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PROGRESS ON CHARACTERIZATION AND OPTIMIZATION OF MULTILAYERS

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Abstract

We present a complete study of a Multilayers Nb/MgO/NbN series with several thicknesses in order to determine the optimum thickness of the NbN layer and to compare experiments and recent theoretical advances proposed by A. Gurevich or T. Kubo. The structure and composition of the samples have been characterized structural point of view (SEM- EDX, XPS), and from the superconducting point of view (Tc, local magnetic penetration field, PCT…).

An optimum thickness has indeed been measured (close from the theoretical predictions), and the protective effect of the dielectric interlayer against avalanche vortex penetration has been evidenced.

INTRODUCTION

Although the multilayer idea was proposed for SRF applications in 2006 [1], more than 10 years later only a few group have been involved in the development of these new metamaterials. The main issue is to master a deposition technique able to produce some 10 nm films inside a cavity, and a second issue is to optimize the proposed structures with the actual superconducting parameters from these thin films which may actually differ a lot from the bulk ideal values.

From the material point of view, it is more convenient to optimize the structures on small samples, but one requires the development of specific tools to measure their performances. Systematic classical characterizations are also mandatory in hope to develop a predictive model able on the role of the particular crystalline defects present with one or another technique, and find out which one are desirable and which one should be prevented.

International Context

Up to now most of the work has been conducted on NbN, even if on paper it is not the most favourable candidate for SRF applications. Indeed NbN has proven to be a material of choice for the fabrication of Josephson junctions in superconducting electronics based on RSFQ [2]. Its fabrication on flat sample by classical techniques (e.g. reactive DC or magnetron sputtering) is well mastered.

Work on Samples: Characterization on NbN or NbTiN samples has initially been conducted at Saclay and collaborators [3-9], and at Jlab and collaborators (see e.g. [10-15]).

Early DC Squid magnetometry have shown indeed an H_{C1} increase on multilayers compared to bulk Nb, but interrogation remains to know if those results are relevant for SRF applications, since the sample is immersed in a DC field with some orientation and edge effects due to demagnetization parameter.

Similar improvement was measured on MgB_2, by LANL and collaborators [11, 16-18], thanks to an hybrid physical chemical vapor deposition technique developed by X.X. Xi at Penn State University [19]. Further work is being done at Temple university [18, 20]

Characterization tools are being developed to overcome the drawbacks of DC magnetometry.

A local magnetometer was developed at Saclay (See below and M. Aburas, [21]), where the magnetic field is applied with a coil which size is much smaller than the sample size. As the field decays quickly away from the coil, the sample can be considered as an infinite plane and no demagnetization effects occur. The same type of magnetometer is now under development at Kyoto University.

For instance in ref [18], the MgB_2 films are deposited on ellipsoids niobium substrates, so that iso-field lines lie parallel to the surface and no demagnetization effect occurs. In ref [22, 23] the same techniques is used along with μSR. μSR is a very powerful technique since it can probe the existence of a magnetic volume (i.e. field penetration) at various thicknesses, but as it is quite heavy, the turnover is slow. More detail can be found in [24]

Fast turnover techniques are being (re)developed, For instance STFC Darresbury is building a system where the sample is a tube surrounded by a coil. A magnetic probe inserted inside the tube allow to determine the field at which the whole tube transit to the mixed state (note that in this configuration, one is insensitive to vortices trapped close to the external surface) [25].

All these set-ups are dedicated to evaluate the maximum field before vortex penetration. On the other hand, it is also mandatory to evaluate the surface resistance.

Although several "sample" cavities exist or are under development ([26] and Fig. 1), they still exhibit severe drawbacks: most of them are operating at relatively high frequency to accommodate small samples. Thus they are limited by the BCS part of the surface resistance of the cavity body (usually Nb) that mask the only parameter that cannot be predicted yet: the residual surface resistance. Getting faster turnovers is also necessary.

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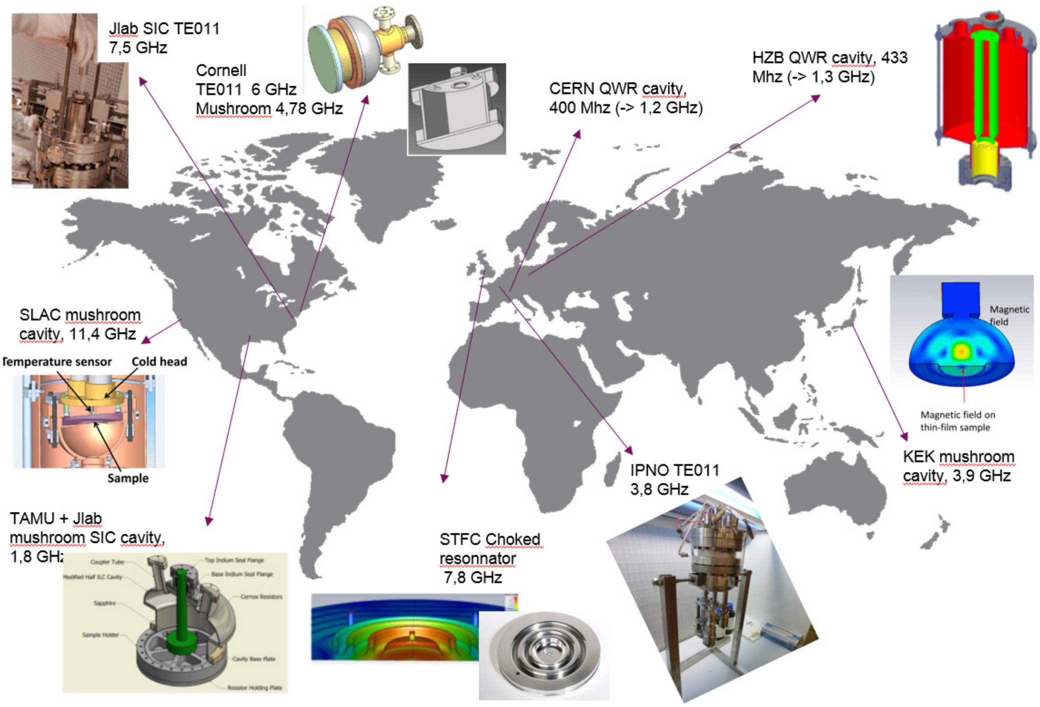

Figure 1: Sample cavities developments worldwide.

Film deposition techniques specific for SIS, like Atomic Layer Deposition (ALD) sample has been initiated at ANL [27-29], and up to now they are the only group that actually deposited ALD films inside a cavity [30]. They are now several groups that are pursuing this goal: Grenoble INP (collaboration with Saclay) [31], ODU/Jlab [32, 33], STFC [25, 34], KEK [35, 36]…

Several of these group are also exploring other deposition methods like energetic condensation [15, 37] or CVD [25, 38]. Most of these groups also develop specific measurement tools as described in the previous paragraph.

Theory. After the initial proposal by A. Gurevich, several improvement were proposed. Kubo and KEK coworkers started to include the role of the dielectric layer in the boundary conditions and showed it should keep below a certain thickness and that there was possibly an optimum thickness [39]. The initial models, based on London approximation, were further improved by Gurevich [40] and/or Kubo [41]. Reference [41] in particular shows very clearly the theoretical progression with its justifications, while in [40], Gurevich discusses the situation where no dielectric layer is deposited between bulk Nb and top SC layer; and draws the attention to the protective role of the dielectric layer against avalanche penetration of vortices.

Indeed, under the impulse of Cornell (see e.g. [42]), it was proposed that Nb cavities could just be protected against vortex penetration thanks to the enhancement of the Bean-Livingston barrier from a film with higher H_{SH} material deposited directly onto Nb and that no multilayer structure was necessary. Indeed when the calculation is done with Nb_3Sn with the simple London model, no improvement is predicted for a multilayer Nb_3Sn structure [39, 42], it is not the case when more accurate models are used [40, 41]. A slight enhancement on bilayers without dielectric is expected [40, 41, 43] and has been observed on thick Nb_3Sn and thin MgB_2 layers deposited directly on Nb [22, 44], but we hope to demonstrate experimentally in this paper that the protective role of the dielectric layer is very efficient and protects the superconductor surface from early penetration of vortex at local defects, opening the route to the use of "realistic" materials with some acceptable amount of defects.

This could have a paramount role in reducing the cost of fabrication and preparation of SRF cavities.

EXPERIMENTALS

Model Trilayers

A series of Nb/MgO/NbN trilayers layers were deposited by magnetron sputtering on Silicon wafer substrates (Collaboration with Grenoble INP and CEA Grenoble) in hope to assess the theoretical calculations. The 500 nm Nb layer is meant to mimic the bulk Nb cavity, whereas several thicknesses were tested for the thin NbN layer on the top of it. All the samples had an interlayer of ~10 nm of MgO as dielectric layer. Thicknesses of the NbN layers are also detailed in Fig. 2 shows the cut of a trilayer deposited onto the Si wafer with expected thickness "100 nm". The thickness different than what was expected: Nb is thinner (~280 nm), MgO (~ 40 nm) and NbN(~150 nm) are thicker than what was foreseen. Full characterization of the films will be published soon [45].

Note that the penetration field of the sputtered naked Nb layer was measured by Squid magnetometry as well as with local magnetometry and is about 18-20 mT [5, 8], about 10

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time less than bulk Nb, as expected for thin films with small grains and high density of defects.

Table 1: Expected Thicknesses of the Layers

Nb (nm)	MgO (nm)	NbN (nm)	T_C (K)
250†	**14**	0	8.9
250†	**14**	25	15.5
500	**10**	50	15*
500	**10**	75	14.1*
500	**10**	100	14*
500	**10**	125	14.3*
500	**10**	150	15.9*
500	**10**	200	15*

† Not same batch, deposited on the same conditions, but substrate = sapphire (actual thicknesses) - *As determined with magnetometry, see below.

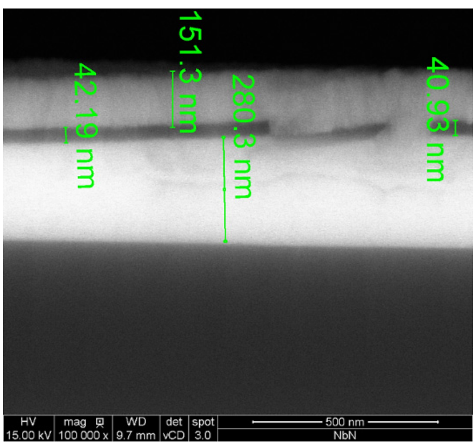

Figure 2: SEM picture of the cut of the "100 nm" trilayer: thicknesses different from what was than expected and will be fully characterized in [44].

Field Penetration

Field penetration has been measured by the local magnetometer developed at Saclay [8, 9]. Similar existing setups that are usually dedicated to irreversibility field measurement on mostly HTC materials, so they usually work in nitrogen-liquid helium, and produce fields in the 15-20 mT at best. As our facility was meant to approach cavity operation condition (T < 2K, field > 200 mT), it required the development of an innovative design (and many versions!). These developments are described in details in [46].

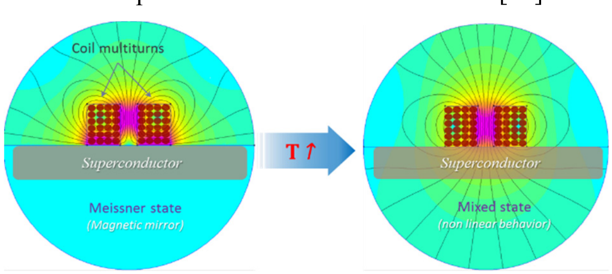

Figure 3: Repartition of the field lines in the Meissner state and in the mixed state.

The measurement principle is quite simple: an AC field (1Khz) is applied on a zero field cooled sample by passing a reference $I_0 cos(\omega t)$ current in a coils which dimension is much smaller than the sample size.

As the field decays quickly away from the coil the sample can be considered as an infinite plane. In the Meissner state the sample act like a perfect magnetic mirror and the reference signal is unaffected (Fig. 3, left).

Then the temperature is slowly increased until the sample crosses the mixed state transition and vortices start to enter the sample (Fig. 3, right).

At this point, the flux lines are pinned inside the sample and exert a dragging force on the electron inside the coil. The current in the coil is affected and starts to behave non-linearly.

By monitoring the intensity and/or the phase of the third harmonic signal (i. e. the highest intensity harmonic) one can detect accurately the transition to the mixed state in a configuration that is close to the cavity operation conditions (~parallel field on one side only). In this configuration, if the samples were devoid of defects, we could in principle measure directly superheating field. In the following, we nevertheless will refer to it as H_{C1} as a precaution, meaning the transition of the composite sample to the mixed state.

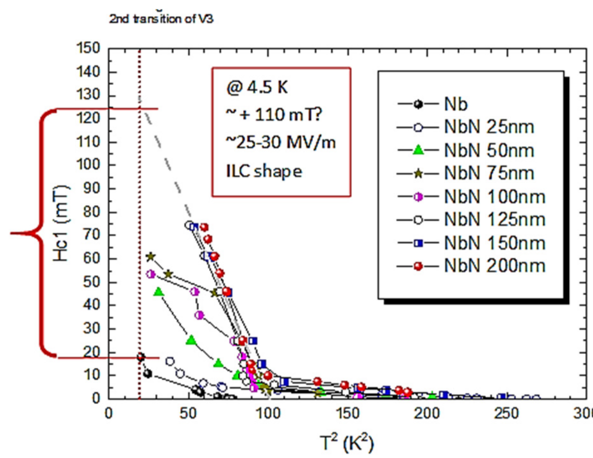

Figure 4: $\mu_0 Hc_1$ vs T^2 values for the NbN series on top of a thick Nb layer. The protective effect of the NbN layer increases with thickness until a saturation is observed, starting around 125-150 nm of NbN.

Figure 4 shows the results of H_{C1} (actually $\mu_0 Hc_1$ when expressed in Tesla) for the whole series of Nb/MgO/NbN trilayers ranging from 25 to 200 nm of NbN top layer. One observe an increase of H_{C1} with thickness, until one reaches saturation around 125-150 nm.

In the present configuration, the local magnetometer is capable to produce up to 150 mT on the sample, but it is very difficult to get a thermal stabilization higher than 80-100 mT. A new design is under construction to check the behaviour of the sample between 80 and 200 mT.

ith the present results, if one fit the experimental values, one can see that for the150 nm NbN layer one expect the field to enter the SC structure nearly 110 mT higher than

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naked Nb (equivalent to 25-30 Mv/m for ILC shaped elliptical cavity). If these results get confirmed on bulk Nb, the field could nearly be doubled with the deposition of one single layer, which is quite simple to achieve.

Comparison with Model

This part of the work will be detailed in [43]. We just give below two examples that show how far the behaviour of trilayers are affected by the materials properties.The pictures below show calculation from T. Kubo (personal communication and in [43]).

The first one (Fig. 5) is the estimation of the maximum field achievable on a "perfect" bulk Nb with $\mu_0 H_{C1}$ ~170 mT, while the second one (Fig. 6) is calculated with a "degraded" Nb with low mean free path (~5.8 nm) and $\mu_0 H_{C1}$ ~50 mT. The calculation includes an η =0.4 factor which takes into account possible surface defects that promote early vortex penetration as described in [41].

In the case of an ideal material, the deposition of a top dielectric + NbN layer is expected to increase the field from ~ 170 mT to 250 mT (nearly 50 %).

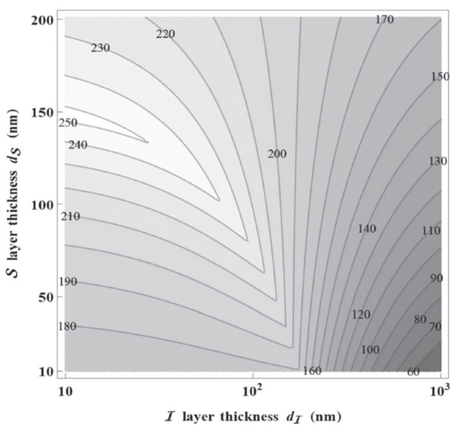

Figure 5: B_{max} of a NbN–I–Nb system in mT, calculated with B_C = 230 mT and λ_1 = 200 nm for NbN, B_{max} = 170 mT and λ_2 = 40 nm for the Nb substrate (from [41]).

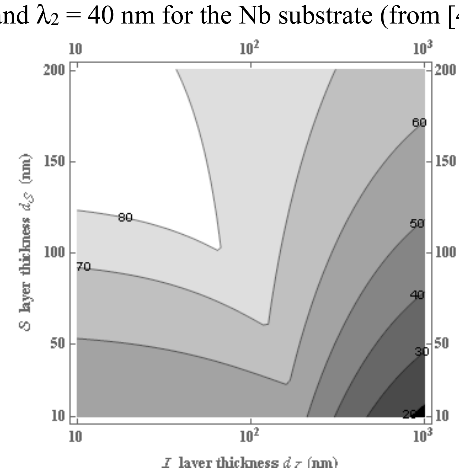

Figure 6: B_{max} of a NbN–I–Nb system in mT, now calculated with B_{max} = 50 mT and λ_2 = 112 nm for the Nb substrate. The calculation includes an η =0.4 factor which takes into account possible surface defects that promote early vortex penetration.

When the calculation is done with the "degraded" Nb, the expected field enhancement is much lower, and the optimum thickness range is much larger, similarly to our experimental observations where the 125,150 and 200 nm NbN layers exhibit the same field enhancement behaviour.

Role of the Dielectric Layer

Note that the 3rd harmonic signal vs T in a classical bulk superconductor affect a bell shape; going through a maximum when all the vortices are trapped, and then decreasing as the temperature further increases and the vortices are progressively unpinned and start to move freely with the field (this experiment is also an indirect way to measure pinning force).

In our experiment on multilayers, we observe a similar behaviour only at low field. At low field the signal takes a ~Bell shape similarly to what is observed on individual superconductors (bulk or thin films).

In this case the transition is above 9 K, which means only NbN is superconducting and obviously the Nb underneath is not affecting the NbN behaviour (if one fit this part of the curve we get an H_{C1} value of a few mT, similar to NbN thin film alone [4]).

At higher field, the transition appears below 9K, when both superconductors were initially in the Meissner state, in the region where H_{C1} enhancement seems to be effective. The observed behaviour is very different (Fig. 7). The transition appears to be dramatic, as if all the vortices entered the material in one shot, at a delayed temperature.

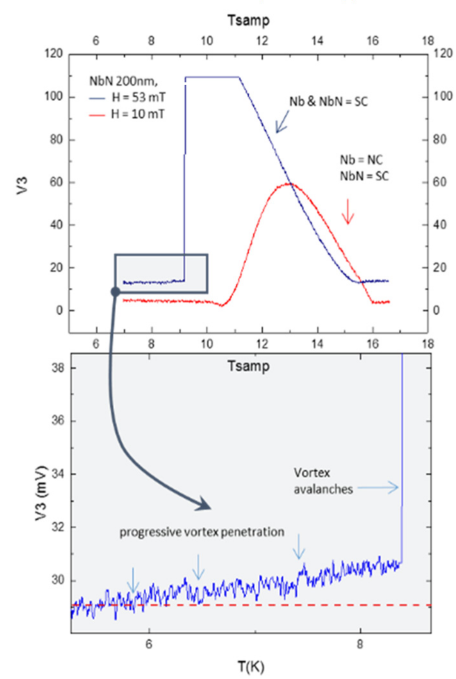

Figure 7: example of 3rd harmonic signals at 10 mT (red curve) and 53 mT (blue curve) and detail below (see text).

In fact if one look in detail the first transition occurred earlier (the earlier signal can also very clearly be observed in the phase signal), but apparently only a few vortices enter the sample, and the dissipation due to their pinning keeps low. Then the second, dramatic transition occurs.

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We interpret these results with the scenario proposed by Gurevich in [40]: that the dielectric layer blocks the first vortices entering the superconductor until an avalanche penetration occurs (see Fig. 8).

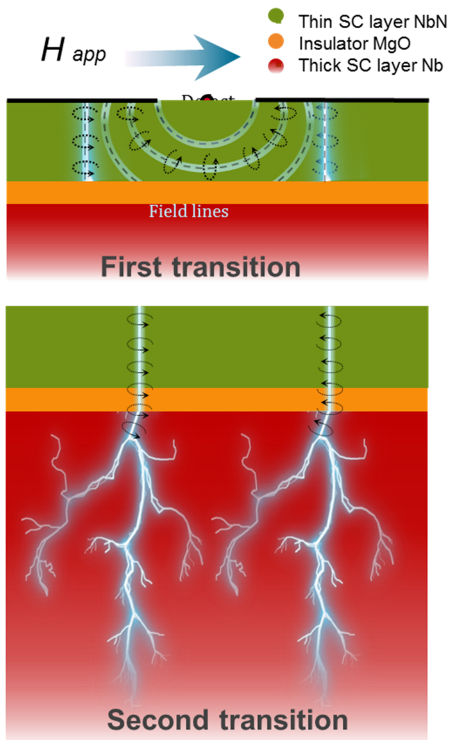

Figure 8: scenario for the first and second transition observed on the 3rd harmonic signal.

CONCLUSION

If superheating field is the ultimate limit for ideal material, in real life was is often limited by local defects that promote early penetration of vortices. The risk of flux jumps (or avalanche penetration corresponding to the entrance of several millions flux quanta) being aggravated in transient states [47], those small defects are all liable to trigger an early quench. Concerning Niobium, we now master the fabrication well enough to be able to get high performance material (most of the time), but one can fear that more complex material like e.g. Nb_3Sn or MgB_2, will be more difficult to produce defect less. Multilayer structure provide an alternative route since the protection brought by the presence of a dielectric layer open the possibility to use realistic materials with a certain amount of defects.

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