# Influence of fast proton irradiation with energies of 12.4 and 12.8 MeV on magnetic characteristics and microstructure changes of superconducting intermetallic compound $Nb_3Sn$

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**Abstract.** Data on magnetization, magnetic susceptibility and microstructure modification of irradiated $Nb_3Sn$ platelets are presented. The irradiation was produced at room temperature by fast protons with the energies of 12.4 and 12.8 MeV with fluencies of $5x10^{17}$ and $1x10^{18}$ $cm^{-2}$. Variation of the superconducting transition temperature *versus* irradiation dose was determined. Temperature dependence of magnetic susceptibility of $Nb_3Sn$ platelet with 160 μm thickness demonstrates several steps corresponding to different superconducting transition temperatures. We supposed that there are layers inside the sample with significantly different radiation damage levels caused by particles movement termination (Bragg peak). It was found that after the irradiation a lot of randomly oriented platelet-like Nb-enriched particles of 0.1-0.5 μm size appear in volumes with maximal damages.

## 1. Introduction

Multifilament wires of superconducting intermetallic compound $Nb_3Sn$ are the main candidates for utilising in superconducting magnetic system of future large projects of accelerators at CERN and in future fusion reactors. So study of influence of fast particle irradiation on critical parameters of these materials is very important at least for estimation of magnetic system lifetime. It is well known, that irradiation of $Nb_3Sn$ leads to strong changes of superconducting parameters [1-5] and the changes depend on both the initial state of material and irradiation parameters. Previous investigations of $Nb_3Sn$ platelets irradiated by protons with energies of 65 MeV and 24 GeV up to fluence of $3x10^{17}$ $cm^{-2}$ revealed almost 100% increase of the critical current density $J_c$ and ~ 5% increase of the upper critical field $B_{c2}$ while at higher fluencies both $J_c$ and $B_{c2}$ decreased [3, 6]. It was found also that

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the critical temperature $T_c$ decreases monotonically with the fluence increase [3, 6, 7]. Irradiation of $Nb_3Sn$ multi-filamentary wires by 32 MeV protons demonstrated similar behavior [8]. However, the investigation of changes of $Nb_3Sn$ microstructure in wires is quite difficult because of complicated design of modern commercial wires. In contrast, irradiation of bulk $Nb_3Sn$ material is very helpful to understand the main physical mechanisms of radiation defects formation in these materials. Therefore, in this paper the results of study of $T_c$ and microstructure changes in $Nb_3Sn$ thin platelets after irradiation by fast protons are presented.

## 2. Platelets and experimental technique

Uniform and pure $Nb_3Sn$ platelets were cut from a cylindrical ingot of $Nb_3Sn$ produced by a reaction under HIP conditions, described in details in Refs. 3 and 6. The platelets parameters are presented in the Table 1. Typical area of platelets was 2x2 $mm^2$. Thickness of the unirradiated platelet was 0.19 mm. In order to observe at the same fluence both the effects of steady loss and Bragg peak (see Figure 1), pairs platelets in a stack were subjected to the irradiation simultaneously. The first pair with platelets of 0.17 and 0.14 mm thicknesses was irradiated by $5x10^{17}$ $cm^{-2}$ fluence, while the second one with 0.22 and 0.16 mm thicknesses was irradiated by $10^{18}$ $cm^{-2}$ fluence.

The irradiations were done on the isochronous cyclotron in NRC "Kurchatov institute" at room temperature using water-cooled targets separated by an aluminum foil from vacuum. The energies and fluencies of protons were 12.4 and 12.8 MeV, $5x10^{17}$ and $1x10^{18}$ $cm^{-2}$, correspondingly. The scheme of irradiation is presented on Figure 1a. The proton energies were adjusted for different fluencies to make equal the penetration depth of protons and total thickness of two platelets. Numerical calculations of damage profiles were made with SRIM-2013 code [9] for $Nb_3Sn$ density 8.9 $g/cm^3$ and displacement energy 25 eV for both Nb and Sn atoms. According to calculations, the mean path for 12.8 MeV protons passing the front platelet exceeds 300 μm (see Fig. 1b). Thus, it was assumed that the stack of #1a and #2a platelets should be completely penetrated by protons and protons should be completely stopped in #1b and #2b stack, forming the peak of radiation damages in the last platelet.

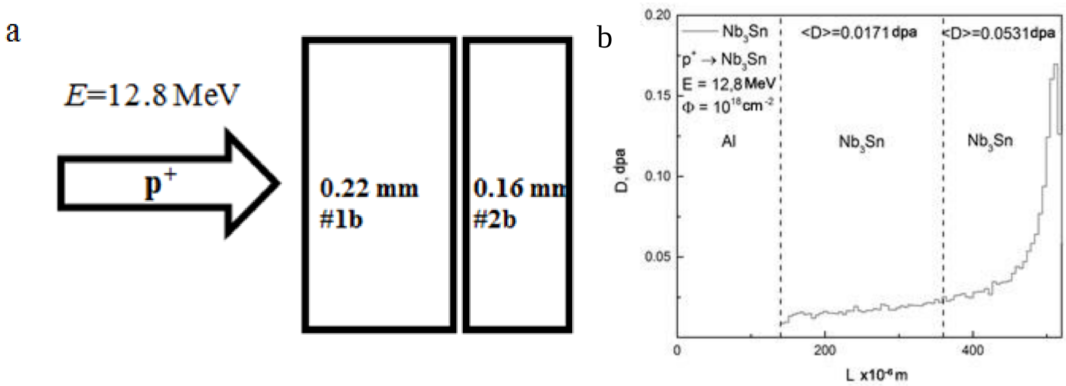


**Figure 1.** (a) The scheme of irradiation; (b) calculated radiation damage profile for $Nb_3Sn$.

Magnetic measurements were performed on the PPMS station in Lebedev Institute in magnetic fields up to 8 T and at temperatures up to 20 K. To estimate the critical current, the magnetization curves at $T$ = 7 K in fields up to 6 T and temperature dependence of remanent magnetization were measured on Vibrating Sample Magnetometer (VSM). For detailed $T_c$ characterization AC magnetic susceptibility was measured in field of 5 Oe at frequency of 37 Hz in the temperature range from 2 up to 20 K.

Detailed microstructural analysis was performed using scanning/transmission electron microscope TITAN 80-300 TEM/STEM (FEI, US) with Probe-Cs corrector and EDX spectrometer (EDAX, US) operated at 300 kV in NRC "Kurchatov Institute". The high angle annular dark field (HAADF) detector (Fischione, US) was used in STEM mode. Platelets were prepared by standard FIB procedure in a Helios (FEI, US) SEM/FIB dual beam microscope.

**Table 1.** The characteristics of $Nb_3Sn$ platelets.

| Type of platelets | Fluence, $cm^{-2}$ | Thickness, mm | Size, $mm^2$ | Mass, g |
|---|---|---|---|---|
| Reference platelet | unirradiated | 0,19 | 2,53x2,47 | 0,00822 |
| Platelet #1a | $5x10^{17}$ | 0,17 | 2,5x1,9 | 0,00620 |
| Platelet #2a | $5x10^{17}$ | 0,14 | 2,5x2,1 | 0,00540 |
| Platelet #1b | $10^{18}$ | 0,22 | 1,1x2,3 | 0,00340 |
| Platelet #2b | $10^{18}$ | 0,16 | 1,5x1,6 | 0,00286 |

## 3. Results and discussion

The results of remanent magnetization measurements for unirradiated and irradiated $Nb_3Sn$ platelets are presented on Figure 3. The curves normalized to magnetization at 7 K demonstrate the essentially different behavior for unirradiated and irradiated samples. For the unirradiated platelets the temperature dependence decreases abruptly and the magnetic moment completely vanishes at the critical temperature 17.9 K of $Nb_3Sn$ [9]. The curves for irradiated platelets #1a, #2a and #1b are similar to the unirradiated one but they drop to zero at lower temperatures. On the contrary, the curve for irradiated sample #2b passes below other curves and drops to zero at 16.2 K. As we assumed, this is the manifestation of non-uniformity of radiation defect on depth in the sample #2b.

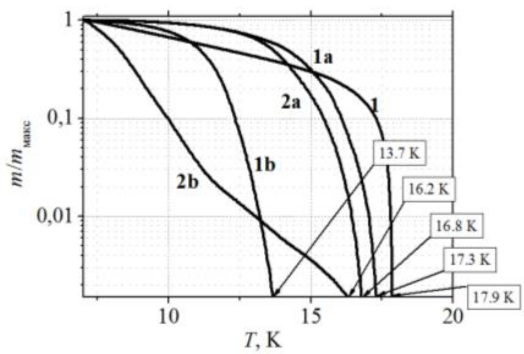


**Figure 2.** Temperature dependence of remanent magnetization of platelets: 1 – 0.19 mm, unirradiated; 1a and 2a – 0,17 and 0,14 mm, irradiated by 12.4 MeV protons with $5x10^{17}$ $cm^{-2}$ fluence; 1b and 2b – 0.22 and 0.16 mm, irradiated by 12.8 MeV protons with $1x10^{18}$ $cm^{-2}$ fluence.

The HAADF STEM image of the platelet with the marks, pointed to the areas of the EDX microanalysis, is presented in Figure 3b. An uniform image contrast indicated the good homogeneous composition of the platelet. Some contrast variations appeared due to the difference in specimen thickness aroused during FIB platelet preparation. Additional confirmation of the homogeneity was obtained from the EDX microanalysis. The Nb:Sn ratio is very close to the $Nb_3Sn$ stoichiometry.

The measured magnetic susceptibility is very sensitive to $T_c$ variations. The superconducting transition for unirradiated platelets corresponds to the critical temperature of pure $Nb_3Sn$ (see Fig. 3). The critical temperature for platelets #1a and #2a (at the fluence $5x10^{17}$ $cm^{-2}$) is close to 15.3 K (Fig. 4). For platelet #1b (at the fluence $1x10^{18}$ $cm^{-2}$) it decreases to 12.4 K (Fig. 5). For platelet #2b the $\chi_{AC}$ curve has several steps at temperatures – 16.2, 13.7, 9.9, 8.7 and 2.7 K, what also confirms our assumption.

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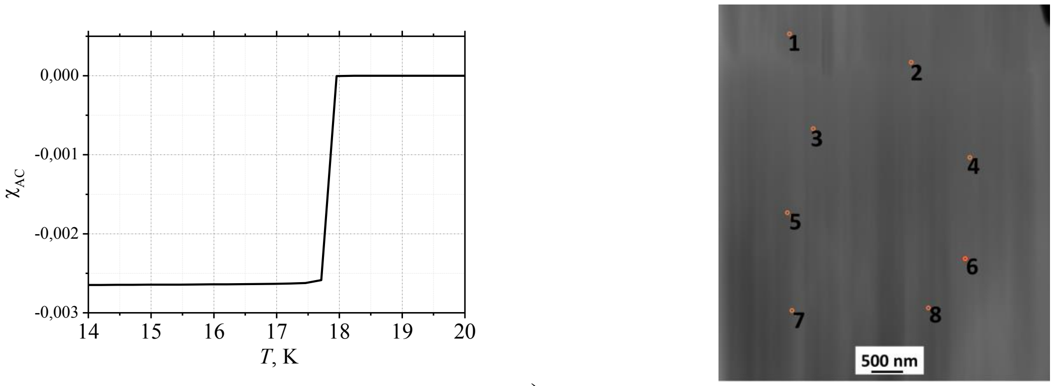
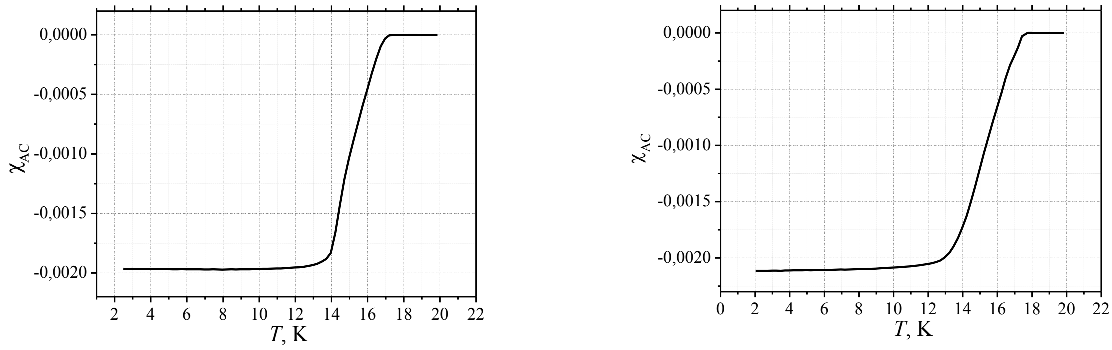


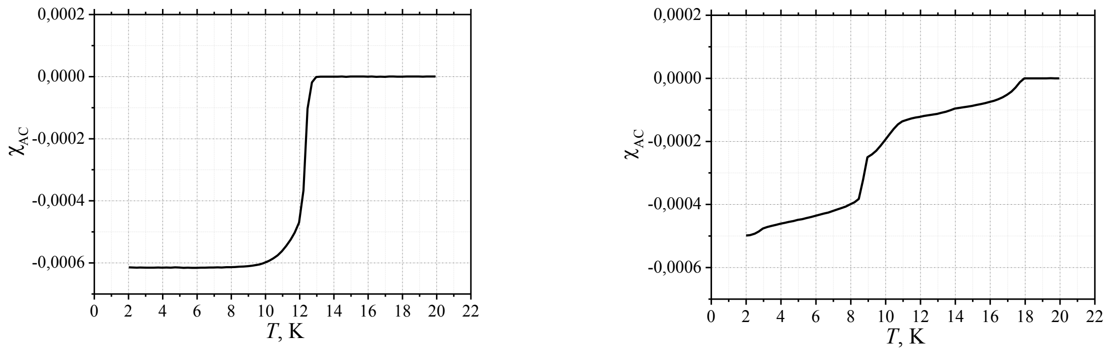

a) b)

**Figure 3.** Temperature dependence of $\chi_{AC}$ magnetic susceptibility (left) and HAADF STEM image (right) for unirradiated platelet #1. The small circles point to the areas of EDX microanalysis (1 – 73 at.% Nb and 27 at.% Sn; 2 – 74 at.% Nb and 26 at.% Sn; 3 – 75 at.% Nb and 25 at.% Sn; 4 – 76 at.% Nb and 24 at.% Sn; 5 – 74 at.% Nb and 26 at.% Sn; 6 – 76 at.% Nb and 24 at.% Sn; 7 – 75 at.% Nb and 25 at.% Sn; 8 – 74 at.% Nb and 26 at.% Sn).




a) b)

**Figure 4.** Temperature dependence of $\chi_{AC}$ magnetic susceptibility for platelets #1a (left) and #2a (right) after proton irradiation with the energy 12.4 MeV and fluence $5x10^{17}$ $cm^{-2}$.

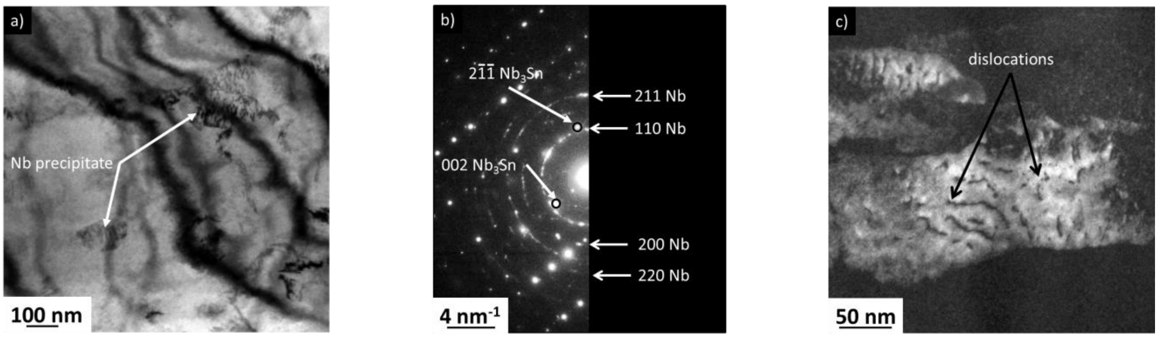



a) b)

**Figure 5.** Temperature dependence of $\chi_{AC}$ magnetic susceptibility for platelets #1b (left) and #2b (right) after proton irradiation with the energy 12.8 MeV and fluence $1x10^{18}$ $cm^{-2}$.

The bright field (BF) TEM image of platelet #2b presented in Figure 6a demonstrates appearance of randomly oriented particles of 0.1-0.5 μm size after irradiation of the sample. The contrast, similar to Moiré fringes, is clearly visible in vicinity of these particles. The selected area diffraction pattern

(SADP) from the area with several similar defects is shown in Figure 6b. The SADP consists of both set of spot reflexes corresponding to A15 structure of $Nb_3Sn$ in [120] zone axis and diffraction rings. Estimation of inter-planar distances, corresponding to these rings, unambiguously demonstrated that they originated from metallic Nb plane indexes (see Fig. 6b). The dark field (DF) TEM images of Nb particle was obtained by selecting part of the $110_{Nb}$ diffraction ring shown in Figure 6b and corresponding image is presented in Figure 6c. The particle exhibits bright contrast in these DF TEM images with dislocations, which are supposed to originate at the Nb particle – $Nb_3Sn$ interface and arise from Nb-$Nb_3Sn$ lattice mismatch. Surprisingly, the EDX microanalysis did not show noticeable presence of extra Nb (see Fig. 7). That means that Nb particle are very thin and do not impact to the EDXS signal. We suppose that these Nb particles adopted platelet morphology with the habit plane parallel to the specimen surface and perpendicular to $e^-$-beam.




a) b) c)

**Figure 6.** BF TEM image of platelets #2b, the areas with typical radiation defects are arrowed (a); SADP from the sample with radiation defects (b) and DF TEM image of Nb precipitate (Note, that dislocations look like dark lines) (c).

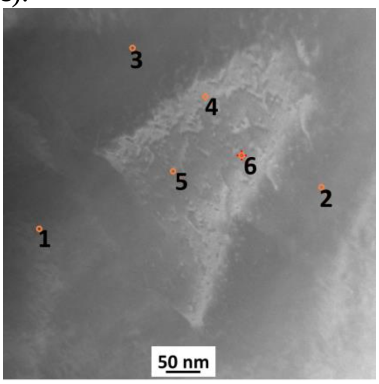



**Figure 7.** HAADF STEM image of the platelet #2b. The small circles point to the areas of EDX microanalysis (1 – 68 at.% Nb and 32 at.% Sn; 2 – 73 at.% Nb and 27 at.% Sn; 3 – 75 at.% Nb and 25 at.% Sn; 4 – 75 at.% Nb and 25 at.% Sn; 5 – 73 at.% Nb and 27 at.% Sn; 6 – 74 at.% Nb and 26 at.% Sn).

## 4. Summary

The experimental investigations of the magnetic characteristics and microstructure analyses of thin $Nb_3Sn$ platelets with different thickness after irradiation by fast protons with the fluencies $5x10^{17}$ (energy 12.4 MeV) and $10^{18}$ $cm^{-2}$ (energy 12.8 MeV) were performed. The curve of temperature dependence of remanent magnetization for irradiated platelet #2b at fluence of $10^{18}$ $cm^{-2}$ passes below the curve for unirradiated platelet. We supposed that an inhomogeneous structure was formed in this platelet under irradiation. This assumption is confirmed by data on magnetic susceptibility. The $\chi_{AC}$ transition curve demonstrated several steps at temperatures 16.2, 13.7, 9.8, 8.7 K (approximately to temperature of critical temperature of pure Nb) and 2.7 K. It was found that thin randomly oriented

Nb-enriched particles 0.1-0.5 μm in size are formed in platelet-like shape under irradiation. The observed structural transformations of sample lead to decrease of critical current density. The results will be extended in further investigations of irradiated $Nb_3Sn$ samples.

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