Suppressed Meissner-effect in Niobium: Visualized with polarized neutron radiography

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Abstract. Low temperature superconductors ideally exhibit the Meissner-effect, i.e. magnetic flux is expelled from the material during superconducting transition. We report about the complete suppression of the Meissner-effect in two differently surface-treated niobium samples by means of polarized neutron radiography. Both samples were studied in the Meissner phase, T < Tc = 9.25[K] with an external magnetic field Bext = 6.3mT and for T < Tc and Bext = 0. Neutron radiographs of both samples were recorded, imaging the depolarization of the neutron spin for T > Tc and Bext = 6.4mT, T < Tc and Bext = 6.3mT, and for T < Tc and Bext = 0. After turning off Bext at a temperature below Tc strong position dependent flux pinning was observed in the untreated sample and more uniform flux pinning in the case of the surface - BCP treatment.

1. Introduction

Flux pinning in type II superconductors has been extensively studied and a large number of publications discuss pinning centres, interactions between flux lines, and the investigations on the geometry of pinning centres. Already very early papers deal with temperature and field dependence of dislocation pinning in Niobium alloys, attributing the pinning process to a second-order elastic interaction between dislocations and the fluxoid lattice [1]. Several mechanisms have been suggested to explain currents in rough surfaces of type II SC such as Meissner currents, the surface sheath, the surface barrier and flux pinning. Meissner currents and flux pinning were connected with a loss of the Meissner state [2], however, the change in flux density between the outside and inside of the superconductor, the greatest contribution is suggested to come from the Meissner currents [2]. In order to understand flux pinning in type II SC a concept of flux-lattice elasticity was developed predicting pinning functions, which explained measured Lorentz force curves [3]. Another aspect was promoted by the observation of the paramagnetic Meissner effect (PME) [4], which was explained by the persistence of the giant vortex state with the fixed orbital quantum number L. The numerical solution of the Ginzburg-Landau equations showed that the compression of the flux trapped inside the giant vortex state results in the PME. The PME is suppressed, and the normal diamagnetic response is recovered, by increasing the applied field as was found out in [5,6]. Corresponding vortex phase diagrams are given in [7], however, they could not observe a multi-critical point in Nb crystals. Some works were performed to investigate Meissner and shielding effects recording zero field cooling (ZFC) and FC on polycrystalline Niobium samples with different pinning strengths [8]. There the Meissner fraction was found almost unity for the annealed sample from which pinning centers were removed and the effect was much less for the unannealed one.
Niobium is commonly used for superconducting radio frequency resonators. The material must fulfil purity criteria concerning dissolved gases like hydrogen, oxygen and nitrogen that decrease the heat conductivity at liquid helium temperature and degrade the cooling of the rf surface. Moreover, any deviation from the ideal Niobium lattice, like contaminations by foreign metals, grain boundaries, dislocations, dissolved oxygen or hydrogen can lead to magnetic flux pinning. Flux-pinning contributes to the total surface resistance of the cavity, and thus leads to increased heat dissipation in rf fields [9]. It can even dominate over the (unavoidable) baseline BCS-resistance making it the cost driver of the cryo-plant and thus the whole superconducting accelerator – a solid reason to put more effort in understanding and possibly avoiding flux pinning in SRF applications.

By means of radiography with polarized neutrons [10]-[15] one can image magnetic fields in the bulk of massive samples and thus get information about the magnetic field distribution inside and outside of a sample. Low temperature superconductors ideally exhibit the Meissner-effect, i.e. magnetic flux is expelled from the material during the superconducting transition. However, a marginal type II superconductor like niobium can behave like a type II superconductor and trap up to 100% of the ambient magnetic flux during the superconducting transition.

Thus, by means of polarized neutron radiography it can be distinguished if the sample is in the Meissner state or the intermediate state where magnetic flux pinning occurs. We have measured the nearly complete suppression of the Meissner-effect in two differently surface-treated Niobium samples by means of polarized neutron radiography. It turned out that the magnetic distribution of the trapped field depended strongly on the surface treatment.

2. Polarized neutron radiography
The neutron radiographies of these Niobium samples were recorded with PONTO, an instrument of the University of Applied Sciences “Beuth Hochschule für Technik” Berlin, dedicated to radiography and tomography with polarized neutrons. It is located in neutron guide hall of the BER II reactor of the Helmholtz Zentrum Berlin. Neutrons are perfect to study bulk materials and magnetic fields, the latter due to the spin of the neutron which interacts with a magnetic field B as

\[
\dot{s}_j(t) = \frac{\mu_N}{\hbar} \cdot g \cdot \left[ \vec{B}(t) \times \vec{S}_j(t) \right], \quad j = x, y, z
\]

with \( \mu_N = 5.05078343 \times 10^{-27} \text{[J/T]} \) is the nuclear magneton of the neutron and \( g = -3.826085 \) the Landé-factor for neutrons, \( \hbar = 6.6260755 \times 10^{-34} \text{[J.s]} \), \( \hbar = \hbar/2\pi \). The spin rotation can be treated like a classical dipole having the Larmor frequency \( \omega_L = \gamma_L \cdot B \). \( \gamma_L \) is the gyromagnetic ratio of the neutron, \( \gamma_L = g \cdot \mu_N/\hbar = -1.83247 \times 10^8 \text{rad.s}^{-1}.\text{T}^{-1} \). Entering a magnetic field the neutron spin starts Larmor precessions, whereas the number of rotations depend the strength of magnetic field and on the path length trough the field, as

\[
\phi = \omega_L \cdot t = \gamma_L \cdot \frac{B \cdot t}{v} \int B \cdot ds = \frac{\gamma_L \cdot m}{\hbar} \cdot B \cdot s \cdot \lambda
\]

\( \phi \) = rotation angle, \( t \) = time the neutron traverses the field, \( v \) = velocity of the neutron, \( B \) = magnetic field, \( m \) = neutron mass, \( s \) path length in the field, \( \lambda \) = wavelength and \( \hbar \) = Planck constant. With a mean neutron wavelength \( 0.39.10^{-9} \text{[m]} \), \( v = \hbar/m.\lambda = 1014[\text{ms}^{-1}] \). Leaving the sample, i.e. the magnetic volume, the neutron spin has made position –and path-length dependent Larmor precessions that can be analyzed with a spin-analyzer in front of a two-dimensional detector. The intensity is two-dimensionally detected as described in [11] as

\[
I(x,y) = I_0(x,y) \exp[-\int \alpha(s) ds \frac{1}{2}(1 + \cos \Theta(x,y))]
\]

\( I(x,y) \) = measured intensity in the (x,y,z) co-ordinate system, the detector plane (x,y) is perpendicular to the neutron flight direction (z-axis), \( I_0 \) = incident intensity, \( \alpha \) = linear attenuation coefficient of the
sample, s is the path through the sample. In order to perform radiography or tomography with polarized neutrons, the neutron beam has to be polarized, of course, in front and analyzed behind the sample which was done with so-called benders [16], polarizing units consisting of Silicon wafers which were coated with an FeSi substrate that were magnetized. The polarization P for the following experiments was determined with two different methods, once with the shim method, measuring the depolarization of polarized neutrons when a demagnetizing shim plate is put in the polarized beam as and by measuring the spin rotation with a spin flipper.

\[
P = \left( \frac{I_{\text{pol}} - I_{\text{background}}}{I_{\text{depol}} - I_{\text{background}}} - 1 \right) \cdot 100
\]

(4)

Niobium is a type II superconductor and the superconductivity occurs immediately after reaching the critical Temperature \( T_c \) which is in the case of Niobium 9.25K. The critical magnetic field \( B_c \) of Niobium at 0K allows an external field of 150mT to be expelled, which depends on the temperature \( T \) as

\[
B_c(T) = B_c(0K) \left[ 1 - \left( \frac{T}{T_c} \right)^2 \right] \cdot (1 - N)
\]

(5)

One has to add a factor \((1-N)\) to the formula taking into account the demagnetisation factor \( N \) which is given by

\[
N = 1 - \frac{1}{1 + \frac{q \cdot a}{b}}
\]

(6)

For \( a = 22.5\text{mm} \) (radius of the sample), \( b = 1.4\text{mm} \) (half height of the sample), \( q \) is yielded as

\[
q = \frac{4}{3\pi} + \frac{2}{3\pi} \cdot \tan \left( 1.27 \cdot \frac{b}{a} \cdot \ln \left( 1 + \frac{a}{b} \right) \right)
\]

(7)

Thus the demagnetisation factor \( N \) was for the samples 0.8837. In these experiments the external field \( B_{\text{ext}} = 6.4\text{mT} \) was for \( T = 5.5\text{K} \) well below the critical \( B_c(5.5\text{K}) = 11\text{mT} \) (see Fig.1).

![Fig.1](image)

Fig.1 The applied external magnetic field \( B_{\text{ext}} = 6.4\text{mT} \) was at \( T = 5.5\text{K} \) well below 10.7mT.
3. Experiments and results
The experimental setup is shown in Fig.2. A graphite monochromator in neutron guide reflected neutrons with a mean wave length = 0.39nm to an optical bench. The neutron beam was horizontally and vertically collimated (0.1° and 0.2°) and polarized with the bender. The overall polarization of the beam P = 85% was measured with both methods, mentioned above. The samples were 2.8mm thick and had a diameter of 45mm. The influence of the surface on magnetic flux trapping was studied by means of a special surface treatment. One of the samples got a BCP = Buffered Chemical Polishing: Hydrogen fluoride (HF) 48%, Nitric acid (HNO₃) 65%, Phosphoric acid (H₃PO₄) 85% at the ratio of 1:1:2. And therefore the treated Niobium sample was 100µm thinner than the other one which was left untreated. The samples were kept in special mountings that fitted in the cryostat (Fig.2). The samples themselves were larger than the field of view of the spin analyzer and then they had to be adjusted in the cryostat properly in order to realize images for both orientations (0° and 90°) with respect to the incident neutron beam and magnetic field, respectively.

At first the flux pinning was investigated for the untreated sample, i.e., it was cooled down with a conventional cryostat below T_c = 9.25K in the presence of an external homogenous magnetic field B_{ext} = 6.4mT, generated with two Helmholtz coils (Fig.3). At each particular cooling temperature (11K, 9K and 5.5K) T was kept constant for more than three hours to ensure a homogeneous temperature all over the sample. The exposure time for each radiograph was again three hours, flat and dark field images were recorded at the beginning and end of each series. In the “Flux-pinning-experiments” (B_{ext} = on) the samples were oriented with the surface first perpendicular (0°-orientation) and then parallel (90°-orientation) to B_{ext}. These experiments were repeated with zero external magnetic field B_{ext} = 0 for both orientations.

Fig.2 Experimental layout of the instrument PONTO (polarized neutron tomography) and sample mounting for the cryostat.

Fig.3 The Niobium sample was kept at cryogenic temperatures in an external magnetic field B_{ext} = 6.4mT, neutrons traversed the sample parallel (0°) (this case is shown in the figure) and perpendicular (90°) the surface.
If the sample was in the Meissner phase in the 0°- position the expelled magnetic field should have caused Larmor precessions of the neutron spins that should have been observed as intensity variations on the 2D detector. The path lengths were long enough to cause spin rotations of more than 2π if the expelled magnetic field would be app. 1mT (at a path length of 45mm) and less for other paths. However, this is not what we observed: The results are shown in Fig.4.

![Fig.4 Nb sample, untreated and surface treated, orientation of surface perpendicular to B_{ext} = 6.4mT, sample being in the Meissner phase for T = 5.5K, black dotted lines indicate the surfaces of the sample, black area between dotted lines shows the pure absorption contrast due to the sample; there is no difference to the 11K – images.](image)

No intensity changes could be detected in the area outside the sample. Inside the sample (black area between the dotted lines) no typical fringe pattern due to spin rotations, as observed in lead [13] were measured. The whole systems behaved in the same way as it would be above Tc. Inhomogeneities were due to the inhomogeneous field of view (FOV) of the spin analyzer. Thus (almost) all of the offered flux must have been pinned into the sample.

The sample was then rotated by 90° such that its surface was perpendicular to the incident neutron beam and the external magnetic field was switched off to measure possible flux pinning. Fig.5 shows the measured spin orientations for untreated and treated Niobium samples below and above Tc (at T = 9K and 11 K, respectively).

![Fig.5: Flux pinning in an untreated and surface treated Nb sample; images show the position dependent depolarisation of the neutron spin due to pinned magnetic fields in the bulk of the sample. Bright areas and dark areas show spin rotations of whole-number multiples of 2π. Note the rather homogeneous field distribution in the case of the surface treated sample. The feature in the 9K – image was caused by a slit in the sample due to the preparation process. BCP smoothen the edges of the slit which lead to less flux pinning. In the case of T = 11K no differences could be observed.](image)
The distinct feature in the pictures of the low temperature measurements hints at the existence of frozen flux. It is due to a cut in the sample and becomes invisible to the neutrons in the normal conducting state. The feature is much less pronounced in the treated sample, meaning that either less flux is frozen and/or the flux is distributed more homogenously.

A more thorough quantitative analysis of the results is necessary, but has not been performed yet. Qualitatively it can be stated that the BCP had a significant influence on the captured magnetic flux. It must be emphasized that the magnetic flux is measured unobtrusively as a bulk effect in these experiments. Other methods like SQUID or flux gate magnetometers can only measure the part of the flux lines that is outside the bulk.

4. Summary

In this work the suppression of the Meissner effect in superconducting niobium was investigated by using polarized neutron radiography. Below the critical temperature and the lower critical magnetic field, niobium should be in the Meissner state and an ambient magnetic field should be expelled. However, defects in the crystal lattice suppress the expulsion of the magnetic field during the superconducting transition so that the magnetic field is trapped inside the material. Radiography via polarized neutrons allows the study of the imperfect Meissner effect and has been performed on two niobium samples with different surface treatment history. Since the neutron radiography allows studying magnetic fields in bulk materials, information of the local field distribution in the samples can be derived. It is shown that both studied samples suppress the Meissner effect and that the surface has a great influence on the local field distribution of the trapped field.

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