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# Electron phase-breaking time in ultra-thin Nb films

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Abstract. Here we study the temperature dependences of the electron phase-breaking time  $\tau_{\phi}$  in ultra-thin superconducting niobium (Nb) films. In Nb films, passivated with a layer of silicon (Si), the observed temperature dependence of the phase-breaking time is  $\tau_{\phi} \sim T^{2.5}$ , is resembling the electron-phonon scattering. However, in the uncovered Nb films, we observe the saturation of  $\tau_{\phi}$  at low temperatures, which may be a signature of the surface magnetic disorder, present in native Nb oxide on the film surface.

Keywords: magnetoresistance, thin films, inelastic scattering, magnetic disorder

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Материалы конференции УДК 537.9 DOI: https://doi.org/10.18721/JPM.153.312

# Время сбоя фазы в ультратонких пленках Nb

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Аннотация. В данной работе мы представляем результаты экспериментального исследования времени сбоя фазы волновой функции электрона в ультратонких сверхпроводящих пленках ниобия (Nb). В Nb пленках, пассированных слоем кремния (Si), наблюдается сильная зависимость времени сбоя фазы от температуры  $\tau_{\phi} \sim T^{2.5}$ , вероятно, обусловленная электрон-фононным рассеянием. Однако в непокрытых кремнием Nb пленках наблюдается насыщение времени сбоя фазы при низких температурах, что может быть обусловлено наличием поверхностного магнитного беспорядка, возникающий в естественном окисле Nb на поверхности пленки.

**Ключевые слова:** магнетосопротивление, тонкие пленки, неупругое рассеяние, магнитный беспорядок

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### Introduction

Magnetic disorder, potentially present in the native oxides on the surface of thin superconducting films, can crucially suppress superconductivity due to breaking of the time-reversal symmetry in superconductors [1, 2]. However, distinction this mechanism from other mechanisms of superconductivity suppression is not straightforward. For example, it is well known that the native oxide of Nb is conductive and, hence, it can diminish superconducting properties due to the inverse proximity effect [3]. Meanwhile some experimental observations show signatures of magnetic disorder on the surface of Nb [4–5], one can to obtain additional information with magnetoresistance transport measurements. In particular, one can expect the saturation of phase-breaking time of the electron wavefunction in case of scattering on magnetic disorder [6–7]. In this work we investigate the dependence of phase-breaking time upon temperature by measuring magnetoresistance in ultrathin superconducting Nb films. We observed a strong dependence of the phase-breaking time on temperature in Nb films, passivated with a layer of silicon (Si). Meanwhile, in uncovered Nb films, we observe the saturation of the electron phase-breaking time at low temperatures.

# **Materials and Methods**

Ultrathin Nb films are sputtered using the magnetron sputtering system (AJA International Inc.) with a background pressure of  $9 \times 10^{-8}$  torr. The samples are deposited on r-cut Al<sub>2</sub>O<sub>3</sub> and Si substrates by sputtering of the Nb target with diameter of 50.8 mm and purity of 99.95% in argon atmosphere (99.998% purity). The working pressure is 3.1 mTorr. During deposition the substrates are heated up to  $T_{dep} = 400$  °C. This heating is controlled with a built-in PID controller, and  $T_{dep}$  is pre-calibrated using an analog thermometer (PTC Instruments). The rotation of substrates during deposition and the relatively large distance between substrates and the target (~ 10 cm) allows for fabrication on Nb films with high uniformity. The acquisition of thin Nb films is controlled by piezoelectric microweighting in the test process. The film growth rate is 0.11 nm/s, and the thickness is determined by the time of film deposition. The films investigated have thicknesses in the range of 3–6 nm. To prevent unintentional oxidation of Nb films in the atmosphere, two films are passivated with a 5-nm thick silicon (Si) layer. In this study, we prepare two passivated Nb samples (A1, A2) and two uncovered samples (B1, B2). In the latter case, the films are exposed to strong unintentional oxidation [8].

To study transport properties, we patterned the films into 500-µm wide and 1000-µm long Hall-bars. Electrical transport measurements are carried out with a 370 AC LakeShore resistance bridge at a bias current less than 1 µA. Normal-state resistance Rs is measured in a four-probe configuration. The measurements are carried out on a custom <sup>4</sup>He cryogenic insert immersed in a dewar in a wide temperature range (from 300 K to 1.7 K). At low temperatures we measure the magnetoresistance  $R_s(B)$ , the temperature dependencies of the second critical magnetic field  $B_{c2}(T)$  and the Hall resistance  $R_H$  at 25 K by applying perpendicular magnetic field B up

to 4 T. The latter allows to determine the carrier density  $n = B/(edR_H)$ . We determine the slope  $dB_{c2}/dT$  at  $T_c$  by measuring R(T)-dependencies on different values of B (not shown here). The latter allows to estimate the critical magnetic field  $B_{c2}(0)$ , the electron diffusivity D using the following expressions  $B_{c2}(0) = -0.69T_c(dB_{c2}/dT)$ ,  $D = -4k_B/(\pi e) (dB_{c2}/dT)^{-1}$ .

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For samples B1 and B2 we estimate the diffusion D coefficient by extrapolation from data for thicker films (not presented here). Here, the critical temperature  $T_c$  is determined as the temperature at  $R_s = R_s^{10K} / 2$ . The parameters of the studied samples are presented in Table 1.

Table 1

	With a Si layer		Without a Si layer	
	Al	A2	B1	B2
d, nm	6	3	5	3
Substrate	Al <sub>2</sub> O <sub>3</sub>	Al <sub>2</sub> O <sub>3</sub>	Al <sub>2</sub> O <sub>3</sub>	Si
$T_{c}, \mathbf{K}$	7.53	3.26	1.10	0.92
$R_{s}^{10K}$ , Ohm	20.0	137.3	186.4	363.8
$R_s^{300\mathrm{K}}$ , Ohm	48.57	181.9	186.8	363.8
$D, \text{ cm}^2/\text{s}$	3.56	1.59	1.3	1.2
<i>n</i> , cm <sup>-3</sup>	4.85×10 <sup>22</sup>	4.06×10 <sup>22</sup>	3.03×10 <sup>22</sup>	3.25×10 <sup>22</sup>
$B_{c2}(0), T$	2.32	2.25	0.93	0.84
$\tau_{0}$ , fs	14.3	6.4	5.2	4.8
$\alpha_{_{e-e}}^{^{-1}}$ , ps	203	97	227	165
$\alpha_{e-ph}^{-1}$ , ps	7.5	20	100	20
р	2.5	1.5	0	0
$\tau_{s}$ , ps	inf	10	2.4	6.7

#### Characteristics of niobium films

## Theory

To determine the electron phase-breaking time  $\tau_{\phi}$  in thin films, one should experimentally study contribution of quantum corrections to magnetoconductance [9]. The dimensionless change in magnetoconductance at a fixed temperature T can be determined from the measured sheet resistance  $R_s(B, T)$  using the expression:

$$\delta G(B,T) = \frac{2\pi^2 \hbar}{e^2} [R_s(B,T)^{-1} - R_s(0,T)^{-1}].$$
<sup>(1)</sup>

There are different contributions to the magnetoconductance: weak localization [10] and superconducting fluctuations (Maki-Thompson (MT), Aslamazov-Larkin (AL), and renormalization of density of states (DOS) contributions) [9]. Since the superconducting fluctuations are stronger than the weak localization in our samples, we refer only to the contribution of the superconducting fluctuations:

$$R_{s}(B,T)^{-1} = \sigma(B,T) = \sigma^{AL}(B,T) + \sigma^{DOS}(B,T) + \sigma^{MT}(B,T) + \sigma^{WL}(B,T),$$
(2)

where

$$\begin{aligned} \sigma^{AL}(B,T) &= \frac{\pi^2 \varepsilon}{4h^2} \bigg[ \psi \bigg( \frac{1}{2} + \frac{\varepsilon}{2h} \bigg) - \psi \bigg( 1 + \frac{\varepsilon}{2h} \bigg) + \frac{\varepsilon}{h} \bigg], \\ \sigma^{DOS}(B,T) &= -\frac{28\zeta(3)}{\pi^2} \bigg[ \ln \bigg( \frac{1}{2h} \bigg) - \psi \bigg( \frac{1}{2} + \frac{\varepsilon}{2h} \bigg) \bigg], \\ \sigma^{MT}(B,T,\tau_{\phi}) &= -\beta_{MT}(T,\tau_{\phi}) \bigg[ \psi \bigg( \frac{1}{2} + \frac{B_{\phi}}{B} \bigg) - \psi \bigg( \frac{1}{2} + \frac{B_{\phi}}{B} \frac{\varepsilon}{\gamma_{\phi}} \bigg) \bigg], \\ \sigma^{WL}(B,T) &= \frac{3}{2} \psi \bigg( \frac{1}{2} + \frac{B_2}{B} \bigg) - \psi \bigg( \frac{1}{2} + \frac{B_2}{B} \bigg) - \frac{1}{2} \psi \bigg( \frac{1}{2} + \frac{B_3}{B} \bigg). \end{aligned}$$

Here  $\psi(x)$  is the Digamma function,  $\varepsilon = \ln(T/T_c)$  and  $h = 0.69B/B_{c2}(0)$  are the reduced temperature and magnetic field, respectively,  $\gamma_{\phi} = \pi \hbar / (8k_B T \tau_{\phi})$  is the phase-breaking parameter 66

with  $\tau_{\phi}$ , which is used as a fitting parameter here. The characteristic fields are defined as  $B_1 = B_0 + B_{so}$ ,  $B_2 = B_{\phi} + 4B_{so}/3 + 2B_s/3$ ,  $B_3 = B_{\phi} + 2B_s$ ,  $B_{so} = \hbar/4eD\tau_{so}$ ,  $B_o = \hbar/4eD\tau$ . The coefficient in MT term  $\beta_{MT}(T, \tau_{\phi})$  can be found in [11].

In order to analyze the dependence of the phase breaking time upon temperature, it is useful to represent  $\tau_{\phi}$  as the sum of four different contributions, namely the scattering on superconducting fluctuations, the electron-electron scattering, the electron-phonon scattering and the scattering on magnetic moments:

$$\tau_{\phi}^{(-1)}(T) = \tau_{SC}^{-1} + \tau_{e-e}^{-1} + \tau_{e-ph}^{-1} + \tau_{s}^{-1}.$$
(3)

While magnetic scattering is independent of temperature, other terms are dependent on T in the following way:

$$\tau_{SC}^{-1} = \frac{\pi g k_B T}{\hbar} \frac{2 \ln 2}{\varepsilon + \beta},$$
  
$$\tau_{e-e}^{-1} = \frac{\pi g k_B T}{\hbar} \ln \frac{1}{2\pi g} = \alpha_{e-e} \frac{T}{T_C},$$
  
$$\tau_{e-ph}^{-1} = \alpha_{e-ph} \left(\frac{T}{T_C}\right)^p,$$

where  $\beta = 4ln2/[\sqrt{(ln^2(2\pi g) + 64/(\pi^2 g))} + ln(2\pi g)]$ ,  $g = e^2 R_s/(2\pi^2 \hbar)$ ,  $\alpha_{e-e}$  and  $\alpha_{e-ph}$  are material-dependent constants, *p* is the power index, which is in the range of 2-3 for Nb films, as shown previously in Ref. [12].

# **Results and Discussion**

Fig. 1, *a* shows the dependence of the sheet resistance  $R_s$  upon temperature for all samples. One can see that the passivated samples have lower  $R_s$  and higher values of the critical temperature  $T_c$  than the uncovered samples. This observation means that 5 nm-Si layer protects films from unintentional oxidation.

a)



Fig. 1. The dependence of the sheet resistance  $R_s$  upon temperature for all investigated samples (*a*); The normalized magnetoconductance  $\delta G(B,T)$  versus magnetic field for a representative sample (A1) (*b*); Different colors of the curves correspond to different operating temperatures marked on the  $R_s(T)$ -curve in the inset. The dashed black lines represent the fits by Eq.(2).

In Fig. 1, b we plot the normalized magnetoconductance for a representative sample A1. The magnitude of the phase-breaking time at each operating temperature is deduced from the fits of the experimental data by Eq.(2) (the dashed black lines in Fig. 1).

Fig. 2 shows the phase-breaking time  $\tau_{\phi}$  as a function of temperature for all studied samples. We fit the experimental dependence  $\tau_{\phi}(T)$  with Eq.(3) (dashed black lines) using  $\alpha_{e-ph}$ , p, and  $\tau_s$  as fitting parameters. The best-fit values, which defined the electron-phonon time  $\tau_{e-ph}$  ( $\alpha_{e-ph}$ <sup>-1</sup> and p), are listed in Table 1. One can see that the best-fit values of electron-phonon time at the T range under study are much smaller than the estimated values of the electron-electron time. Thus we exclude electron-electron scattering from further analysis. Since  $\tau_s$  is supposed to be a temperature independent parameter and  $\tau_{sc} \sim \ln(T)/T$ , we assume that the increase in  $\tau_{\phi}$  with against decreasing of *T* is determined by the electron-phonon inelastic scattering time. First of all, we observe the close power-law *T*-dependence of  $\tau_{\phi}$  for the passivated samples ( $\tau_{\phi}(T) \sim T^{2.5}$  for A1 and  $\tau_{\phi}(T) \sim T^{1.5}$  for A2), meanwhile  $\tau_{\phi}$  for the uncovered samples does not show a pronounced dependence on *T*. The observed results for  $\tau_{e-ph}(T)$  in the passivated samples are also close to previous reported data for thin Nb films [12]. The sign of saturation in *T*-dependence of  $\tau_{\phi}$  is observed for A2, B1, and B2 samples and can be explained by the finite value of  $\tau_s$  (see Table 1). Now, we can compare  $\tau_s$  derived from  $\tau_{\phi}$  with  $\tau_s$  obtained from the quantitative analysis of suppression of  $T_c$  in Nb films [13].

The estimated value of  $\tau_s$  for passivated sample A2 is an order of magnitude longer than value of  $\tau_s$  reported in [13], which indicates that the magnetic disorder cannot be the dominant factor in this film. In additional, we assume that the suppression of  $T_s$  with the film thickness in the passivated samples can be related to the inverse proximity effect due to metallic silicide at the Nb-Si interface [3] rather than the magnetic disorder [13].

In contrast, we observe the enhanced phase-breaking rate  $\tau_{\phi}^{-1}$  for the uncovered samples B1 and B2, which evidences an additional phase-breaking mechanism. We also notice that the low-temperature value of  $\tau_{\phi} \approx 1.5$  ps is consistent with the spin-flip scattering time  $\tau_{s}$  in Nb reported in [13]. This result, together with the observed decrease of  $\tau_{\phi}$  and the saturation in  $\tau_{\phi}(T)$ -dependence, indicates that the electron dephasing in the uncovered samples may be caused by the magnetic disorder in the native oxide layer [6]. It is the worth remark that the magnetic disorder concentrated in native Nb oxide is known to be a source of parasitic magnetic flux noise in uncovered Nb-based superconducting quantum interference devices [14] and power-independent losses in Nb-based resonators [15].



Fig. 2. Temperature dependence of the phase-breaking time  $\tau_{\phi}$  upon temperature extracted from the magnetoconductance measurements. The data are plotted with symbols on a log-log scale

#### Conclusion

We investigated the influence of the Si passivating layer on the electron phase-breaking rate in ultrathin Nb films. We observed the power-law type  $\tau_{\phi}(T)$ -dependence in the passivated Nb samples and a tendency towards saturation in the  $\tau_{\phi}(T)$ -dependence for the uncovered Nb samples. The latter may indicate the presence of the magnetic disorder in the native niobium oxide on the Nb surface. The study can be useful for the design of microelectronic Nb-based devices.

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