Thickness dependence of the superconductivity in thin disordered NbSi films

O Crauste, C A Marrache–Kikuchi, L Bergé, D Stanescu and L Dumoulin
C.S.N.S.M, CNRS-IN2P3, UMR 8609, Paris XI University, Orsay, France
E-mail: Olivier.Crauste@csnsm.in2p3.fr

Abstract. Superconducting a-Nb$_x$Si$_{1-x}$ thin films experience a lowering of the $T_c$ until the superconductivity disappears through a Superconductor–Insulator Transition (SIT). We here present transport measurements on 2D a-Nb$_x$Si$_{1-x}$ films, close to the SIT, for different compositions and thicknesses. We investigate the lowering of the $T_c$ in light of existing theories, especially the one developed by Finkel’stein.

1. Introduction
Since the pioneering work of Hebard et al. on InO$_x$ [1], it has been decades now that transport properties in disordered supraconducting thin films have aroused great excitation. These 2D systems, where the thickness $d_\perp$ is lower than the superconducting coherence length $\xi$, exhibit long studied Superconductor–Insulator Transitions (SIT) which can be induced by various external parameters such as the magnetic field, the disorder, etc. [2]. These SITs have been described by M.P.A. Fisher [3] and the "Dirty Boson Model" which analyses the transition in terms of Quantum Phase Transition.

When the thickness of such superconducting thin films is reduced, the superconductivity is progressively destroyed before the film undergoes a disorder–induced SIT and hence becomes insulating. M.P.A. Fisher’s model well describes this transition but does not give a clear physical interpretation for the initial $T_c$ reduction.

Superconducting a-Nb$_x$Si$_{1-x}$ thin films have been shown to provide an interesting example of system undergoing a SIT [4]. In this paper, we focus on the understanding of the $T_c$ reduction in this system before it turns to insulating.

2. Experimental Setup
The amorphous NbSi thin films have been prepared under ultrahigh vacuum by e-beam co-deposition of Nb and Si. Different series of four samples have been deposited onto sapphire substrates coated with a 50 nm-thick SiO underlayer. The stoichiometry and the thicknesses of the various samples are given in Table 1. The four 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 piezo-electric quartz in order to precisely monitor the composition and the thickness of the deposition. These two characteristics were then controlled $ex$ $situ$ by Rutherford
Table 1. Description of Nb$_x$Si$_{1-x}$ thin films: composition $x$, name, thickness $d_\perp$, and superconducting transition temperature $T_{cb}$ for bulk samples of the same stoichiometry.

| Composition $x$ | Name | $d_\perp$ [Å] | $T_{cb}$ [K] |
|----------------|------|----------------|--------------|
| $x = 14\%$    | OC02 | 75, 125, 175, 500 | 0.28         |
|                | OC03 | 90, 105, 150, 175 |              |
| $x = 15\%$    | CKJ1 | 125, 250, 500, 1000 | 0.59         |
|                | CKJ2 | 25, 50, 75, 125 |              |
| $x = 17\%$    | CKJ3 | 40, 50, 75, 125 | 0.8          |

Table 2. Results obtained from least square fit of Eq 1.

| Composition $x$ | $\tau$ [s] |
|----------------|------------|
| $x = 14\%$    | $1.2 \times 10^{-13}$ |
| $x = 15\%$    | $0.8 \times 10^{-13}$ |
| $x = 17\%$    | $1.4 \times 10^{-13}$ |

Figure 1. Resistance for $x = 15\%$. Our samples are assumed to be homogeneously disordered: there is no reentrant behavior, the transition is sharp and the onset of the superconductivity decreases with the thickness.

Figure 2. Decrease of $T_c$ vs $R_{\square}$. The symbols represent the experimental data. The solid lines are the least square fit of Eq 1. The dashed curves are obtained with $\tau = 10^{-13}$ s for the different compositions.

Back Scattering and the results well fitted with the in situ monitoring. Each samples’ edges were etched by Reactive Ion Etching in order to suppress any edge effect that might affect the transport measurements.

We then measured the electrical transport properties of the samples with a dedicated dilution fridge down to 7 mK. All leads were filtered from RF at room temperature. Transport measurements were done with a TRMC2 measuring bridge. We double-checked our measurements by performing AC lock-in detection measurements. Moreover, in all cases, special care was taken to ensure we were not overheating the films.

Our samples are believed to be homogeneous as AFM and SEM measurements show no morphological granularity. Temperature dependance of the sheet resistivity shows sharp transitions (Figure 1) and no reentrant behavior as it is the case for granular systems [5]. Another argument for the samples’ homogeneity is that the onset temperature of the superconductivity decreases as the film thickness is reduced.
3. Theoretical background
The influence of the disorder on the superconductivity has been a long-standing problem. For weakly disordered systems, \( k_F l \gg 1 \) where \( k_F \) is the Fermi wave vector and \( l \) the elastic mean free path, Anderson’s theorem claims that superconductivity is not affected by non-magnetic impurities. However, as the disorder increases, this theorem does not hold: the elastic scattering caused by the disorder reduces the dynamical charge screening and thus enhance the Coulomb interaction. This induces a \( T_c \) reduction as Maekawa and Fukuyama [6] have explained within a linear approximation in the first order of \( k_F l \). Finkel’stein [7] then completed this theory and described the whole reduction of \( T_c \) down to very disordered system (\( k_F l \approx 1 \)). He derives the following equation:

\[
\frac{T_c}{T_{c0}} = \exp \left( -\frac{1}{\gamma} \left[ \left( 1 + \frac{\sqrt{r/4}}{\gamma - r/4} \right) \times \left( 1 - \frac{\sqrt{r/4}}{\gamma - r/4} \right) \right]^{-1} \right)^{1/\sqrt{2r}}
\]

where \( \gamma = 1/\ln \left( \frac{k_B T_{c0}}{2\pi e} \right), \quad r = (\pi^2/2\gamma)R_{\square} \) [8], \( R_{\square} \) is the normal sheet resistance, and \( T_{c0} \) is the BCS superconductivity transition temperature for the bulk material and \( \tau \) the elastic mean free path time. This theory was applied on different systems: MoGe [7], Ta [8], TiN [9].

4. Thickness dependence of \( T_c \)
Since a-NbSi thin films are homogeneous and highly disordered (\( k_F l \approx 1 \)), we have tried to see whether Eq 1 could account for the \( T_c \) reduction we observe. We measured \( R_{\square} \) and \( T_c \), \( T_{c0} \) was taken to be \( T_{c0} \), the superconducting transition temperature measured for thicker samples, and \( \tau \) was the fitting parameter. The mean free path times obtained by the least square fit of our experimental data are given in Table 2. The fits and the experimental data are shown on Figure 2. Since all \( \tau \) have a value of roughly \( 10^{-13} \) s, we tried to perform a fit with a fixed \( \tau \). The results is shown on Fig. 2 (dashed lines).

Let us compare the \( \tau \) obtained by fitting the experimental data with Eq. 1 with what we can reasonably expect in this material. In our systems, the inter-atomic distance was measured to be of about 2.6 Å [10]. Therefore the mean free path can be expected to lie between 2.6 and 10 Å. Moreover specific heat measurements on NbSi films have given \( c = 1.5 \times 10^{-5} \) J/K.cm\(^3\) [11] which is a value comparable to that of gold. Taking into account that the effective mass might not be the same in Au and in NbSi, we have an acceptable range of Fermi velocity between \( 10^7 \) and \( 2.10^7 \) cm.s\(^{-1}\). This leads to an acceptable range for the mean-free-path times between \( 10^{-16} \) and \( 10^{-14} \) s. The \( \tau \) obtained from the fit is one order of magnitude above this acceptable range. Eq. 1 therefore does not describe satisfiingly our data.

However, Finkel’stein mentions a correction that has to be made for very disordered films. Indeed, for \( l < d_\perp \), the mean free path time \( \tau \) in Eq. 1 should be replaced by \( \frac{d_\perp^2}{2} \tau \). One must note that this introduces an additional dependance on \( l \) which value is not precisely known in our films. In order to fit our data, we must therefore make an hypothesis on the value of \( l \) and adjust \( \tau \) and \( T_{c0} \). We performed the fits for two values of \( l \) (\( l=2.65 \) Å and \( l=5 \) Å). The results are summarized in Figure 4 and the adjustment plotted together with the experimental data on Figure 3. Whatever the precise value of \( l \), we now obtain mean free path times that are plausible. Both \( \tau \) and \( T_{c0} \) are found to be not significantly dependant on \( l \). However, the \( T_{c0} \) inferred from this fit are systematically lower than the \( T_{c0} \) measured in bulk samples.

5. Conclusion
Our samples’ behavior is qualitatively described by Finkel’stein theory provided that we take into account the correction necessary for very disordered thin films (\( d_\perp > l \)). This is in agreement
Figure 3. Decrease of $T_c$ vs $R_{\Box}$. The solid lines are the least square fit of Eq 1 with the correction $\tau \rightarrow d^2/\tau_l$. The fits performed for $l = 2.6\, \text{Å}$ and $l = 5\, \text{Å}$ are indistinguishable.

with the fact that a-NbSi is a strongly disordered system: for bulk $x = 14\%$ samples, the resistivity $\rho \simeq 1900\, \mu\Omega\, \text{cm} \pm 100$, which is 10 times larger than the bulk resistivity in MoGe [7][12] ($\rho \simeq 160\mu\Omega\, \text{cm}$). Quantitatively, however, the inferred mean free path time corresponds to what is expected for these films, but the $T_{c0}$ do not match the superconducting temperatures measured for 3D samples. Equivalently, this means that the theory doesn’t account for thick samples. A possible explanation for this is that these thick samples approach the vicinity of the 2D-3D crossover which is the limit for Finkel’stein’s theory (for $x=15\%$, $T_{cb}=0.59\, \text{K}$, $\xi_{\text{eff}} = \sqrt{\hbar v_F l/k_B T_c} = 409\, \text{Å}$ with $l=2.6\, \text{Å}$ and $v_F=5 \times 10^5\, \text{m.s}^{-1}$).

References

[1] Hebard A F and Paalanen M A 1984 Phys. Rev. B 30 4063
[2] Sondhi S L, Girvin S M, Carini J P and Shahar D 1997 Rev. Mod. Phys. 69 315
[3] Fisher M P A 1990 Phys. Rev. Letters 65 923
[4] Aubin H, Marrache-Kikuchi C A, Pourret A, Behnia K, Bergé L, Dumoulin L and Lesueur J 2006 Phys. Rev. B 73 094521
[5] Haviland D B, Liu Y and Goldman A M 1989 Phys. Rev. Lett. 62 2180
[6] Maekawa S and Fukuyama H 1983 J. Phys. Soc. Jpn. 52 1352
[7] Finkel’stein A M 1994 Physica B 197 636
[8] Astrakharchik E G and Adkins C J 1994 Phys. Rev. B 50 13622
[9] Hadacek N, Sanquer M and Villégier J C 2004 Phys. Rev. B 69 024505
[10] Hucknall P K, Walker C G H, Greig D, Matthew J A D, Norman D and Turton J 2004 Surface and Interface Analysis 19 23
[11] Marnieros S, Bergé L, Juillard A and Dumoulin L 1999 Physica B 261 862
[12] Graybeal J M and Beasley M R 1984 Phys. Rev. B 29 4167

| Composition | $\tau(l = 2.65\, \text{Å})$ | $\tau(l = 5.0\, \text{Å})$ | $T_{c0}$   |
|-------------|-----------------------------|-----------------------------|------------|
| $x = 14\%$  | $0.9 \times 10^{-16}\, \text{s}$ | $3.4 \times 10^{-16}\, \text{s}$ | $0.21\, \text{K}$ |
| $x = 15\%$  | $1.8 \times 10^{-16}\, \text{s}$ | $6.5 \times 10^{-16}\, \text{s}$ | $0.43\, \text{K}$ |
| $x = 17\%$  | $8.6 \times 10^{-16}\, \text{s}$ | $31 \times 10^{-16}\, \text{s}$ | $0.56\, \text{K}$ |

Figure 4. Results obtained from least square fit of Eq 1 with the correction $\tau \rightarrow d^2/\tau_l$. Two different values of $l$ are taken to perform the fit. $T_{c0}$ is now also a fitting parameter.