to purely orbital (in perpendicular field) or to purely spin (in parallel field) degrees of freedom. While the results suggest the power-law dependence of the $B_c$ on conductance in both limits, the exponent of the power law is not directly comparable to the exponent observed in our experiment, in which both orbital and spin effects are important.

Turning now to the problem of the suppression of the $B_c$, we propose two scenarios. One possibility is that the paramagnetic pair breaking may be responsible for the suppression of SC correlations on the approach to $T_0$. This idea is supported by the fact that the dependencies $B_c(T)$ and $H_{c2}(T)$ are closely correlated. We illustrate this correlation in Fig. 3(b) by the data for film with $d = 1.4$ nm, together with the color map of the derivative $dR_{sq}/dT$. The dependence of the $B_c$ on $T$ traces the path along which $dR_{sq}/dT$ is close to zero. In particular, the transition from clean to disordered regime at $T_0$ appears in close vicinity of the maximum of $H_{c2}$ at $T_{max}$. Since the dependence of $H_{c2}$ on $T$ is explained by taking into account the paramagnetic pair breaking, it is likely that
the paramagnetism of Nb films plays a decisive role in the \( B_c \) suppression as well. It follows that the apparent transition from the clean limit critical regime to disordered regime may also be related to the paramagnetism of thin Nb films. We note that the shape of \( H_{c2}(T) \), with the decrease below \( T_{\text{max}} \), resembles the phase diagram of Zeeman-limited superconductivity in the presence of parallel magnetic field in thin Al or Be films\cite{24, 25}, which displays two characteristic regions: second order transition from SC to normal state at \( T > T_{\text{max}} \), and the first order transition from SC to spin polarized normal state below \( T_{\text{max}} \). In our experiment with perpendicular magnetic field no first order transition is present. Nevertheless it is possible that the thinnest films at low-\( T \) resemble a system of locally SC islands with zero spin magnetization, immersed in the metallic background with non-zero Pauli spin susceptibility. The experiments on the \( H_{c2} \) in parallel magnetic field, similar to experiments on Zeeman-limited superconductivity\cite{24, 25} should verify this scenario.

Another possibility is that the suppression of SC correlations is somehow related to the development of the quantum metal state, observed at the lowest \( T \). In the Nb films quantum metal develops smoothly from the disordered regime with \( \nu_z > 1 \), without any obvious signature of this development in the value of the \( B_c \). This poses an interesting question: is it possible that the disordered regime is in fact a precursor of the quantum metal, i.e. is it a transition region between the clean limit regime and the quantum metal? We recall here that the recent studies of the ac-conductance in the weakly disordered InO\(_x\) find that the quantum metal is a highly unusual state, with no cyclotron resonance\cite{21}, in agreement with earlier suggestions of the absence of the Hall effect in InO\(_x\) and TaN\(_x\) films\cite{70}. Moreover, the ac conductance data indicate that with the increasing \( B \) the SC correlations acquire gradually decreasing length scales, until they are suppressed completely above the \( B_c \)\cite{21, 25}. It is unclear if similar behavior could be expected in Nb films, because the peculiar properties, which we observe near \( T_0 \), have not been reported for any other system with resistance saturation\cite{12, 28, 61}. It is possible that in thin Nb films both the quantum metal state, and the paramagnetic pair breaking coexist, producing behaviors unlike those in any other system.

**IV. SUMMARY**

We have examined the \( B \)-induced SMT in ultrathin Nb films of thickness \( d \) ranging from 1.2 nm to 20 nm, which undergo a transition from polycrystalline to amorphous structure at the thickness \( d \approx 3.3 \) nm. The surprising effect observed here is the robust paramagnetic limit of the upper critical field in the amorphous films leading to double-valued \( H_{c2} \) in the thinnest films, in which spin-orbit interaction is suppressed.

The SMT is observed in all films with \( d < 11.3 \) nm, with the critical field \( B_c \) approximately constant in polycrystalline films, and decreasing as a power-law with the film conductance in the amorphous films. The scaling analysis identifies two different critical regimes in the amorphous films, with the critical exponents consistent with the clean limit at high-\( T \), and with the disordered (dirty) limit at low-\( T \). In addition, at the lowest \( T \) quantum metal phase appears. The transition from the clean to disordered regime occurs in the vicinity of the temperature \( T_0 \), which scales as \( \sqrt{B_c} \). These observations suggest the developments of inhomogeneities in the vicinity of the SMT, in qualitative agreement with a model of SC puddles coupled by a metallic background through a proximity effect\cite{21}. However, there are some unusual features, not observed in other systems. These include the suppression of SC correlations on cooling on the approach to \( T_0 \), and the exponent of the power law in the dependence of \( B_c \) on \( G \), which differs from theoretical predictions. These features may be caused by paramagnetic pair breaking, or by the proximity of quantum metal phase.

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**Appendix A: Film preparation and characterization**

The films were made by magnetron sputtering at room temperature on glass substrates, with niobium sandwiched between Si-buffer layers to prevent Nb oxidation\cite{38}. High-resolution transmission electron microscopy and X-ray diffraction studies\cite{38} indicate that the films with \( d < 3.3 \) nm are amorphous, without any indications of gross structural inhomogeneity. The thickness of the films is quite uniform, without any traces of polycrystalline grains. On the other hand, all polycrystalline films contain a uniformly thin layer (of thickness of about 1.5 nm) of amorphous Nb at the Nb/Si interface, formed in the initial stage of the Nb deposition. Note that this amorphous layer is thinner than 3.3 nm. This suggests that, when \( d \) exceeds 3.3 nm during Nb deposition, the polycrystalline grains begin to form, and part of the amorphous layer at the interface recrystallizes. X-ray Photoelectron Spectroscopy of the thinnest films is consistent with small admixture of Si ions (5 to 10 at.%) into Nb layer closest to the interface\cite{38}. The change of
sign of the Hall coefficient is observed, from positive in thick polycrystalline films, to negative in thinnest films at the lowest temperatures; it is most likely related to strong surface scattering of hole carriers.

For magnetoresistance measurements the films were cut into 5 × 5 mm² size and they were lithographically patterned into "Hall bar" structure, with the current path 2 mm long and 200 μm wide, as shown in Fig. 7(a) (current contacts are marked A-B). The measurements at low temperatures, T < 1 K, were carried out in LHe dilution refrigerator with low-pass filters. Resistance was measured by low-frequency (f = 19 Hz) ac lock-in techniques with I = 10 nA.

Appendix B: Isotherm crossing

Fig. 7(b) shows the dependence of R_{sq} on B for series of temperatures below 1K for a film with d = 1.4 nm. To extract B_c and R_c we determine the crossing points of consecutive isotherms. Starting from high temperatures, when T is reduced from 1 K towards 0.4 K, as shown by grey curved arrow, the crossing points, marked by crosses at the intersections of dashed lines, shift towards lower B. However, when T is reduced below 0.36 K, as shown by black curved arrow, the crossings points (marked by stars at intersections of continuous lines) reach some minimum value of B, and possibly even start to increase slightly (this increase is within experimental error of the measurement).

The inset to Fig. 7(b) shows the T-dependence of B_c and R_c values determined by this procedure. Within experimental accuracy the B_c and R_c are seen to be constant below the temperature T_0 marked in the figure. The value of B_c, averaged for T < T_0, is equal to 1.46 ± 0.01 T.

Appendix C: Upper critical field

According to WHH theory the upper critical field in the dirty limit can be calculated using the following equation:

\[
\ln \frac{1}{T} = \left( \frac{1}{2} + \frac{i\lambda_{so}}{4\gamma} \right) \psi \left( \frac{1}{2} + \frac{\bar{h} + \lambda_{so}/2 + i\gamma}{2t} \right) + \left( \frac{1}{2} - \frac{i\lambda_{so}}{4\gamma} \right) \psi \left( \frac{1}{2} + \frac{\bar{h} + \lambda_{so}/2 - i\gamma}{2t} \right) - \psi \left( \frac{1}{2} \right),
\]

where t = T/T_c, γ ≡ [(α\bar{h})^2 - (λ_{so}/2)^2]^{1/2} and

\[
\frac{\bar{h}}{(-d\bar{H}/dt)_{t=1}} = \frac{\pi^2\bar{h}}{4} = \frac{H_{c2}}{(-dH_{c2}/dt)_{t=1}}.
\]

When α = 0 and λ_{so} = 0, in the absence of the spin-paramagnetic effect and the weak spin-orbit interaction upper critical field is described by:

\[
\ln \frac{1}{T} = \psi \left( \frac{1}{2} + \frac{\bar{h}}{2t} \right) - \psi \left( \frac{1}{2} \right),
\]

than μ_0H_{c2}(0) = -A(dμ_0H_{c2}/dT)_{T=T_c}, where the numerical factor A is proportional to 2Δ/kT_c.

In BCS weak-coupling superconductors with 2Δ/kT_c = 3.52, A is equal to 0.69. Since the Nb is an intermediate-coupling superconductor, 2Δ/kT_c is larger, so that A is larger as well. It has been shown by Park and Geballe\(^{38}\) that the value of the 2Δ/kT_c increases with decrease of the film thickness for amorphous Nb-films. Therefore, in our fits we have used the estimates of 2Δ/kT_c reported in Ref.\(^{38}\). They increase from 3.9 in thick films up to 4.5 in the thinnest films\(^{28}\).

In the fitting procedure, we first determine the slope dH_{c2}/dT|T_c from linear fits to H_{c2}(T) line in the vicinity of the T_c. Subsequently, the slope is treated as fixed parameter, while Maki parameters α and λ_{so} are treated as adjustable parameters. The values of dH_{c2}/dT|T_c and 2Δ/kT_c used in the fits, together with Maki parameters estimated from the fits, are shown in Table II.
| $d$ (nm) | $dH_{c2}/dT | T_c$ | $2\Delta/kT_c$ | $\alpha$ | $\lambda_{so}$ |
|----------|-----------------|----------------|----------------|----------|----------------|
| 20       | 448 (14)         | 3.9            | 0              | 0        |                |
| 9.5      | 672 (44)         | 3.9            | 0.35 (0.1)     | 0        |                |
| 5.3      | 1114 (42)        | 4              | 1.23 (0.3)     | 0.5 (0.4)|                |
| 3.2      | 2277 (102)       | 4              | 2.4 (0.2)      | 0.7 (0.15)|                |
| 2.2      | 2778 (238)       | 4.1            | 2.6 (0.3)      | 0.3 (0.1)|                |
| 1.4      | 2648 (124)       | 4.5            | 3 (0.1)        | 0.09 (0.01)|                |
| 1.3a     | 2684 (57)        | 4.5            | 2.5 (0.1)      | 0.09 (0.01)|                |

TABLE II: The Maki parameters $\alpha$ and $\lambda_{so}$ estimated from the fit of WHH model to experimental data (the uncertainties are shown in parentheses) for Nb films of thickness $d$. $2\Delta/kT_c$ are estimates from Ref.28.
higher fields, so it cannot be explained by the failure of charge carriers to cool.

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