

## THIN FILMS ACTIVITIES IN THE IFAST PROGRAM

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### Abstract

Now that bulk Nb technology has reached its full maturity, improving SRF technology demands that new materials need to be developed. For reasons explained in the talk, all next generation SRF materials will be in the form of thin films. The IFAST project has the ambition to coordinate European activities on that topic, not only throughout its own program (that will be presented here), but also by keeping in touch with all actors worldwide, with the hope of developing a more efficient collaborative actions in a limited funding context. In this paper, we will present the challenges presented by the development of new thin films materials, each developed for tailored applications and the main research direction proposed by the thin film community.

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### TAILORED MATERIAL FOR SRF

The SRF technology is mostly based on ultra pure bulk niobium, which is not optimized to maximize its superconducting properties (surface resistance), but rather to maximize thermal stabilization of dissipating defects. By separating each functions (mechanical structure, thermal transfer, surface resistance, surface protection...), one can achieve superconducting cavities with enhanced performance (Fig. 1). One can even hope to tune their performances for specific applications.

This process is already "en marche". For instance, the "doping procedure" proposed by FNAL [1] consist in diffusing interstitial atoms in a shallow part of the surface.

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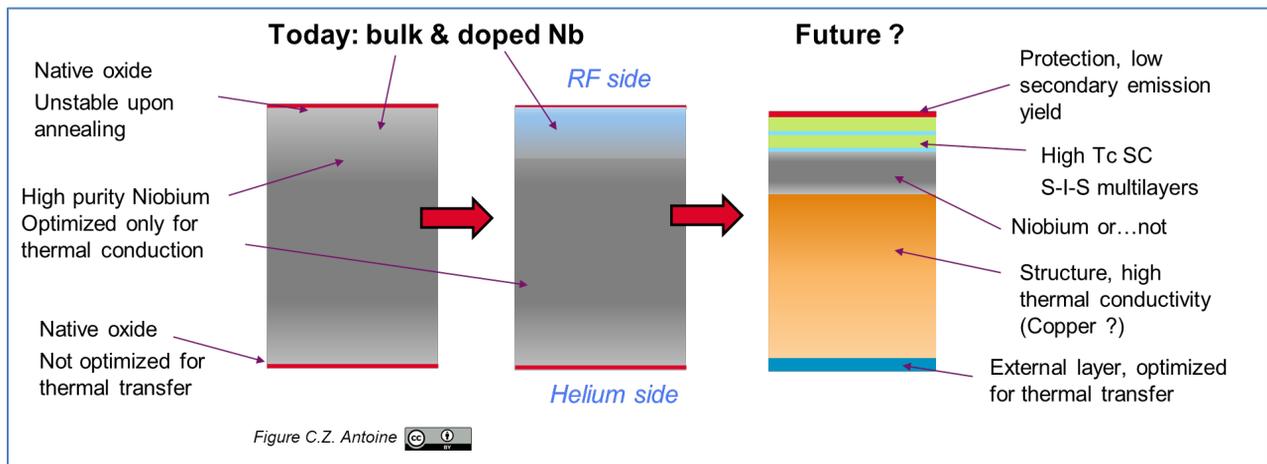


Figure 1: Expected evolution of the functionalization of SRF materials.

It is sufficient to tune the superconducting properties of the cavities' inner surface without affecting the bulk thermal conductivity. Replacing the external part of the cavity by a copper, as a highly thermally conducting and mechanical support, keeping only a thin niobium layer at the inner surface has been tried for decades, but it is only recently, with new deposition processes that dense enough films have been achieved, which exhibit improved performances.

### Technological Challenges

Improving RF technology presents huge challenges in material science. Indeed, when one deals with classical copper cavities, the main requirements on the material are based on metallurgy, a science that started to be explored by humanity 6000 years ago. When one switches to niobium, still a pure metal, one has to face new challenges. The main one arises from the fact that the penetration depth of the field is  $\sim 40$  nm, hence everything that happens on surface starts to become of paramount importance. Surface science, especially at the nm level was much more recently mastered.

Then higher  $T_c$  materials are all compounds, so it also requires mastering the chemical aspects in order to get the proper superconducting phase. Most of these materials are brittle and cannot be considered to build bulk cavities. They have to be deposited in "thick" ( $\mu\text{m}$ ) or "thin" (nm) films. Here again, Physical Vapor deposition (PVD) techniques or Chemical Vapor deposition (CVD) techniques have been mainly developed in the course of XX<sup>th</sup> century and are still in development.

The parameter space to be explored is vast, and needs a substantial investment in material science, as has been done in the superconducting magnet community over the past 70 years.

The understanding of the physics of superconductors for magnet applications has led to tremendous progresses and opened a large domain of applications.

Unfortunately, the requirement for SRF materials are literally opposite to the requirement for magnets applications, and somehow the exploration has to be started over, but in a very different direction.

### Type II Superconductors, Domains of Application

Actually thousands of superconductors (SC) have been listed but only a dozen have found applications. They are all type II. In short, magnets use type II superconductors in the mixed state. Defects are voluntarily introduced to enhance the critical current density, which in turn decreases the transition between the Meissner and mixed state (Fig. 2) by reducing the lower critical magnetic field,  $H_{C1}$ .

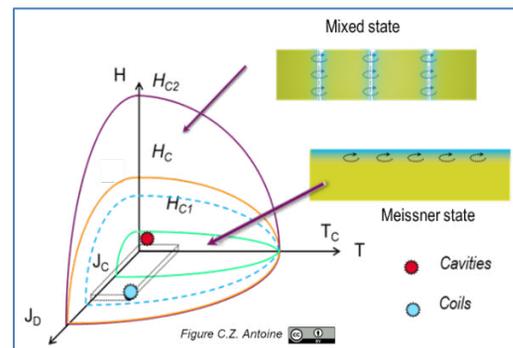


Figure 2: Meissner and mixed state. At low magnetic field, supercurrents screen the external magnetic field and the SC is in the Meissner state. Above  $H_{C1}$ , it is energetically favourable for the magnetic field lines to enter the SC as normal conducting zones surrounded by screening currents, while the rest of the material remains fully superconducting; this is the mixed state.

In RF, the mixed state is too dissipative, and cavities must be kept in the Meissner state. It means reducing the density of defects that could promote early entry of the field lines (vortex), characteristic of mixed state. Niobium it the material with the highest first critical field  $H_{C1}$ , which explain why it has become the material of choice for SRF applications [2].

### Superheating Field and Multilayer Concept

In fact, in RF cavities, where the magnetic field is parallel to the surface, it is difficult to nucleate a vortex. This configuration helps to maintain the Meissner state as a metastable state above  $H_{C1}$  up to the “superheating field” in theory (Fig. 3). This rationale is used to predict the maximum accelerating field in cavities.

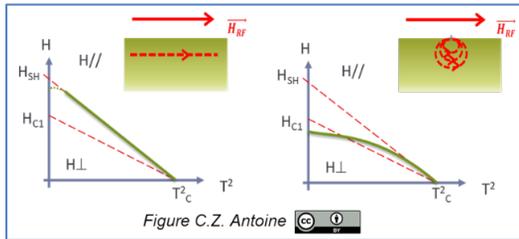


Figure 3. Vortex penetration in parallel field, without (left) or with (right) defects. The green curves are the actual transition field in the projection of the phase diagram from Figure 2 in the  $H$  vs  $T^2$  dimensions.

In realistic condition, complex materials tend to exhibit many defects which can prevent the superheated state to be maintained.

In 2006, A. Gurevich proposed a new multi-layered structure that could overcome that issue [3]. If one places a dielectric layer a few 10s of nm below the surface, it will break any vortex loop in a vortex plus antivortex that coalesce together within a few RF periods (see Fig. 4). Moreover, if the top layer is a superconductor with a higher  $T_c$  than Nb, the surface resistance will be lower. Limiting its thickness to a few 10s of nm (i.e. below its field penetration depth  $\lambda$ ) it a way to artificially enhance its transition field [4]. With this structure, one becomes less sensitive to defects [5], and it is one of the ways that are explored to get higher performances, as described below.

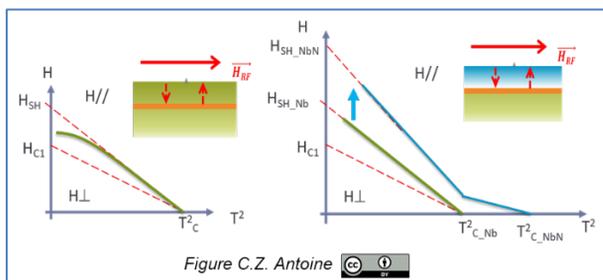


Figure 4: Effect of a dielectric layer and multilayer concept. With multilayers, one can both gain on the quality factor and the accelerating field.

### IFAST THIN FILM ACTIVITIES

The WP9 from IFAST, “Innovative superconducting cavities”, is focused on improving performance and reduce cost of SRF acceleration systems based mostly on the use of thin films. It comes after several European projects on the topics (WP12.2 within Eucard2, WP15 within Aries) that helped bringing together the few teams working on that topic in Europe and keep in touch with the international community [6-8].

The European members are from France (CEA, CNRS), from Germany (HZB, USI), from Italy (INFN, PICCOLI srl), from Latvia (RTU), from Slovakia (IEE), and from United Kingdom (UKRI), but we have also external collaborators, both formal (JALB, PTI, MEPHI) and informal (CERN, DESY).

One part of the job (task 9.1) is contributing at building together a global strategy to be able to produce Superconducting RF (SRF) cavities coated with a superconducting films, and participating to the corresponding chapters (thin films) initiatives like e.g. the Snowmass propositions [9] or the European Accelerator R&D Roadmap Implementation [10].

Functionalizing SRF material requires 4 main actions:

- Mastering thin film deposition techniques in terms of final composition and structure.
- Adapting known deposition techniques to the internal complex shape of the cavities (not always compatible with standard techniques).
- Mastering interfaces quality (substrate preparation, interlayers).
- Finding the proper compromise between optimum superconducting quality and fabrication cost (choice of the superconducting material).

Past projects have shown promising results at least on flat samples. The objective of IFAST is to pass from developments on samples to the first RF prototypes and merge all the developments that have been mastered over the last years.

Among the recent achievements, here are the most compelling steps:

- Nb thin film layers with performance close to bulk Nb (mitigation of the Q-slope, high transition field) were observed at CERN [11, 12] and at JLab [13, 14]. It opens the route to Cu cavities deposited with functionalized layers on the top of a thick Nb film. The quality of the substrate (Cu) appears to have a paramount importance on these performances [15-17].
- Bulk niobium cavities deposited with high  $T_c$  material like  $Nb_3Sn$  start to exhibit very high  $Q_0$  opening the route to operation at higher temperature and alternate cooling schemes. These higher  $T_c$  materials are very sensitive to defects and do not reach high fields yet [18, 19]. Successful alternative fabrication routes have been explored on samples (direct deposition on copper [19-22], bronze route...).
- The multilayers concept has proven to be effective both in increasing the penetration field of vortices (which drives the maximum accelerating field), and reducing the surface resistance [5]. Moreover, the “protective” effect of such structures opens the route to more realistic materials and less sensitivity to defects, including  $Nb_3Sn$  or  $NbTiN$  multilayers.

### General Strategy

The general strategy (Task 9.1 of WP9) consist into pursuing the optimization and the industrialization of key steps:

- Substrates preparation (Nb, Cu), e.g. Plasma Electropolishing (PEP) developed at INFN, metallographic polishing(developed at CNRS), pre-and post-treatment (laser at RTU, flash annealing at HZDR).
- Production of seamless copper cavities as the risk of poor film deposition at welding has been assessed [16]
- Optimization of deposition techniques: Energetic deposition techniques, atomic layer deposition (ALD)... to get Nb, NbN, Nb<sub>3</sub>Sn, V<sub>3</sub>Si... thick films (μm) and/or SIS Multilayers (nm)
- Firstly producing and RF testing prototypes of SRF cavities at 6 GHz. These cavities are easier to fabricate, handle, and dissect... so that fast feedback can be provided.
- Finally producing accelerator type 1.3 GHz cavities (as a feasibility assessment). 1.3 GHz cavities present the advantage that they allow evaluating both residual and BCS resistance.

There is also a strong necessity to develop advanced characterizations tools to be able to measure superconducting properties in condition close to cavity operation, a condition which is not available with conventional techniques.

### *Seamless Elliptical Cavities (Task 9.2)*

Producing seamless copper substrate is mandatory. Many 6 GHz cavities are required for destructible tests during the optimization stage; automated production of 1.3 GHz cavities will be necessary at the prototyping stage. In the task 9.2, the goal is to switch from a semi-automatic to a fully automatic process using a CNC machine. The work is developed in collaboration between INFN and the company Piccoli. The process is now assessed, including the optimization of the annealing temperature. Up to now, twenty 6 GHz cavities and three 1.3 GHz cavities have been produced and distributed among collaborators for further deposition. Further improvement of the automation is regarded [23, 24]. Surface treatment of copper is also an important part of the process [15, 25]

### *From Samples to Cavities (Task 9.3)*

**Thick Nb layers on copper.** As mentioned earlier, getting bulk like performance on Nb films is an important step. Activities have been conducted at UKRI, INFN and USI [6, 8, 12, 17, 26-28].

**Higher Tc materials.** Because the material of choice will combine enhanced superconducting properties with relative easiness to produce it, several higher Tc materials are still in the optimization process: Nb<sub>3</sub>Sn, NbTiN, NbN, MgB<sub>2</sub>... They are complex (compound) materials, composition needs to be adjusted to get best SRF performance, and then the optimized recipes need to be adapted for complex geometries [29]. Deposition set-ups for 6 GHz cavities have been designed, build and commissioned at INFN, STFC and USI [30], along with specific developments on sputtering targets [31].

First attempts of deposition in 6 GHz cavities are undergoing [22][32].

In between, the work on flat sample allow assessing their structures and evaluate their RF properties on QPR flat samples (see below). A specific deposition set-up was built in Legnaro, while the existing set-up was used at USI .

Deposition of thick films and multilayers structures are studied in parallel. Nb/AlN/NbN have been deposited on bulk and copper QPR supports.

### *Surface Engineering with ALD (Task 9.4)*

Atomic Layer Deposition is a particular technique based on chemical reaction of precursors adsorbed on the reactor surface (which is the cavity itself). It is a highly conformational technique particularly well adapted to complex shapes encountered in our domain, and it is easily scalable to industrial production.

A wide range of compounds are manageable in the same deposition set-up so that in situ composite fabrication is achievable.

A 1.3 GHz deposition set-up has been constructed so that results obtained on samples can be now tested on actual cavities.

Three types of functionalized layers are being explored:

- SIS multilayers: good quality NbTiN/AlN multilayers have been deposited on Nb samples with a Tc between 14 and 15.5 K after a final annealing. A first 1.3 GHz has been deposited with the same recipe [33].
- Dielectric surface engineering and doping. The native oxide on Nb is defective. It can be replaced by e.g. Al<sub>2</sub>O<sub>3</sub> deposited by ALD followed by an annealing. The process has been tested on 2 1.3 GHz cavities with an observed increase of the quality factor [34].
- Low secondary yield cap layer. The secondary electron yield (SEY) is at the origin of the multipacting phenomena. Depositing a thin capping layer by ALD (~10 nm) proved to decrease the SEY on both samples [35] and cavities [36].

### *Surface Engineering with Heat (Task 9.5)*

Among the pre- and post- treatments of substrates as well as films, laser and flash lamp annealing are being explored. Both treatment are very superficial, which is a plus since copper has a very low melting point and some of the films require higher temperature treatments. Only surface is affected, and these heat treatments are liable to smooth the surfaces [37-39], help to recrystallize, improve films adhesion and decrease porosities [27]. Such surface treatment could also be used to stabilize the high temperature phase of A15 compound (with the highest Tc) or built specific alloys [40]. Here also set-ups adapted to cavities treatments are under development at RTU and HZDR.

### *QPR Cavity: RF Evaluation (Task 9.6)*

Material characterization techniques, even the most advanced ones, are still not predictive of future RF behaviour. The development of a QPR cavity at HZB [41, 42] has permitted to start to explore RF behaviour at 3 different frequencies on flat samples, small enough to be easy to handle,

It has allowed the measurements of Nb, NbN and NbN multilayers developed by HiPIMS at USI [43-48]; Nb and Nb<sub>3</sub>Sn and multilayer structures prepared by DCMS or HiPIMS at STFC, and Nb and Nb<sub>3</sub>Sn thick films prepared by DCMS at INFN. Preparation of bulk Niobium like PEP (developed at INFN) or metallographic polishing (developed at CNRS [49]) are also under study [43, 44, 50].

### Material Characterization

The development of complex material requires thorough characterization tools. Classical material characterization: optical and confocal microscopy, SEM, EDX and EBSD, Ion beam miller for cross-section, X-Ray, TEM, and basic superconducting properties are measured: T<sub>c</sub>, RRR, DC magnetometry, AC susceptibility.

In addition, specific original characterization tools have been developed to measure the superconducting samples behaviour in condition closest to the operating cavities condition: Tunnelling microscopy (Superconducting gap, density of superconducting states cartography), flux penetration measurement set-ups (at UKRI [51] and CEA [52]), Surface resistance on small sample (7.8 GHz cavity at UKRI).

### CONCLUSION

Thin films SRF activities are still conducted in a few small groups, with few resources. Coordination and exchanges help to derive maximum benefit of the vast space parameter to be explored.

The next generation of SRF material is “en route” with already very nice results on samples, and early progress on R&D cavities. We hope IFAST WP9 (and collaborators) will bridge the gap between lab R&D and 1<sup>st</sup> prototypes development. If successful this step will prove that a full change of technology is possible after more than 50 years of bulk Nb domination.

If accelerator community wants SRF technology to evolve in that direction, strong investments are needed in the near future.

To end on a positive note, several of the techniques underdevelopment are liable to be applied on bulk Nb cavities, opening the route to future up-grades of existing machines.

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