Thickness-tuned superconductor-insulator transitions under magnetic field in *a*-NbSi

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We have studied the thickness-induced superconductor-to-insulator transition in the presence of a magnetic field for *a*-NbSi thin films. Analyzing the critical behavior of this system within the "dirty boson model," we have found a critical exponent product of $\nu_d z \sim 0.4$. The corresponding phase diagram in the (H,d) plane is inferred. This small exponent product, as well as the nonuniversal value of the critical resistance found at the transition, calls for further investigations in order to thoroughly understand these transitions.

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I. INTRODUCTION

Low-temperature transport in disordered conducting materials implies quantum interferences, Coulomb repulsion, and superconducting fluctuations. Since two dimensions (2D) is the lower critical dimension for the existence of both the superconducting and the metallic states, transport properties of such disordered thin films have attracted continuous attention since the 1960s in order to understand what ground states are allowed in those systems and study the nature of the quantum phase transitions between the different phases.[1–3]

Quantum phase transitions (QPT) occur when a parameter in the Hamiltonian is varied, resulting in a change in the system's ground state. These transitions, therefore, take place at zero temperature and are driven by quantum fluctuations, contrary to classical phase transitions, which are controlled by thermal fluctuations. Near a QPT, the quantum fluctuations have a characteristic length scale—the correlation length ξ—diverging as $\xi \propto \delta^{-\nu}$, where ν is the correlation length critical exponent, $\delta_K = \frac{|K-K_c|}{K_c}$ is the distance of the considered system to the K-driven transition, and K is an experimentally tunable parameter which critical value is K_c. The fluctuations are also characterized by a vanishing frequency $\Omega \propto \xi^{-z}$, where z is the dynamical critical exponent. The two critical exponents ν and z define the universality class to which the transition belongs.

In the case of superconductor-to-insulator transitions (SITs) in disordered thin films, the tunable parameter in the Hamiltonian can be the disorder or the magnetic field H. The most popular theoretical model to explain these SITs is the "dirty boson model" developed by M.P.A. Fisher.[2] In this model, the coherence of the superconducting state is destroyed by quantum fluctuations of the order parameter's phase, and the system amounts to interacting bosons in the presence of disorder. The superconducting and insulating phases are then dual to one another: the superconducting phase consists of localized vortices and condensed Cooper pairs, whereas the insulating phase is characterized by condensed vortices and localized Cooper pairs. Both disorder and magnetic-field-driven transitions have similar description within this frame: in the quantum regime, for dc measurements, the sheet resistance obeys a scaling law that is solely dependent on the variable $\delta * T^{-1/\nu z}$ (Refs. 1 and 2),

$$R(\delta,T) = R_c f(\alpha \delta T^{-1/\nu z}), \tag{1}$$

where R_c is the critical sheet resistance and f is a universal scaling function having a unique constraint: $f(0)=1$. α is a nonuniversal constant.[4] $z=1$ is expected due to the long-range Coulomb interactions and the dirty boson model predicts $\nu > \frac{2}{d} = 1$, as well as a universal value of the system's sheet resistance at the transition $R_c = R_Q = \frac{h}{4e^2} = 6500\ \Omega$.[3] Despite obeying to the same scaling laws [Eq. (1)], the field-induced transition and the disorder-induced transitions have different physical grounds: in the magnetic-field-induced SIT, the vortex density increases with the magnetic field until they delocalize and Bose condense; in the disorder-induced SIT at zero field, the Bose condensation is undergone by the vortex/antivortex pairs. These two SITs, hence, have no reason to have the same critical exponents.[3]

Experimentally, number of disordered superconducting films experiences a SIT when submitted to a perpendicular magnetic field. However, they do not all behave in the same way. Following Gantmakher's[5] comment, one can separate them into two different categories. Some compounds exhibit an insulating phase in which low-temperature resistance is only 10% above their high-temperature resistance. This behavior resembles more the one of a conductor in the presence of weak localization than the one of an actual insulator.[6] This is the case of Mo_xGe_{1-x},[7] Mo_xSi_{1-x},[8] Be,[9] *a*-Bi,[10] or $Nd_{2-x}Ce_xCuO_{4+y}$.[5] Other systems, such as amorphous indium oxyde[11] or TiN,[12] have, in the same conditions, a much more important increase in resistance—up to a factor of 10. Their resistances then have an exponential increase with the temperature.[11,13] The renormalization analysis of these field-induced SIT gives $0.75 \leq \nu_H z \leq 1.35$, independently of the above-mentioned categories.

The experimental realizations of the thickness-induced SIT, where tuning the system's thickness is taken to be a mean of varying its disorder, are far more rare because of the experimental difficulty of synthesizing microscopically identical films, which only differ by their thicknesses. In the case of this transition, the distinction previously made no longer exists: all studied compounds show a drastic increase in resistance of many orders in magnitude when their thickness is lowered.[10] However, one can make another distinction. Some systems, such as *a*-Bi,[10] are very sensitive to any thickness variation: a fraction of angstrom difference engenders resistance increases of several orders of magnitude at low temperature. This behavior is comparable to the one observed in granular systems.[14] On the other hand, systems such as MoC present a more progressive thickness dependence.[15] Values of the critical exponents have only been reported for *a*-Bi (Ref. 10): $\nu_d z \sim 1.3$.

Whichever the parameter tuned to induce the SIT, and contrary to the predictions of the dirty boson model, experiments show an important variation in the values of the critical sheet resistance at the transition R_c.[7–10,12,16] Within one system, R_c can vary between 2000 Ω to 9000 Ω (Ref. 9), depending on the applied magnetic field or the normal resistance of the sample. Theories introducing a fermionic channel of electronic conduction have been developed to explain the nonuniversality of R_c,[7] but these are not entirely satisfactory since they do not account for values of R_c larger than R_Q.[10]

As one can see, all the experimental realizations of the SITs in thin disordered films show a large variation in the measured critical exponents, as well as in the critical resistance. This has led to the questioning of the dirty boson model. Some have suggested a percolation-based mechanism,[17] others the contribution of fermions to the conduction near the transition.[7] Moreover, the flat $R(T)$ curves found near the transition have put into question Fisher's picture of a unique metallic separatrix between the superconducting and insulating regimes. Some[18] have suggested the existence of an intermediate metallic phase—the Bose metal.

In this context, it seemed to us particularly interesting to provide another example of such transition. 2D Nb_xSi_{1-x} films are interesting systems for this study. We have previously shown that these films experience a magnetic-field-tuned SIT (Ref. 19) with a product of critical exponents $\nu_H z = 0.67$, in agreement with other experimental data[20] but in contradiction with the dirty boson model. In this paper, we concentrate on the thickness-driven SIT in this compound. The following sections will be organized as follows: first, Sec. II will detail the experimental procedures. Section III will explain the finite-size scaling method we have used to analyze our results concerning the disorder-induced transition under nonzero magnetic fields, and show that we have obtained surprisingly small critical exponents for the transition. Combining this analysis with our previously obtained results,[19] we infer the phase diagram for Nb_xSi_{1-x} (Sec. IV). Finally, Sec. V will provide a discussion on the interpretation of these sets of experiments on disordered superconducting thin films and on the domain of validity of the dirty boson model.

II. EXPERIMENTAL PROCEDURE

The NbSi films have been prepared under ultrahigh vacuum by electron beam (e-beam) codeposition of Nb and Si. A series of four samples with stoichiometry $Nb_{0.15}Si_{0.85}$ and thicknesses of 100, 50, 25, and 12.5 nm has been deposited onto sapphire substrates coated with a 50-nm-thick SiO underlayer. The films were synthesized during a single run in order to have the samples' niobium concentrations as similar as possible. We also took special care over the control of the sample's parameters: the evaporation was controlled *in situ* by a special set of piezoelectric quartz in order to precisely monitor the composition and the thickness of the deposition. These two characteristics were then controlled *ex situ* by Rutherford back scattering (RBS), and the results were well fitted with the *in situ* monitoring. Samples of the same stoichiometry with thicknesses down to 2.5 nm have been characterized by atomic force microscopy and showed no sign of morphological granularity nor inhomogeneity. The superconducting transitions of these samples in zero magnetic field are a few tens of mK sharp and show no sign of reentrant behavior as usually observed for granular systems. Besides, all samples showed the same resistivity at high temperature within 4%. All these arguments lead us to think that our samples are homogeneous in composition, nongranular, and only differ from one another by their thickness. This conclusion is corroborated by a transmission electron microscopy (TEM) study,[21] showing that only Nb_xSi_{1-x} alloys annealed at 500 °C present Nb-rich clusters. The electrical characteristics of the four films were measured down to 150 mK using a dilution refrigerator. A perpendicular magnetic field could be applied and was made to vary between 5 and 11 kOe. Resistance measurements were performed using a standard ac lock-in detection technique operated at 23 Hz. A current of 100 nA was applied to the sample, which is within the linear regime of the *I*-*V* characteristics for the considered films. All electrical leads were filtered from radio frequency at room temperature.

III. *d*-INDUCED TRANSITION

Before describing the renormalization procedure we have used and the results thus obtained, let us establish the dimensionality of our samples. In our system, the mean free path l is of the order of the interactomic distance: $l \simeq 2.65$ Å (Ref. 22) and, hence, much smaller than the superconducting coherence length ξ_0 given by the Bardeen Cooper Schrieffer theory $[\xi_0 = 0.18\frac{\hbar v_F}{k_B T_{c0}}$, where v_F is the Fermi velocity estimated to be 2×10^8 cm s^{-1} (Ref. 23)]. In the "dirty" limit the effective coherence length of the system is given by $\xi_{\text{eff}} = \sqrt{\xi_0 l}$. We also have to consider the dephasing length, which acts near the SIT as a cutoff length due to the finite temperature:[2,3,11] $L_\Phi = \frac{\hbar^2}{m_e k_B \xi_{eff} T}$, where m_e is the mass of the electron. The smallest length between L_Φ and ξ_{eff}, hence, determines the dimensionality of the film. The different relevant lengths are given in Table I. The films with thicknesses ranging from 12.5 to 50 nm can be considered to be 2D, whereas the 100 nm film is three dimensions (3D). In the renormalization procedure, we shall focus on the 2D films so

TABLE I. Relevant parameters for our samples: the thickness d, the superconducting transition temperature T_{c0}, the BCS coherence length ξ_0, the effective coherence length $\xi_{\rm eff}$, and the dephasing length L_Φ at 0.3 K.

d (nm)	T_{c0} (mK)	ξ_0 (μm)	$\xi_{\rm eff}$ (nm)	L_Φ(0.3 K) (nm)
12.5	213	12.8	58.2	50
25	347	7.9	45.7	64
50	480	5.7	38.9	75
100	530	5.2	37.1	79

that the resistances mentioned below are sheet resistances. Let us also note that, in what follows, we used the usual convention found in the SIT-related literature:[24] the term "superconducting" applies to curves that have a positive temperature coefficient of resistance (TCR: $\frac{dR}{dT}$), and, by contrast, we shall label as "insulating" all curves having a negative TCR.

As shown by the rarity of experimental data concerning the thickness-induced SIT, it is difficult to obtain a series of samples that are identical except for their thickness: unlike the magnetic field, d cannot be tuned continuously. We have, therefore, developed an analysis method, which enables us to interpolate the system's transport behavior between the discrete values of d we experimentally have access to.

All four samples were superconducting at zero magnetic field (inset of Fig. 1) and were progressively tuned through the transition by a finite H. For each value of H, all four samples were studied (Fig. 1), and the diagram (R,d) traced for different temperatures presents a crossing point (Fig. 2). This is the signature of the QPT (Ref. 10) and allows us to estimate the critical thickness d_c associated to the magnetic field H. We repeat this process for all values of H, obtaining a collection of critical parameters couples (d_c,H).

When H is fixed, the thickness-induced transition is solely governed by the distance to the transition $\delta_d=\frac{|d-d_c|}{d_c}$. If these

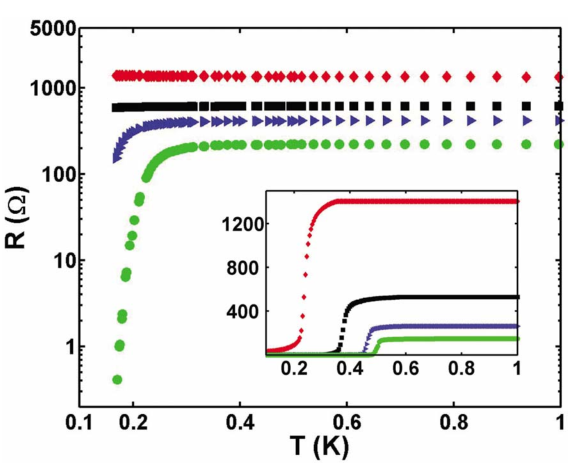

FIG. 1. (Color online) Resistance per square as function of temperature for $H=6.8$ kOe. The curves for all four samples are represented. For this particular value of the magnetic field, the 25-, 50- and 100-nm-thick films are superconducting, whereas the 12.5-nm-thick film is insulating. Inset: The same data at zero magnetic field.

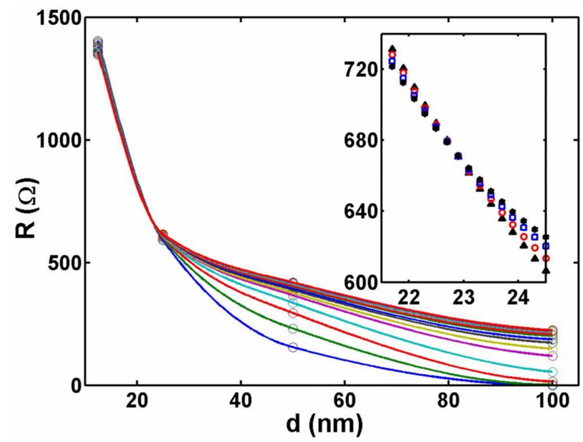

FIG. 2. (Color online) Resistance per square as function of sample thickness for $H=6.8$ kOe. The curves are represented for 16 different values of the temperature between 168 and 831 mK. Inset: the same data are shown around the crossing point at about d_c $=23$ nm for four particular temperatures: $T=168$, 186, 239, and 831 mK. This crossing point is interpreted as the signature of a QPT.

d-driven transitions all belong to the same universality class, independent of the particular value of H, the only relevant parameter for the scaling of all our data is the value of δ_d $=\frac{|d-d_c(H)|}{d_c(H)}$. This means that *all* curves $R(\frac{|d-d_c(H)|}{d_c(H)},T)$ should collapse on two universal curves. Note that the renormalized quantity we consider is R and not $\frac{R}{R_c}$ as in Ref. 10, for we do not find a universal critical resistance.[4] For each individual sample, this means that by tuning H, d_c is made to vary and so does δ_d. In other words, the thickness d being fixed, the critical thickness d_c is changed via the magnetic field. Since the only relevant parameter for the scaling is the distance δ_d to the transition, this situation is ultimately equivalent to having a fixed critical thickness and variable sample thicknesses (as in Ref. 10 for example).

For each sample, the results were analyzed using two independent scaling methods.[7,10] First, for the derivative method, we plot $\frac{DR}{D\delta_d}|_{\delta_d=0}\propto R_c T^{-\frac{1}{\nu_d z}} f'(0)$ as function of $\frac{1}{T}$, which, in a log-log diagram, gives a straight line of slope $\frac{1}{\nu_d z}$ (left insert Fig. 3). The second method consists in numerically finding $t(T)$ such that $R[\delta_d,t(T)]=R_c f[\delta_d t(T)]$ and that $t(T)$ yields the best collapse between the data measured at the temperature T and the data measured at our lowest temperature (150 mK). To obey the scaling law [Eq. (1)], $t(T)$ should be of the form $T^{-1/\nu_d z}$, and we can, hence, infer the value of $\nu_d z$ (right insert Fig. 3).

For all 2D samples, we obtained a product of critical exponents of $\nu_d z=0.4\pm0.15$;. We can check this value of the exponent product by plotting R as function of $\delta_d * T^{-1/\nu_d z}$ (Fig. 3) for the 25-nm-thick sample. All data superimpose nicely in the ranges $0.16\leq T\leq0.35$ K and $|\delta_d|\leq1$, forming two curves only: one representing the superconducting behavior and the other the insulating side of the transition. $|\delta_d|=1$ still exhibits a critical behavior since the corresponding data collapse on the same curves. It is quite surprising that the scaling continues to work that far from the critical

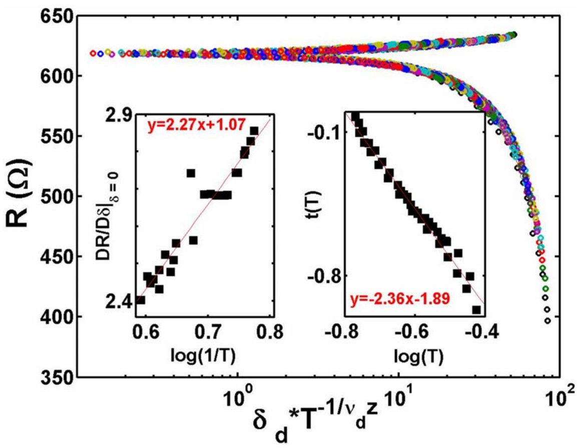

FIG. 3. (Color online) Renormalization of the resistance R for the critical exponents $\nu_d z=0.4$ for the 25-nm-thick sample. Each color is affected to a particular value of δ_d. Left inset: determination of the critical exponent product by the derivation method. Right inset: determination of the critical exponent product by the $t(T)$ minimization method.

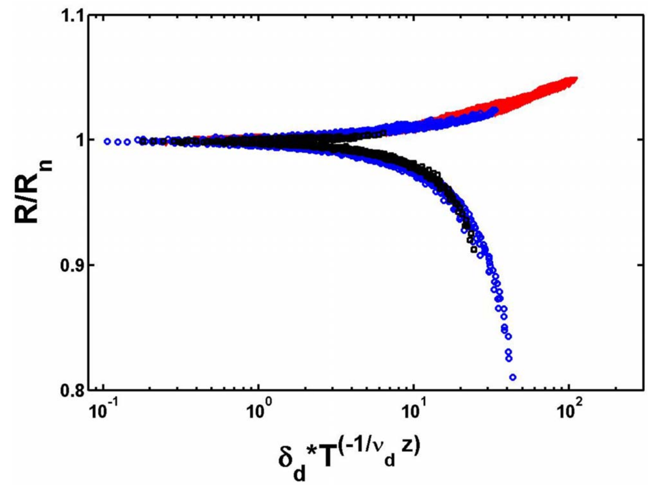

FIG. 4. (Color online) Renormalization of the renormalized resistance $\frac{R}{R_n}$ for the critical exponents $\nu_d z=0.4$ for the 12.5- (triangles), 25- (circles), and 50- (squares) nm-thick samples.

point. The analysis performed on the 12.5-nm- and the 50-nm-thick samples gave the same value of the product $\nu_d z$ within the uncertainty.

This far, we have only considered the renormalization of the resistance for one sample at a time. In order to compare the critical behavior of the different samples, we have to take into account their different normal resistances. We, therefore, have to compare the quantity $\frac{R}{R_n}$, where R_n is the resistance taken at high temperature, typically at 1K. This procedure is not usual in the literature and directly derives from the fact that, in our experiment, R_c is not universal and varies over one order of magnitude (see Sec. V). The scaling of $\frac{R}{R_c}$ then has no significance.[4]

We then looked for a critical exponent product that allowed *all* curves from *all* samples to collapse. For each sample, we adjusted the nonuniversal parameter α of Eq. (1) for the curves to superimpose. We found $\alpha_{12.5\text{ nm}}=1.9$, $\alpha_{25\text{ nm}}=0.9$, and $\alpha_{50\text{nm}}=0.5$ for a product of $\nu_d z=0.4\pm0.1$. The corresponding criteria for the renormalization are then very clearly defined: (i) the magnetic field was made to vary between 5.1 and 10.5 kOe by increments of 0.1 kOe; all critical points (d_c, H_c) corresponding to these fields have been taken into account; (ii) the only constraint on the distance to the transition is $\delta<0.8$; (iii) $0.17<T<0.39$ K. The result of the renormalization is given in Fig. 4. This graph is particularly remarkable: even if our samples have normal resistances varying by nearly one order of magnitude, the corresponding resistances all collapse on a single renormalization plot.

IV. PHASE DIAGRAM

The renormalization method has enabled us to measure a number of critical parameter couples (H_c, d_c), although we only had four different samples. We can, hence, draw part of the phase diagram for Nb_xSi_{1-x} thin films (Fig. 5). The line formed by the critical points separates an insulating region at high fields, and small thicknesses form a superconducting region at low field and large thicknesses. Of course, these critical points coincide with those determined from the magnetic-field-induced SIT.[19] As for *a*-Bi,[10] depending on the parameter tuned to cross this line, the critical exponent product found is different: $\nu_H z=0.7$ when the field is varied, whereas a variation in the sample's thickness gives $\nu_d z=0.4$. We, thus, confirm that these two SITs belong to two separate universality classes.

V. DISCUSSION

First let us comment on the value found for the critical exponent product. For *a*-NbSi in a thickness-induced SIT, we have found $\nu_d z=0.4$. This value is surprising when compared

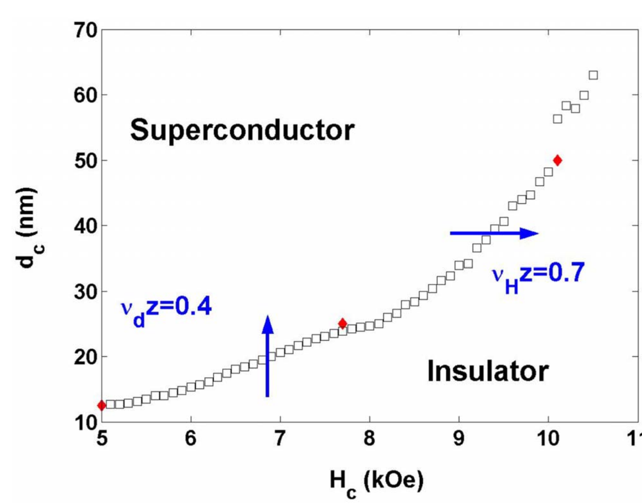

FIG. 5. (Color online) Phase diagram for a-$Nb_{15}Si_{85}$ in the (H_c, d_c) plane. The open symbols were obtained from the thickness-tuned SIT, whereas the full symbols were obtained in Ref. 19 for the magnetic-field-tuned transition.

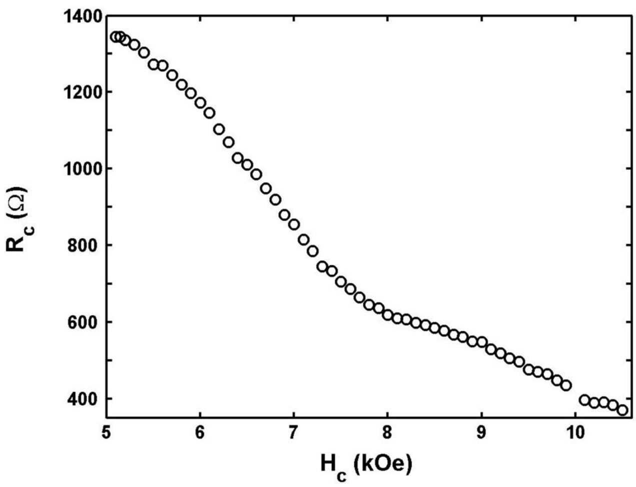

FIG. 6. Critical resistance as function of the critical field for *a*-NbSi films.

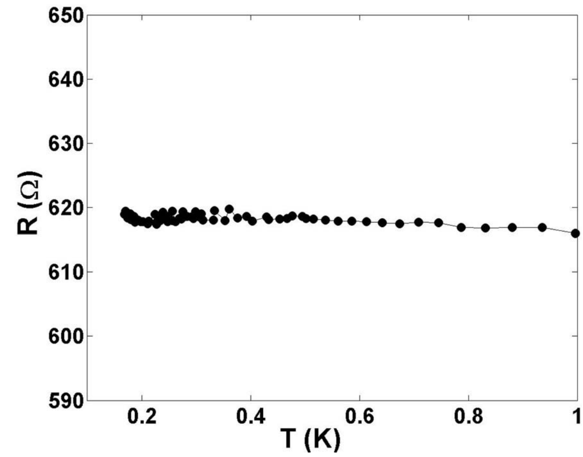

FIG. 7. Resistance as function of the temperature for the 50-nm-thick sample at $H=7.9$ kOe. Over one decade variation in temperature, the film's resistance only varies within 3.5 Ω (0.5% in relative value), which is our experimental uncertainty in this range.

to other critical exponents found by other groups, thin *a*-Bi films for instance, for which $\nu_d z=1.4$. At this point, we do not have any clear explanation for this important difference. However, $\nu_d z=1.4$ is close to what is predicted for classical 2D percolative systems ($\nu_d=4/3$), and *a*-Bi thin films present a thickness-induced SIT for very shallow thicknesses (a few angstroms, 20 Å at most). In this sense also, our system is particularly interesting since it allows 2D samples to experience a thickness-driven SIT at reasonable thicknesses, where the roughness of the film, the surface state of the substrate, or the microscopic details of the film's growth should not be important factors. $\nu_d z=0.4$ is also surprisingly small considering the theoretical predictions that have been made to this day.[1] Although the exact value of this product might be affected by the uncertainty on the determination of the exponents (± 0.1) and by the small number of samples we have, at any rate, we can confidently say that $\nu_d z<1$, which is inconsistent with the dirty boson model. If we assume that $z=1$, the consequence of this is that $\nu_d<1$. Many authors have pointed to the fact that this violates the so-called "Harris criterion" ($\nu>2/d$).[25] However, this criterion is valid for small disorder, and since our system consists in amorphous films in which the mean free path is of the order of the interatomic distance, it is not all that shocking that the value found for the localization length exponent does not obey this inequality.[26]

Another point that has much been discussed related to the dirty boson model is the value of the critical sheet resistance. In this set of experiments, we show that R_c varies over a large range when either the magnetic field or the thickness is varied (Fig. 6).

Until now, we have analyzed our results by comparing them to the dirty boson model. Although the renormalization procedure works remarkably well for all systems studied to date—which means the SIT is indeed a QPT (Ref. 1)—two important predictions of this model ($\nu>2/d$ and $R_c=h/4e^2$) are not verified by *a*-NbSi thin films, as well as in other systems [*a*-Bi,[10] *a*-Be,[9] NdCeCuO,[27] MoGe,[28] InOx,[13,16] TiN,[12] and MoSi (Ref. 8)]. One might, therefore, put this model into question. Tunneling effect experiments suggest[29–31] that, for homogeneous systems, amplitude fluctuations of the order parameter play a role even in the vicinity of the SIT: when the films' thicknesses decrease, the superconducting gap Δ and the critical temperature decrease together, monotonically, such that $2\Delta/T_c \simeq$ constant. In this picture, near the SIT, the amplitude of the superconducting order parameter can become very small, whereas an essential point in the dirty boson model is that its amplitude is finite near the transition. The same studies show that, even in the "superconducting"—in the previously-defined sense of the TCR—region, the one-particle density of state is not zero, meaning that there are normal excitations coming from electrons that are not involved in any Cooper pair. This would mean that amplitude fluctuations of the system must be taken into account for a correct description of the transition, which is not the case in M.P.A. Fisher's model. The suggestion by some authors that other phase(s) may be involved in between the superconducting and the insulating regimes is particularly interesting. Some have suggested a vortex-liquid phase,[32,33] which has recently[34] been linked to the problem of anomalous Nernst effect in the cuprates. As recent measurements on amorphous superconductors have shown,[35–37] Nernst effect is a very sensitive probe of amplitude fluctuations[35,36] and phase fluctuations[37] of superconducting order parameter. These last works suggest that measurements of the Nernst effect should be a relevant probe to test the existence of this vortex-liquid phase. However, there have not been clear predictions on how the thickness variation should affect this phase. Also very appealing is the suggestion that there is a bosonic metallic phase, such as the Bose metal,[18] involved. This hypothesis is very interesting, in particular when one takes a close look at the resistive behavior of our films. Indeed, at low temperatures, the resistance of some samples seem to saturate at a finite value (Fig. 7), displaying a large temperature range where the resistance is independent of the temperature. However, a study at lower temperatures should be undertaken to confirm this tendency. Let us restate that the qualification of insulating or superconducting have been arbitrarily attributed to $\frac{\partial R}{\partial T}<0$ (respectively, $\frac{\partial R}{\partial T}>0$) curves without any other ground than the assumption made by the dirty boson model that only these two phases existed. All these arguments (the amplitude fluctuations of the order parameter, a possible fermionic channel, and the suggestion of a Bose metal) plead in favor of a reconsideration of the dirty boson model and further experimental investigations of these systems.

In conclusion, we have studied the thickness-induced SIT in the presence of a perpendicular magnetic field on a-$Nb_{15}Si_{85}$ thin films of thicknesses ranging from 12.5 to 100 nm. We have found the signature of a QPT when the sample thickness is lowered. The corresponding critical exponent product is $\nu_d z \simeq 0.4 \pm 0.1$. This value is different from the one found in the analysis of the magnetic-field-induced transition in the same compound for which $\nu_H z = 0.65$. These two SITs, therefore, belong to two different universality classes. However, the very small value of $\nu_d z$ cannot be explained by the existing models for this transition. Further experimental investigations are needed to understand the growing discrepancies between the various experimental results and between these results and the theory.

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