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MAGNETIC PROPERTIES OF NITROGEN DOPING NIOBIUM SAMPLES*

Z. Q. Yang, X. Y. Lu#, W. W. Tan, D. Y. Yang, Y. J. Yang, J. F. Zhao
State Key Laboratory of Nuclear Physics and Technology, Peking University, Beijing, China

Abstract

Nitrogen doping study on Niobium samples used for the fabrication of superconducting radio frequency (SRF) cavities was carried out. The samples' surface treatments were attempted to replicate that of the cavities, which included heavy electropolishing (EP), nitrogen doping and the subsequent successive EP with different amounts of material removal. The magnetization curves of both doped and un-doped samples have been measured, from which the lower critical field H_{ffp} (First Flux Penetration, ffp) and upper critical field H_{c2} was extracted. The thermodynamic critical field H_c, superheating field H_{sh} and superconducting parameters of samples with different treatments was calculated from the determined reversible magnetization curves. H_{sh} of doped samples is obviously smaller than that of un-doped samples, which may be a possible reason for the reduction of achievable accelerating gradient in SRF niobium cavities after nitrogen doping treatments.

INTRODUCTION

Fundamental understanding of the nitrogen doping mechanism is being carried out extensively [1-5], but yet remains unclear. Two years after the discovery of nitrogen doping phenomenon, A. Romanenko [6] discovered that fast cooling down through T_c in both single cell and 9-cell nitrogen doping cavities helps to much more efficient flux expulsion and results in lower residual resistance. While this effect is not so obvious for un-doped cavities, especially for the cavities with standard preparation which consists of EP 120μm, 800^{0}C heat treatment for 3 hours, EP 20μm and 120^{0}C baking for 24-48 hours. A. Gurevich and G. Ciovati [7] studied the impact of vortex on the residual resistance of SRF niobium cavities. They predicted that the residual resistance has a high sensitivity to trapped flux. Dan Gonnella [8] studied the sensitivity of surface resistance to trapped magnetic flux on both doped and un-doped cavities. Dan Gonnella's experiments showed that nitrogen doping cavities have higher sensitivity of residual resistance to trapped flux. Vertical tests also showed that the sensitivity decreases with the EP removal and will be reduced gradually to the level of un-doped cavities after a certain amount of material removal, which is determined by the specific nitrogen doping condition.

For the nitrogen doping cavities, vertical tests [9-10] showed that the residual resistance has minimum value when the EP removal reaches an optimized value, where also corresponds to the largest quality factor. When the EP removal is smaller or larger than the optimized value, the residual resistance becomes greater. So the variation of residual resistance with EP removal after nitrogen doping treatments is also an important aspect of the physical mechanism of nitrogen doping phenomenon. Limited to our experimental conditions, we tried to study the impact of nitrogen doping treatments on the residual resistance through experiments on Nb samples to seek the physical explanation of it.

As mentioned above, the diffused N may have an impact on the material's flux pinning behaviour and thereby affects the cavities' residual resistance. So it is necessary to study the magnetic properties of Nb samples before and after nitrogen doping treatment. Both doped and un-doped samples were electropolished with different amounts of material removal. Magnetic measurements on Nb samples were carried out with a SQUID magnetometer (Quantum Design MPMS-XL-7).

EXPERIMENTAL PROCEDURE

To avoid heat generation, instead of wire electro-discharge machining (EDM), the niobium samples were manually processed. The niobium strips were polished smoothly on 180-grit sandpaper, 300-grit sandpaper and 1200-grit sandpaper. The samples' treatments were attempted to replicate that of the cavities. So the niobium strips were etched by EP ($HF:H_2SO_4$=1:9), with the material removal of about 150μm, to remove the mechanical damage layers and surface contaminations introduced during handling or exposing to the air.

Nitrogen Doping Treatment

The nitrogen doping treatments and 800^{0}C heat treatments of the niobium samples used for the magnetic measurements can be seen in [11].

Magnetization Measurement

The magnetization was measured by using a commercial SQUID magnetometer (Quantum Design MPMS-XL-7) at Peking University. The weight of experimental samples lies between 80-260mg. For the considerations of measurement range, the DC module was used with the measurement accuracy of 1×10^{-8} emu. Mounted in a sample holder, the sample was transported to the sample space together with a rigid sample rod during which the sample was cooled from room temperature at zero field (ZFC). The sample's motion length is 4cm. The magnetic field is homogeneous to 0.05% for this regime. The measuring range of DC applied field H_a lies between zero

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† Email address: xylu@pku.edu.cn

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and 5000Oe, including an ascending branch and a descending branch. At each measuring point, the sample was driven through the detection coil and stopped at a number of positions over the specific scan length. The current induced by the magnetic moment of the sample would be detected by the superconducting detection coil, which can provide a highly accurate measurement of the sample's magnetic moment.

Experimental Results

Two groups of samples were used in the magnetization measurements, labeled as "Test 1" and "Test 2", respectively. Magnetization curve of doped sample is shown in Figure 1 for reference, which is typical of type II superconductors.

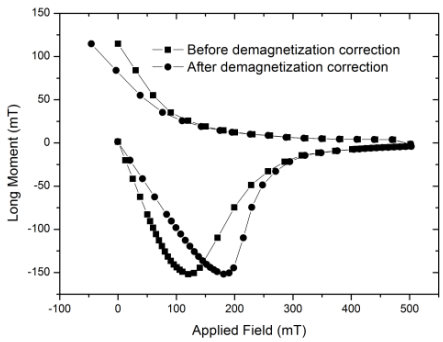

Figure 1: Magnetization curves of nitrogen doping sample before and after demagnetization correction.

H_{ffp} (First Flux Penetration, ffp), which is H_{c1} for reversible magnetization curve, was determined by the field at which a deviation started from the linear magnetization $M(H_a)$ in the Meissner state [12]. Deviation from the linear behavior means the disappearance of complete diamagnetism.

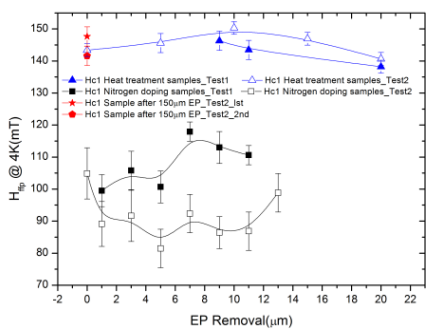

Figure 2: H_{ffp} of samples with different treatments.

H_{c2} of all the samples is defined as the field at which the magnetization M(Ha) reaches zero. H_{c2} of all samples with different treatments is shown in Figure 3

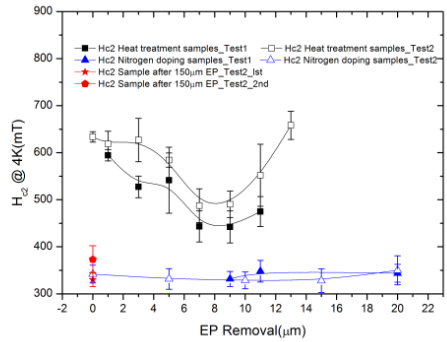

Figure 3: Hc2 of samples with different treatments.

It can be seen in Fig. 2 that H_{ffp} of all doped samples is about 30% lower compared to the un-doped samples. H_{c2} of doped samples is significantly higher due to the diffused nitrogen. H_{c2} of doped samples in both Test 1 and Test 2 have the same change trend with the EP removal. The doped samples with EP removal 7-9μm have minimum H_{c2} in both Test 1 and Test 2. Both H_{ffp} and H_{c2} of doped samples failed to recover to the value of un-doped samples up to 13μm. This means the nitrogen doping effect of depth exceeds 13μm under our doping recipe.

DETERMINATION OF REVERSIBLE MAGNETIZATION CURVE

A set of superconducting material parameters such as the Ginzburg-Landau parameter κ, the penetration depth λ and the coherence length ξ can be deduced from the relationship between H_{c2} and the thermodynamic critical field H_c. So H_c is the key of the analysis of magnetization measurements experimental data.

For niobium (RRR≈ 300) used for SRF cavity fabrication, the magnetization curve shows significantly hysteresis. The reversible magnetization curve should be determined to assessment the superconducting parameters correctly.

The specific heat and magnetization measurements [13] showed that for $H_c < H_a < H_{c2}$, the thermodynamically deduced reversible magnetization curve is approximately equal to the average magnetization between increasing and decreasing fields: $M_{rev} = (M_+ + M_-)/2$, where M_+ corresponds to the ascending branch and M_- to the descending branch. Torque magnetometry measurements by Willemin and co-workers [14] have shown the validity of the above procedure. Theoretical study of Kes [15] showed the reversible magnetization is indeed the average magnetization of the increasing and decreasing branches in first-order approximation. This will be the foundation of the determination of the reversible magnetization curve.

The complete reversible magnetization curve can be divided into four parts. That is the Meissner state part, the part near H_{ffp}, the part of intermediate field region (London region) and the part close to H_{c2} (Abrikosov region).

The completely determined reversible magnetization curve is shown in Fig. 4.

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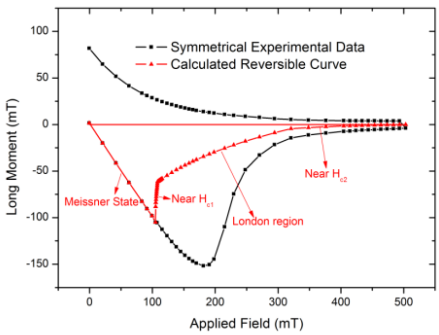

Figure 4: The calculated complete reversible magnetization curve that reflects the typical physical characteristics of ideal type II superconductor.

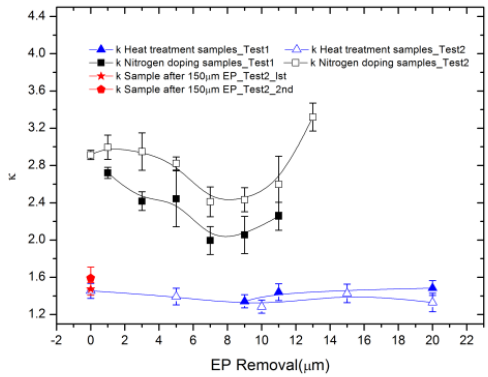

Figure 5: Ginzburg-Landau parameter κ of samples with different treatments.

Table 1: Thermodynamic Critical Field of Samples with Different Treatments in both Test 1 and Test 2

Test 1		Test 2	
Samples	**H_c at 0K/mT**	**Samples**	**H_c at 0K/mT**
N-D 1μm	190±1	N-Doping 0μm	189±12
N-D 3μm	189±1	N-Doping 1μm	180±1
N-D 5μm	192±1	N-Doping 3μm	185±4
N-D 7μm	193±1	N-Doping 5μm	181±1
N-D 9μm	187±2	N-Doping 7μm	187±9
N-D 11μm	183±2	N-Doping 9μm	185±1
H-T 9μm	214±6	N-Doping 11μm	185±8
H-T 11μm	210±2	N-Doping 13μm	172±1
H-T 20μm	201±1	NoHTnoND 1st*	195±4
		NoHTnoND 2nd*	204±6
		HT-0μm	204±6
		HT-5μm	207±1
		HT-10μm	219±1
		HT-15μm	200±2
		HT-20um	229±2

*Sample after EP 150μm without neither nitrogen doping treatment nor 800 ℃ heat treatment.

The calculated superconducting parameters such as κ, λ and ξ can be seen in Figure 5, Figure 6 and Figure 7. The κ and λ of all doped samples is higher than that of un-doped samples, while the ξ of doped samples is lower. The diffused nitrogen can influence the niobium's κ, λ and ξ value by changing the electron mean free path [16-17]. In both Test 1 and Test 2, κ and λ of doped samples have a minimum value between EP removals of 7 μm to 9μm, which is also the location of the maximum value of ξ. This means the amount of trapped flux is least when the EP removal of doped samples is 7-9 μm under our experimental conditions. When the subsequent EP removal is less than 7μm, the amount of trapped flux decreases as the material removal increases. However, when the subsequent EP removal is more than 9μm, the amount of trapped flux increases with the sustained reduction of nitrogen impurity.

H_c can be calculated from the reversible magnetization curve. H_c of samples with different treatments in both Test 1 and Test 2 is listed in Table 1. The thermodynamic critical field averaged over un-doped samples is 208±10mT. H_c of doped samples is slightly smaller compared to the un-doped samples because of their lower H_{ffp}.

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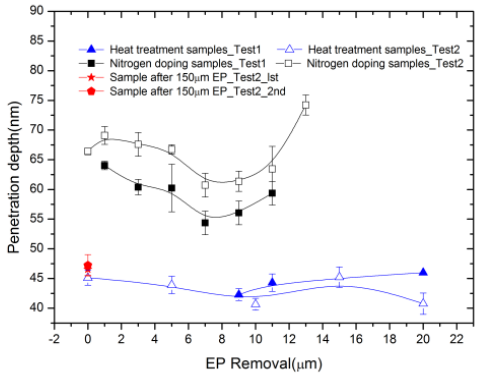

Figure 6: The penetration depth λ of samples with different treatments.

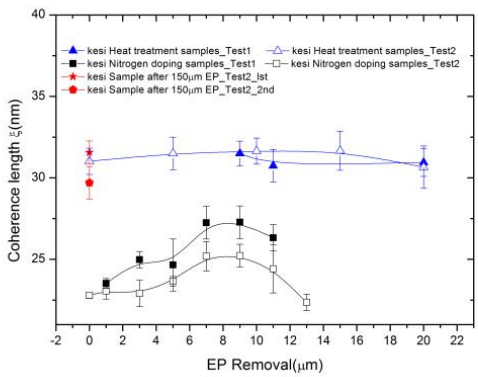

Figure 7: The coherence length ξ of samples with different treatments.

SUPERHEATING FIELD

In the usual nanosecond timescale of an RF field, the RF critical field $H_{rf,\,crit}$ is expected to be up to H_{sh}, which has been verified by high pulsed power measurements on both nitrogen doping cavity [18] and cavity with standard treatment [19]. Therefore, H_{sh} is the fundamental limit to the accelerating field for niobium cavity.

H_{sh} can be estimated through the surface energy considerations. For microwave conditions, the H_{sh} can be calculated by [20]:

$$H_{sh} = \sqrt{2}\frac{H_c}{\sqrt{\lambda/\xi}} = \frac{\sqrt{2}}{\sqrt{\kappa}}H_c \qquad (1)$$

H_{sh} of samples with different treatments at 0K is shown in Figure 8. H_{sh} averaged over un-doped samples is 227±1 6mT, which is in consistent with the estimated value of 240mT in pure Nb. H_{sh} of doped samples is obviously smaller than that of un-doped samples. H_{sh} of doped samples in both Test1 and Test2 have the same change trend with the EP removal. The largest H_{sh} value of doped samples in Test1 and Test2 at 0K is 177±6mT and 161±6 mT, respectively. This agrees well with the H_{sh} value of nitrogen doping cavity at 0K calculated from material parameters using Ginzburg-Landau theory by Cornell University. The doped samples with EP removal 7-9μm have maximum H_{sh} in both Test 1 and Test 2. Measurements on nitrogen doping ingot niobium cavity, carried out by P. Dhakal and co-workers [9] from Jefferson Lab, have shown the breakdown field varied with successive material removal in the same way under their experimental condition.

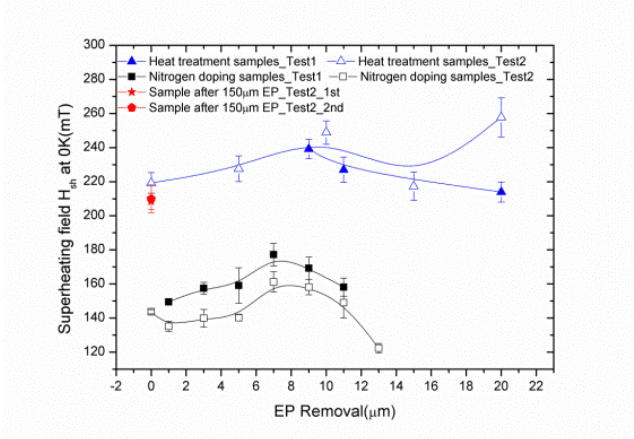

Figure 8: H_{sh} of samples with different treatments.

CONCLUSION

DC magnetization measurements have been carried out to study the magnetic properties of Nb before and after nitrogen doping treatment. H_{ffp} of all doped samples is about 30% lower compared to the un-doped samples. H_{c2} of doped samples is significantly larger than that of un-doped samples due to the decreased electron mean free path affected by diffused nitrogen. H_c and superconducting parameters such as κ, λ and ξ were calculated from the determined reversible magnetization curves and they are consistent with each other. The dependence of H_{sh} on EP removal under our nitrogen doping condition is obtained. H_{sh} of doped samples is obviously smaller than that of un-doped samples, which may be a possible reason for the reduction of achievable E_{acc} in SRF niobium cavities after nitrogen doping treatments. For typical elliptical cavity geometry, H_p/E_{acc}=4.2mT/MV/m leads to E_{acc}~40MV/m as the fundamental limit to the nitrogen doping defect free Nb cavities under our experimental condition. Flux vortex may penetrate into the cavity at a field-enhanced defect, which may cause a smaller breakdown field.

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