Dendritic flux avalanches in high-quality NbN superconducting films

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***Abstract*— Niobium nitride (NbN) thin films are extensively used in superconducting devices such as single–photon detectors, hot electron bolometers, microwave resonators and kinetic inductance detectors. The operation of these devices is strongly influenced by the quality of the films, especially by their resistivity and superconducting transition temperatures (T_c). NbN films have rather high T_c of ~16.5 K and high resistivity of few hundreds micro-Ohm cm, which is perfect for operation of many superconducting devices. However, at low temperatures films are vulnerable to thermomagnetic instabilities in form of dendritic avalanches promoted by high resistivity in the normal state. Recently, new production route for NbN films has been established using high-temperature chemical vapor deposition (HTCVD). Transport measurements show low resistivity in normal state and suggest low level of lattice disorder. The highest for NbN T_c of 17.06 K was also reported in the films grown by HTCVD. According to previous study, these films should be thermo-magnetically stable. This work clarifies if it is the case and searches in one of them for dendritic flux avalanches. The nanoscale origin of avalanches is discussed.**

Keywords*— superconductivity, niobium nitride, thin films, dendritic flux avalanches, magneto-optical imaging.

I. Introduction

Niobium nitride (NbN) thin films are used in a range of superconducting electronics devices for detection and processing of signals. Among these are microwave resonators, kinetic inductance detectors, hot electron bolometers and single–photon detectors [1, 2]. NbN films are also used as components of superconducting circuits linked to devices above. The high quality of the films is a prerequisite for stable operation of the devices and circuits. It is, however, not obvious how to define high quality and, more important, what technique to use to prepare NbN films for particular applications.

A traditional technique for deposition of NbN films is reactive dc-magnetron sputtering [3]. It produces excellent films with critical temperature (T_c) of about 16 K, high critical current density (j_c) and low resistivity in normal state (ρ_n). High T_c, well above critical temperature of Nb (9.3 K), and simplicity of preparation makes NbN films attractive for superconducting electronics. Also large values of energy gap Δ ~ 2.5 meV and upper critical field B_{c2} ~ 40 T are of interest for application and fundamental research. The small coherence length (ξ (0) ~ 5 nm), on the other hand, allows preparation of ultra-thin films (≤5 nm) that are ideal for superconducting single photon detectors. It is, however, a problem for applications that most of NbN films prepared by this technique are thermo-magnetically unstable, showing dendritic flux avalanches at low temperatures and relatively weak magnetic fields of few mT. They demonstrate a fascinating behaviour [4,5], but are also an obstacle for applications.

Few measures were suggested to decrease probability of the appearance of dendritic flux avalanches. One of them is covering the edges of the films, where they are nucleated, by a layer of conductive metal [6]. In the case when it is not possible to avoid formation of avalanches, a measure of protecting particular part of the film with conducting frame was suggested in [7]. Ideally, however, would be not to have dendritic instabilities in the film at all. In [8] a NbN film intrinsically free from avalanches was demonstrated. The film was prepared by an epitaxial growth and had high T_c and low ρ_n, which are usually considered as features of high-quality films. The value of j_c in the film, however, was lower than in those showing dendritic avalanches. This could be an obstacle for some applications. So preparing ideal high-quality films with high T_c and j_c still remained a challenge.

Recently, use of the technique of high-temperature chemical vapor deposition (HTCVD) was reported for preparing NbN films [9]. The films appeared to be of exceptional quality, much better than films that were produced by other techniques. The logical step is to check if they are free from thermo-magnetic instabilities. It is what is investigated in this paper. One of the films that were analyzed in [9] was used to investigate flux distribution in it by magneto-optical study.

II. Experimental Methods

The NbN sample of a thickness of 49 nm was grown by HTCVD at 1300 °C on (0001) oriented sapphire (Al_2O_3) substrate. Synthesis details, structural and electrical characterizations of similar and this film are reported elsewhere [9,10]. Briefly, the thin-film deposition of NbN is done in homemade cold-wall vertical two-chamber quartz CVD reactor. In the synthesis chamber, $NbCl_x$ species react with NH_3 to produce NbN. The $NbCl_x$ is in-situ produced in a chlorination chamber (through the reaction of Nb with Cl_2). Deposited NbN film showed a high T_c value of 17.06 K [9].

High-resolution transmission electron microscopy revealed that the film contains face centred cubic crystal lattice as primary phase grown preferentially along the (111) orientation. Additional orientations, mainly (200), were also present in the film. To reveal the surface morphology and elemental content, the sample was investigated by scanning electron microscopy.

The main technique in this paper is magneto-optical imaging (MOI), which is an efficient technique for mapping distribution of magnetic field in superconducting and magnetic materials using Faraday effect (change of polarization of light in presence of magnetic field). It proved to be very useful for practical applications [7,11,12], but also important for fundamental studies [4,5]. Details of the technique can be found in references above.

III. RESULTS AND DISCUSSION

Fig. 1 shows magneto optical images of HTCVD NbN film at temperature of 3.7 K and four different magnetic fields: a) 0.17, b) 0.34, c) 0.68 and d) 0.85 mT applied after zero-field cooling the sample. The outline of the sample is clearly seen at field of 0.17 mT confirming that it is in superconducting state. Magnetic flux accumulates around the sample and starts penetrating inside from the edges (Fig. 1a). There is also some penetration of magnetic flux from inside the film marked by white arrow, the effect described earlier in [13]. No rapid change in magnetic field pattern is seen on this stage.

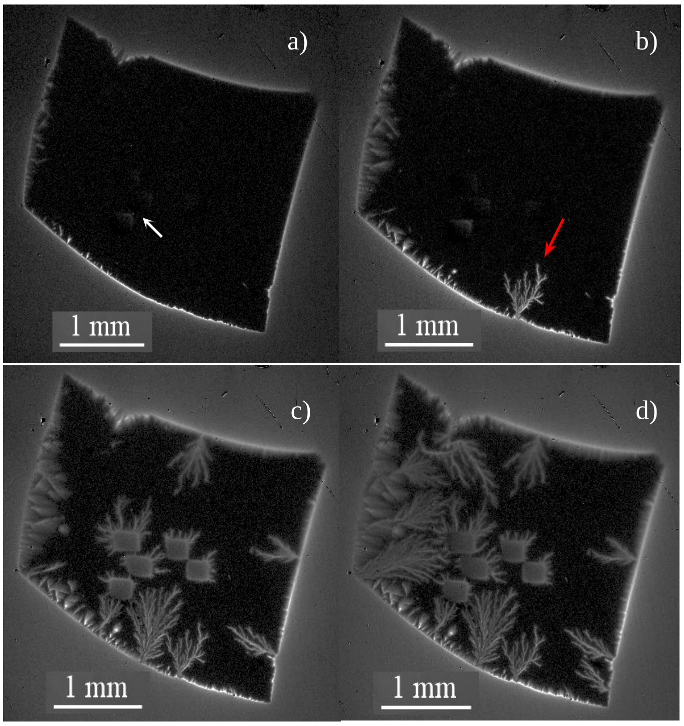


Fig. 1. Magneto-optical images of HTCVD NbN film at temperature of 3.7 K and four different magnetic fields: a) 0.17, b) 0.34, c) 0.68 and d) 0.85 mT applied after zero-field cooling the sample. White arrow in a) marks magnetic flux penetrating from inside the film into patterned rectangular holes. Dendritic flux avalanches, like one marked by red arrow in b), are evident in the film.

At magnetic field of 0.34 mT (Fig. 1b), however, first dendritic avalanche is abruptly entering the sample (indicated by red arrow). The appearance of dendrite means that film is thermo-magnetically unstable. Further increase of magnetic field to 0.68 (Fig. 1c) and 0.85 mT (Fig. 1d) produces additional avalanches. An interesting phenomenon of cascade excitation of avalanches [14] is observed in Fig. 1c. When dendrites reach the lowest of rectangular holes, it becomes instantaneously filled with magnetic field. Moreover, the hole starts emitting its own avalanches reaching neighboring holes. One by one, in a very short time, all holes become homogeneously filled with magnetic flux. According to voltage pulses measurements on similar samples [4], it is likely that this time is on the order of tens of nanoseconds.

Fig. 1d shows appearance of additional to Fig. 1c dendritic avalanches that enter mainly from the left side of the film. One of them is also reaching hole on the left, which increases number of small dendrites emitted by the holes. All additional dendrites are seen as white in differential image of Fig. 2, which is obtained by subtracting Fig. 1c from Fig. 1d. It is interesting, that dendrites on the left are entering not from left edge of the sample, but from the dark lines inside the sample that mark extended defects in the film. The bright dendrite of unusual form on the top repeats profile of a curved scratch in the sample.

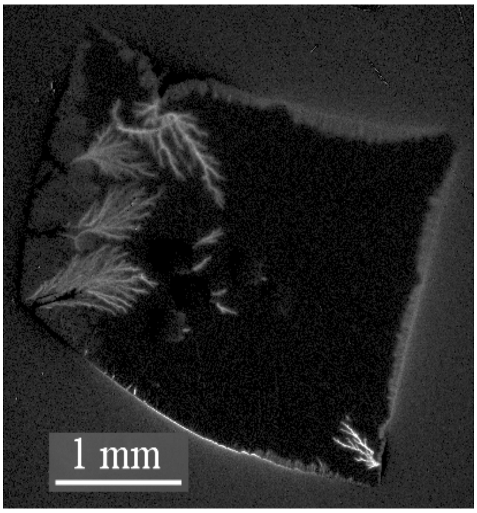


Fig. 2. Differential magneto-optical image of HTCVD NbN film obtained by subtracting Fig. 1c from Fig. 1d. The image shows additional dendritic avalanches that appear after increasing magnetic field from 0.68 to 0.85 mT. Dendrites on the left are generated inside the film.

To further investigate behaviour of dendritic avalanches, field-cooling experiment has been performed. The results of this experiment are shown in Fig. 3. During the procedure, the film was warmed to a temperature above T_c (20 K) and was cooled again to 3.7 K in the presence of magnetic field of 4.42 mT. After cooling, there was no Meissner effect or expulsion of the magnetic field from the sample seen due to high aspect ratio of the film and strong pinning that kept magnetic flux inside. The outline of the sample starts only to appear upon decrease of the field, for example, to 4.40 mT, as in Fig. 3a. The white contrast in the sample represents trapped magnetic flux. Figs. 3b and 3c correspond to further reduction of magnetic field to 3.91 and 3.74 mT, respectively.

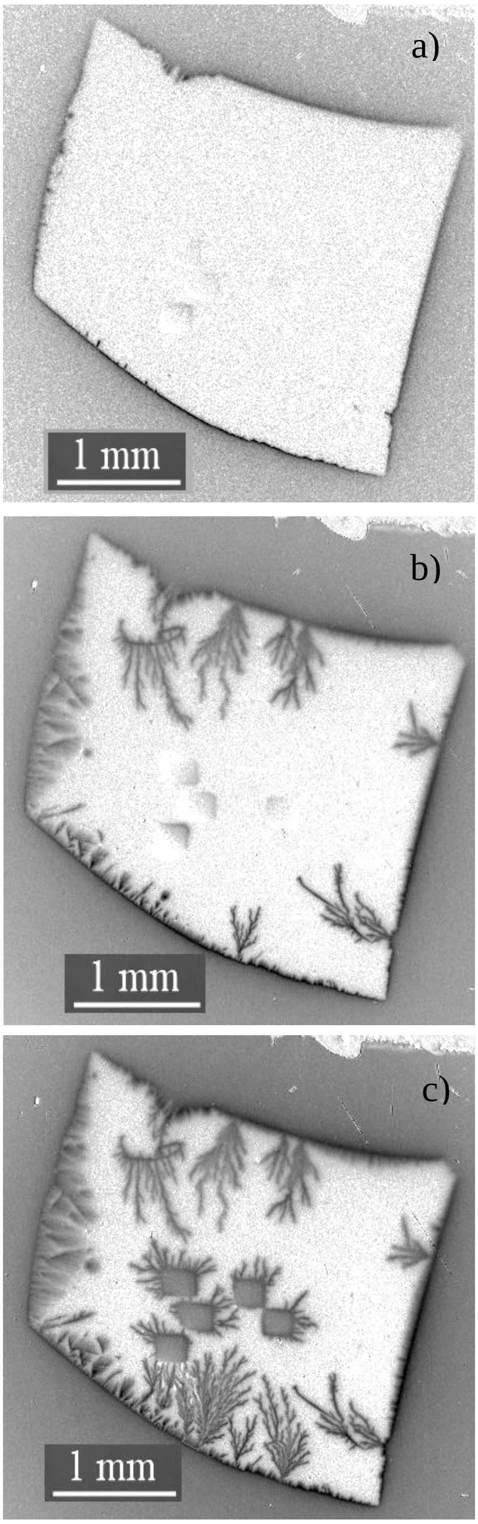


Fig. 3. Magneto-optical images of HTCVD NbN film after cooling it to 3.7 K in field of 4.42 mT and decreasing field to a) 4.40 mT, b) 3.91 and c) 3.74 mT. Anti-flux (black) dendrites are evident in b) and c).

Instead of expected exit of magnetic flux trapped in the sample, with the reduction of external field the antiflux (magnetic flux of opposite sign) enters the sample, again, in the form of dendritic flux instabilities. It makes the zero-field-cooling and field-cooling experiments symmetric. The symmetry is also in the entrance of magnetic flux inside the film (Fig. 3a and 3b) and cascade excitation of dendrites (Fig. 3c). It should be noted, however, that dendrites are never exactly repeating each other. Their branches are rarely following the same paths. An example is given in Fig. 4a.

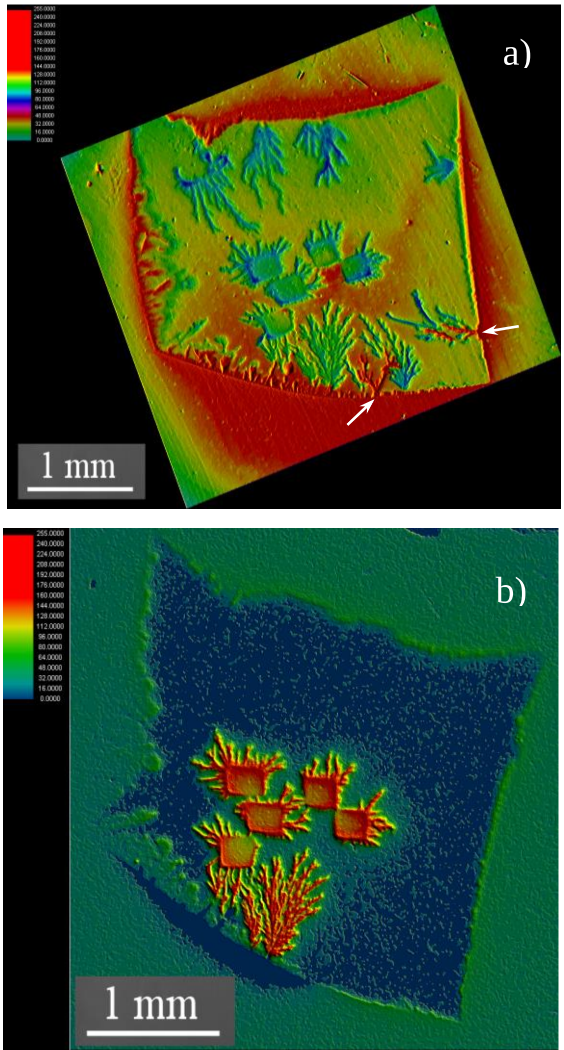
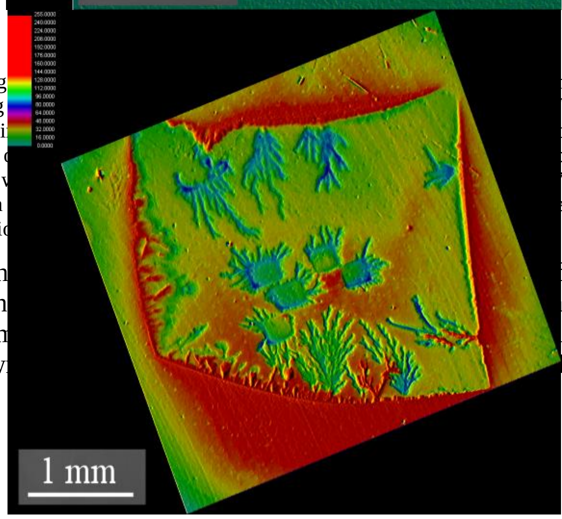


Fig ... after cooling ... T and increasi... optical image ... one of which ... 74 mT (shown ... endrite excitatio...



Th ... field-coolin ... in to 4.59 m ... arity (show ... he to the right appears exactly at the same place where previously formed anti-avalanche started. However, three branches only partially follow the branches of the anti-avalanche. The avalanche in the bottom, in contrast, is induced at a new position, but its branches partially overlap branches of two anti-avalanches to the left and right. Such an overlap is natural, since it is the easiest way to dissipate energy annihilating flux and anti-flux.

Fig. 4b is color-coded differential magneto-optical image obtained by subtracting images of field-cooling experiment, similar to those shown in Fig. 3, at fields of 3.83 and 3.74 mT. It shows isolated event of cascade dendrite excitation.

The experiments above clearly demonstrate thermo-magnetically-unstable behaviour of the film. It could be a desirable property for detection of small signals, but the obstacle for operation of superconducting electronics circuits. This behaviour is unexpected. The film has parameters such as critical temperature or resistivity much better than in any NbN films deposited previously, and according to [8], it should not show dendritic instabilities.

In [8], comparing two films with different T_c and ρ_n, it was assumed that improvement in these parameters leads to stable flux penetration into the film. In contrast, theoretical analysis in the same work showed that both films investigated in it should be thermo-magnetically stable. From this point of view, the origin of dendritic instabilities would remain unexplained. However, independent scanning electron microscopy (SEM) study in [8] showed that, seemingly in correlation with T_c and ρ_n, one of the films showed granular nanostructure, and another did not. It was the film with well-defined nano-grains that demonstrated dendritic instabilities.

Following the same idea, SEM images of HTCVD NbN film investigated in this work has also been recorded. One of them is shown in Fig. 5.

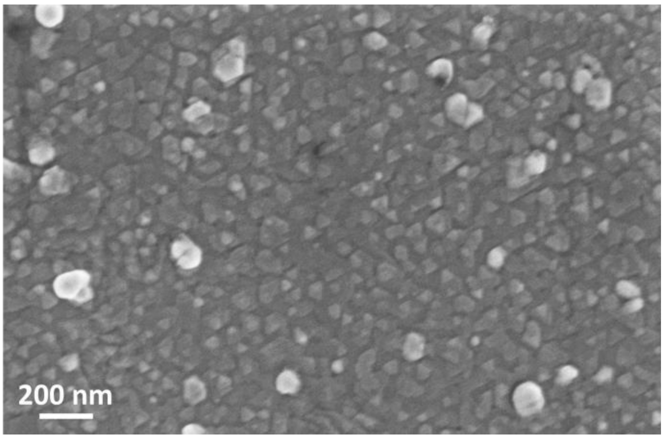


Fig. 5. A scanning electron microscopy image of HTCVD NbN film. It shows well-expressed granular nanostructure.

Despite the high quality of the film, which exhibits excellent T_c and ρ_n, SEM images of the surface, like one in Fig. 5, show a granular structure. Also, a roughness of 4.1 nm was measured by atomic force microscope in the sample (random mean square value from the area of 5 μm x 5 μm). Inside, the film contains grain boundaries that are associated with the surface grains and two in-plane orientations, as discussed in [10]. It is known that grain boundaries in NbN are structurally disordered and less conductive than grains [15]. We believe that this granular nanostructure is responsible for the formation of dendritic flux avalanches. When vortices are moving between the grains, they are able to generate high electrical field that, in combination with high current, leads to dissipation of large amount of heat. This heat, in its turn, leads to formation of dendritic avalanches. Since there are many routes along which vortices can travel between grains, dendritic avalanches never exactly repeat each other.

It is difficult to conclude how well-expressed grains are and what are the properties of grain boundaries from the values of macroscopic parameters, such as T_c and ρ_n, especially when different deposition techniques are involved. To insure that films are thermo-magnetically stable, nanostructured study needs to be performed for every deposition route. Following this rule, the subsequent nano-engineering could be very useful in designing right type of the films for right kind of applications.

IV. Conclusions

Magneto-optical study of niobium nitride thin film prepared by high-temperature chemical vapor deposition reveals a rich variety of thermo-magnetic instabilities in it in spite of its very high critical temperature and low resistivity in the normal state. Among specific features of the film is are appearance of dendritic flux avalanches in very low magnetic fields and their cascade excitation in the sample with holes. It was found that it is granular nanostructure that leads to formation of avalanches irrespectively of the values of macroscopic parameters such as critical temperature and resistivity in the normal state. Nano-engineering would control granularity and allow preparing NbN films with desirable thermo-magnetic properties.

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