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# Influence of hydrostatic pressure on superconducting properties of niobium thin film

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We have studied superconducting properties of niobium thin films under hydrostatic pressures up to 3 GPa. The films with thickness of 100 nm were prepared in the high vacuum DC magnetron sputtering system (with critical temperature TC = 8.95 K at ambient pressure). The produced high quality films have been characterized using electrical resistivity and magnetization measurements, X-ray diffraction, and atomic force microscope imaging. We have observed increase of TC with increasing value of applied pressure (dTC/dp = 73 mK/GPa) up to 3 GPa. This observation is different to pressure effect observed on bulk sample of Nb. In this paper we are discussing the origin of this discrepancy.



## 1. Introduction

Experimental study of superconducting (SC) thin films under extreme conditions is still an uncharted field of experimental physics. Combination of several extreme conditions such as low temperature, high magnetic field and application of high hydrostatic pressure can reveal interesting phenomena in superconductors. In such experiments one can expect to observe unusual behavior of critical temperature ($T_C$) and other SC parameters. Since the discovery of superconductivity in the iron based materials, great efforts have also been devoted for the preparation of thin films of such materials [1,2]. Especially, the iron pnictides are very sensitive to external and chemical pressure and these effects can be induced also by the strain in thin films [1]. Other related materials are single or multilayer SC dichalcogenides [3–5]. However there are still open questions what happens with thin films of elemental superconductors under external hydrostatic pressure. Very important elemental superconductor from application point of view is niobium. Niobium is widely used in many applications, e.g. SC quantum processors for quantum computers [6], the SC magnets ($Nb_3Ge$, $Nb_3Sn$, NbTi) and niobium-containing superalloys. Therefore it is interesting to compare SC properties of niobium thin film with bulk niobium sample under pressure. In this paper we will present results of our experiments performed on niobium thin films and we will compare these results with the bulk niobium properties. Shock-wave experiments on bulk Nb up to 170 GPa have shown no evidence of structural phase transition [7], therefore any changes in $T_C$ are attributed to the changes in electronic properties [8]. In case of niobium thin films we have to also take into account residual strains in film which come from the mismatch of lattice parameter between Nb and substrate.

Bose et al. [9–12] have shown that the grain size dependence of superconductivity in niobium thin films is mainly governed by the changes in the electronic density of states and not by the electron–phonon coupling due to surface effects. On the other hand, in case of weak coupling type I superconductors such as Al and Sb, the increase of the $T_C$ is caused by softening of surface phonon modes [13]. Direct measurement of SC energy gap in nano-structured thin films revealed that energy gap is suppressed with decreasing of the size of grains [9]. The value of $T_C$ is also decreasing with decreasing Nb grain size, which was shown in transport and magnetic experiments performed by Bose et al. [9,10]. In accordance with Anderson criterion, thin films with grain size below 8 nm do not show SC transition [9]. With decreasing of the typical grain size, the films transform from metallic to weak insulating state. Measurements of $H_{C2}$ shown that in films with the grain size in order of 10 nm, the $H_{C2}$ is connected with the changes in Ginzburg–Landau coherence length which is dependent on *a*) decreasing of size of grain and subsequently by increase of disorder and *b*) effective decrease of density of states on Fermi level due to formation of Kubo gap [10]. Therefore the $H_{C2}$ in thin film of Nb is a nonmonotonic function of grain size. In the range between 60 and 20 nm, the value of $H_{C2}$ is increasing with decreasing size of grain. For case of grain size about 20 nm the $H_{C2}$ is already increased by a factor of ~2.5 in comparison with bulk $H_{C2}$. On the other side, the value of $T_C$ is very close to bulk value of 9.21 K [14]. This effect can be attributed to the decrease of coherence length due to shorter mean free path of electrons with smaller size of grains. Below 20 nm size of grains, $H_{C2}$ is again decreasing with decreasing

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size of grains, the value of $T_C$ and energy gap is decreasing as well. This effect can be explained by a quantization of electron levels on Fermi level [10].

## 2. Experimental details

Nb thin films were prepared in the high vacuum DC magnetron sputtering system (AJA International, USA). Films were deposited on the glass microscope cover slides, which were sonicated in acetone and isopropyl alcohol prior to film deposition. The slides were also cleaned in argon plasma just before film deposition. The slides have an RMS (root mean square) roughness of ≈1 nm determined from atomic force microscopy (AFM) measurement. The niobium films were deposited in ultrapure argon (99.999%) plasma at 0.4 Pa. We produced several samples under different preparation conditions in order to gain $T_C$ as high as possible. Quartz crystal thickness monitor has been used to measure deposition rates. Films were cut to desired size by the diamond cutter and four gold contacts were bonded by ultrasonic contact bonder. We choose 100 nm thickness because such thickness is far from quantum size effect (QSE) region and bulk properties are expected [11]. Also, size of grains was optimalized (typical size is around 57 nm) in order to avoid QSE. Therefore we can compare our results with bulk sample and differences in behavior can be attributed to changes in microstructure of thin film and changes in electronic properties.

High pressure measurements of electrical resistivity have been performed in piston-cylinder cell with an NiCrAl alloy insert which allows to achieve a maximum pressure about 3 GPa. As manometer we used Pb ($T_C$ = 7.19 K at ambient pressure). Daphne oil has been used as a pressure medium. It is known that silica (quartz) is stable up to 18–35 GPa, where quartz amorphizes [15]. Therefore no substrate changes are expected from the substrate phase transitions. Applied magnetic field was oriented perpendicular to Nb thin film surface.

The structure of films before applying pressure and after pressure treatment has been probed by X-ray diffraction (XRD) and AFM. XRD measurements have been performed in Argonne National Laboratory using rotating anode X-ray generator with Cu α radiation (Rigaku). Imaging by AFM has been performed at Faculty of Science, P. J. Šafárik University on ICON Bruker in tapping mode using silicon tips from Micromasch.

## 3. Results and discussion

In order to investigate the influence of hydrostatic pressure on SC properties of niobium thin films, we have used several experimental techniques. The pressure dependence of $T_C$ has been studied by the measurement of electrical resistivity and magnetization. The structure of thin films has been probed by XRD and AFM imaging. In the next subsections we will discuss results of above mentioned experimental techniques.

### 3.1. Electrical resistivity

Fig. 1 shows pressure dependence of $T_C$ for niobium thin film and bulk niobium sample (data from [14]). In inset is a temperature dependence of resistivity of Nb thin film for different values of applied pressure. The $T_C$ was estimated as temperature at which the resistance dropped to 50% of its normal state value. The very narrow width of transition (about 30 mK) points to a high purity of sample and also high homogeneity of applied pressure. The value of $T_C$ for ambient pressure is 8.95 K and increases with pressure. At pressure 3 GPa it achieves 9.16 K. This result shows different behaviors than bulk sample. T. F. Smith [14] observed pressure dependence of several bulk samples with very high residual resistivity ratio (RRR from 500 to 11,000) and achieved $T_C$ pressure dependence ($dT_C/dp$) between −16 and −25 mK/GPa. In Fig. 1 are the shown data of single crystal bulk sample with the highest $T_C$ = 9.204 K and RRR ≈ 1300 [14]. The value of RRR of our niobium thin film is approximately 3

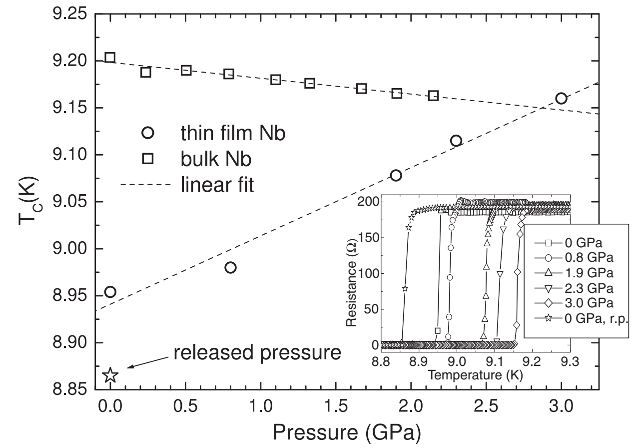


**Fig. 1.** Pressure dependence of $T_C$ for niobium thin film (circles) and for bulk sample of Nb (squares) from Ref. [14]. Star represents $T_C$ after releasing of pressure measured at ambient pressure. Inset shows the temperature dependence of resistivity of Nb thin film for different values of applied pressure (r.p. means released pressure).

and we have positive pressure effect with $dT_C/dp$ = 71 mK/GPa. At the highest pressure about 3 GPa, the value of $T_C$ is even higher than that of bulk sample at the same pressure. After the releasing of pressure, we performed measurement of $T_C$ (see Fig. 1) and observed a decrease of $T_C$ value to 8.86 K, which is lower than before applying of pressure. We suppose that the application of pressure can release the strains induced in thin film during sputtering. The change of microstructure by application of pressure was studied by XRD and by AFM imaging (see next sections).

From the measurements of electrical resistivity at several applied pressures we constructed phase diagram (see Fig. 2). Significant change with pressure can be seen only at low value of magnetic fields. The value of the upper critical field $H_{C2}$ ≈ 2.3 T is much higher than in case of bulk sample ($H_{C2}$ ≈ 0.4 T) [16] and is in good agreement with previous work [10].

### 3.2. Magnetic moment measurement

In measurements of electrical resistivity, the percolative path for electrical current can play important role. Therefore we also performed measurements of magnetization, which probes that the whole sample and the value of $T_C$ are independent on percolation effect. Fig. 3 shows the DC magnetization data obtained by using MPMS (magnetic property measurement system) magnetometer for the both samples. The film was oriented perpendicular to applied magnetic field B = 10 mT. As prepared film has $T_C$ ≈ 8.85 K and film after pressure treatment

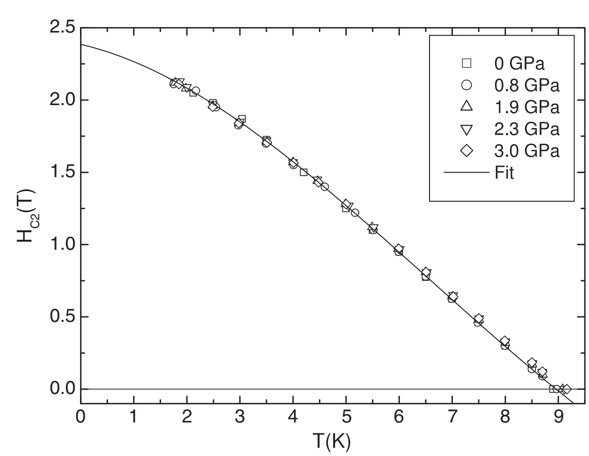


**Fig. 2.** Phase diagram $H_{C2}$ vs. $T$ of niobium thin film under several applied pressures. Significant difference can be seen only at low fields.