MATERIALS FOR SUPERCONDUCTING CAVITIES

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Abstract
The ideal material for superconducting cavities should exhibit a high critical temperature, a high critical field, and, above all, a low surface resistance. Unfortunately, these requirements can be conflicting and a compromise has to be found. To date, most superconducting cavities for accelerators are made of niobium. We shall discuss here the reasons for this choice. Thin films of other materials such as NbN, Nb_3Sn, or even YBCO compounds can also be envisaged and are presently investigated in various laboratories. We show here that their success will depend critically on the crystalline perfection of these films.

1. INTRODUCTION

The advantages of superconducting cavities over normal conducting cavities are well known. These advantages can be exploited in many different ways since they permit continuous operation of the accelerator, improve the energetic conversion to the beam, relax the constraints on cavity design and minimize the cavity impedance seen by the beam. According to the accelerator characteristics, e.g. duty cycle, intensity, beam dynamics properties, a different priority is given to each of these advantages. Nevertheless, it should be remembered that all the advantages of superconducting cavities stem from one single property of the superconducting material, namely its very low *surface resistance.* We shall discuss first the requirements imposed on the superconductor by this criterion of minimal surface resistance.

2. CRITERIA OF CHOICE DERIVED FROM THE SURFACE RESISTANCE

The power dissipation per unit area of a superconductor in RF regime is related to the surface resistance of the material R_s via:

$$P = \frac{1}{2}.R_S.H^2, \qquad (1)$$

where H is the RF magnetic field amplitude. A well known expression for the surface resistance of a superconductor can be derived from the two-fluid model [1]:

$$R_S = \frac{A}{T}.\sigma_n.\omega^2.\lambda^3.e^{-B.T_c/T} + R_{residual}, \qquad (2)$$

where A and B are two constants which depend only weakly on the material, ω is the RF pulsation, σ_n the normal state conductivity of the material, λ its effective penetration depth, and T_c the superconductor critical temperature. The first term, improperly called R_{BCS}, corresponds to the dissipation due to normal electrons, and the second, $R_{residual}$, is associated with imperfections in the superconductor structure and behavior.

In many practical cases, the BCS term takes a non negligible fraction of the total surface resistance. As can be seen from Eq. (2), R_{BCS} depends strongly on the superconductor penetration depth and critical temperature. It is thus crucial to maximize T_c (requirement 1) and to minimize λ (requirement 2).

In Eq. (2), the penetration depth λ is an effective value, related to the London superconductor penetration depth λ_L by $\lambda = \lambda_L \cdot \sqrt{\frac{\xi_0}{\xi}}$, where ξ_0 and ξ are the coherence lengths in the pure and real material respectively. In the real material, the coherence length is given by

$$\xi^{-1} = \xi_0^{-1} + l^{-1},$$

where l is the electron mean free path. Two extreme cases can then be envisaged:

For clean superconductors, i.e. those with a large electron mean free path, $l >> \xi_0$, thus $\xi \approx \xi_0$, and Eq. (2) gives $R_{\mathrm{BCS}}^{\mathrm{clean}} \propto l$. For very clean material, Eq. (2) is no longer valid, and more sophisticated calculations based on BCS theory [2] predict a roughly constant surface resistance, independent of l, and thus, of the material purity.

For dirty superconductors, i.e. those with a small electron mean free path, $l << \xi_0$, thus $\xi \approx l$, hence $R_{\mathrm{BCS}}^{\mathrm{clean}} \propto l^{-1/2}$. The surface resistance thus diverges for very dirty superconductors (Fig. 1).

Between the clean and dirty limits, R_{BCS} takes a minimum value when the electron mean free path becomes comparable to the coherence length.

Pure metals and pure intermetallic compounds with a well defined stoechiometric composition, like Nb_3Sn, are usually clean superconductors, except if they have many crystalline defects; on the other hand, alloys enter the category of dirty superconductors, due to their very small electron mean free path. Consequently, they display large BCS surface resistance. For the same reason, i.e. a small electron mean free path, alloys also have a poor thermal conductivity at cryogenic temperatures, thus hampering the thermal stability of a cavity. For these two reasons, alloys are not suitable materials for superconducting cavities.

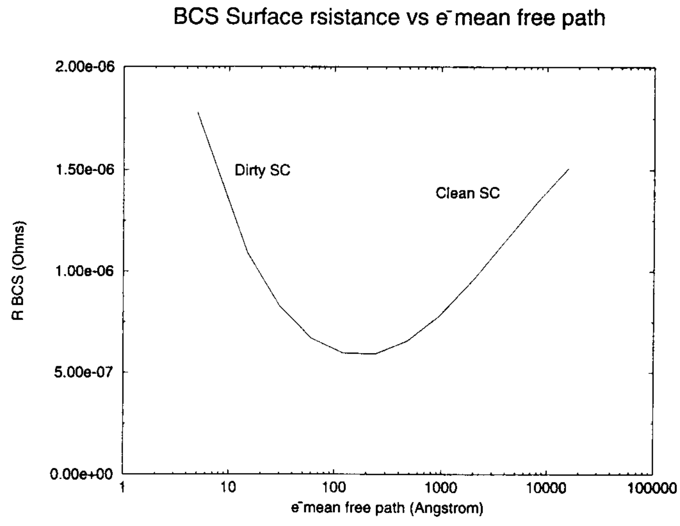

Fig. 1 The BCS surface resistance of niobium at 1.5 GHz and 4.2 K, as a function of the electron mean free path.

Usually, the optimum working conditions of the superconducting cavity are met when $R_{\rm BCS} \approx R_{\rm residual}$. It is therefore very important to minimize this last term. Many causes contribute to the residual surface resistance [3]. Some of these are extrinsic (trapped flux), and can be avoided. Some causes are intrinsic and are due to structural imperfections of the material, like inhomogeneities, grain boundaries or surface serration. The superconducting wave function or order parameter is sensitive to defects larger or of the same size as the coherence length ξ [4]. Materials with a large ξ–value will thus "forgive" large defects without an appreciable increase of the residual surface resistance. This is clearly a desirable feature for cavity applications where square meters of superconducting surface are exposed to the RF field, and are presumably difficult to prepare completely "defect-free".

Putting requirements 2 (small penetration depth) and 3 (large coherence length) together, we get the description of a type I superconductor. These are universally known as low T_c superconductors, and this is clearly in contradiction with requirement 1. The BCS theory [2] gives a relationship between the coherence length and the critical temperature:

$\xi_0 = 0.18 \frac{\hbar . v_F}{k . T_c}$, where v_F is the Fermi velocity. The inverse relationship between the coherence length and the critical temperature indicates that the contradiction between requirements 1 and 3 is very deep indeed. Therefore, the ideal superconductor for RF applications does not exist, and subsequent choices clearly result from a compromise.

Lead, as an archetype of a type I superconductor, has been used for low frequency cavities, and has yielded a very low residual surface resistance. It is cheap, and easily available in a pure form. Unfortunately, at frequencies higher than a few hundred MHz, the BCS surface resistance becomes prohibitive, due to the low critical temperature of this material. Moreover, it has poor mechanical characteristics and oxidizes easily, with a subsequent degradation of the properties of the superconducting surface. For these reasons, lead tends to be progressively replaced by niobium, and is now confined to low frequency applications.

Type II superconductors can have a large T_c and a reasonably small penetration depth, so that their BCS surface resistance can be small, even at rather high cryogenic temperature. But their coherence length ξ is small, so type II superconductors tend to display rather high residual surface resistance, unless they are prepared "defect-free" (Fig. 2). Table 1 summarizes the characteristics of various superconductors.

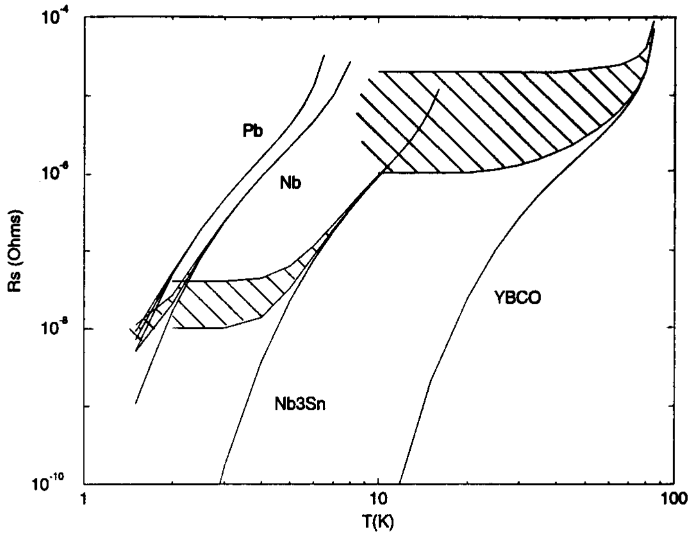

Fig. 2 The surface resistance vs temperature for a few typical superconductors at 1.5 GHz. In this diagram, the full lines show the BCS contribution, and the hatched areas represent the total surface resistance for "state-of-the-art" materials.

Table 1
Characteristics of various superconductors

Material	T_c (K)	λ (nm)	ξ_0 (nm)
Pb	7.2	39	83-92
TYPE I ↑			
TYPE II ↓			
Nb	9.2	32-44	30-60
$Nb_{0.6}Ti_{0.4}$	9.8	250-320	4
NbN	15-17	200-350	3-5
Nb_3Sn	18	110-170	3-6
YBCO	94	140	0.2-1.5

3. CRITICAL FIELD

High magnetic fields are present in accelerating cavities. In many cases, the amplitude of the RF magnetic fields approaches the order of magnitude of the superconductor critical fields. A superconducting material with a high critical field is thus desirable for RF applications (requirement 4). Since the nucleation time of a vortex is usually large compared with the RF period, the relevant critical field of an RF superconductor is not the usual critical fields H_{c1} or H_{c2}, but is believed to be the superheating field H_{sh} [5]. H_{sh} is related to the thermodynamic critical field H_{th} via:

$$H_{sh} = 0.89.\frac{H_{th}}{\sqrt{\kappa}} \qquad \kappa << 1$$

$$H_{sh} \approx H_{th} \qquad \kappa >> 1$$

It should be noted that for type II superconductors, H_{sh} can be significantly larger than H_{c1}.

Theoretical arguments in favor of the hypothesis that the limiting RF field is H_{sh} are supported by the fact that experimentally, the superheating limit has been approached, but never broken [6]. Table 2 gives the thermodynamic field, superheating field and maximum attained RF field for various superconductors.

4. NIOBIUM

In view of the above criteria, Nb appears as a serious candidate for superconducting cavities. It has the highest T_c and H_{sh} of all pure metals. Being a soft type II superconductor, it occupies a position of compromise between the four requirements mentioned above.

Niobium homogeneity and purity are important issues for RF applications because it determines the thermal stability of the cavity. It was quickly realized that a frequent gradient limitation in superconducting cavities is due to thermal instabilities triggered by microscopic hot spots, for example normal conducting inclusions. This led researchers to investigate in detail the thermal behavior of niobium cavities, in relation to the material characteristics [7].

Table 2

Thermodynamic, superheating and maximum obtained RF field of some superconductors

Material	H_{th} (A/m)	H_{sh} (A/m)	H_{RF} max (A/m)
Pb	$6.4\ 10^4$	$8.4\ 10^4$	$6.4\ 10^4$
Nb	$1.6\ 10^5$	$1.9\ 10^5$	$1.6\ 10^5$
Nb_3Sn	$2.5\ 10^5$	$3.2\ 10^5$	$8.0\ 10^4$
YBCO	$\approx 6\ 10^5$ -10^6	$\approx 6\ 10^5$ - $8\ 10^5$	$8.0\ 10^4$

Niobium thermal conductivity

Two parameters are relevant for the description of the thermal behavior of the cavity, namely the niobium thermal conductivity and the Kapitza resistance at the niobium-helium interface. In the case of a hot spot, most of the thermal gradient is located in the niobium sheet, and the thermal properties of the interface play a minor role [7]. For a good thermal stability, a niobium cavity must thus be made from a material with high thermal conductivity. At cryogenic temperatures, the main heat carriers in niobium are electrons, and their mean free path is limited primarily by collisions with impurity atoms [8].

The electron mean free path is usually given in terms of a quantity (residual resistivity ratio), defined as:

$$\mathrm{RRR} = \frac{\rho_{300\mathrm{K}}}{\rho_{0\mathrm{K}}}$$

where ρ_{300K} is the room temperature resistivity (this term is constant, ρ_{300K} = $1.45\ 10^{-7}\ \Omega^{-1}.\mathrm{m}^{-1}$) and ρ_{0K} the normal state resistivity of niobium at zero temperature.

The approximate relationship giving the electron mean free path in niobium as a function of RRR is:

$$l(T = 0\mathrm{K})(\mathrm{Angstrom}) \approx 27.\mathrm{RRR}.$$

The thermal conductivity of niobium (Fig. 3) has been measured as a function of purity and past metallurgical history. A useful rule of thumb is:

$$\lambda(T = 4.2\mathrm{K})(\mathrm{W.m^{-1}.K^{-1}}) \approx \mathrm{RRR} / 4.$$

With usual values of RRR (a few hundreds), this relationship gives a thermal conductivity significantly smaller for niobium than for OFHC copper.

In the superconducting state, paired electrons in niobium decouple from the lattice and no longer participate in heat conduction. Heat is then carried by the small fraction of unpaired electrons. The poor thermal conductivity of niobium is thus intrinsically due to its superconducting nature. A Nb layer of 1 µm thickness would be sufficient for superconducting

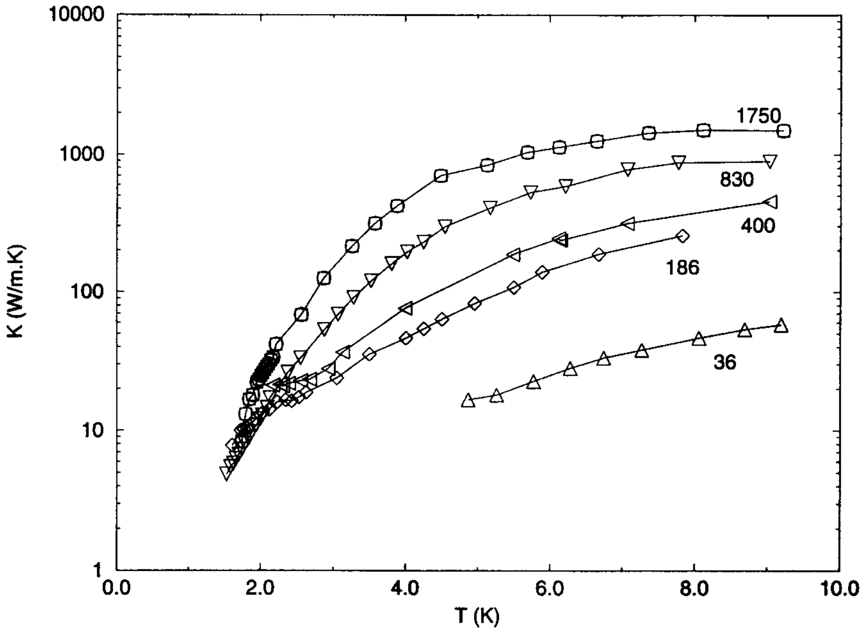

Fig. 3 The thermal conductivity of niobium as a function of temperature, for various RRR values.

cavities. Expensive as it is, the rest of the material serves merely as a substrate, despite its poor mechanical and thermal properties!

The above arguments show how important it is to make superconducting cavities with very pure material. One of the most significant advances in cavity performance is due to a recent effort from niobium suppliers to produce niobium of improved purity.

Niobium production

Niobium production from the ore to the raw ingot involves a complicated path, with chemical treatments and high temperature steps, which have been described in [9]. The purified ingot is then rolled into sheet form. Recrystallization annealings are usually performed by the supplier, between the rolling steps or before the sheet delivery, in order to warrant a uniform and well controlled grain size (typically 50 µm, for good sheet deformation capabilities). The cavity can then be formed.

The main impurities which contribute to the RRR degradation are C, N and O [9]. For state-of-the-art material (RRR 300), these are present at a concentration of about 100 at. ppm; Tantalum is also present in large amounts (1000 at. ppm). This element is difficult to separate from niobium because both have very similar chemical properties.

Niobium purity is improved by electron beam melting (2400 °C) in high vacuum. Light impurities (C, N, O, H) are vaporized during the process; four to six passes are generally considered to be necessary to reach a RRR level of the order of 300 [9].

Niobium post-purification

Post-purification of niobium can be achieved by a heat treatment associated with solid state gettering [10]. The material, either in sheet or in finished cavity form, is heated under high vacuum at temperatures ranging between 1000 °C and 1400 °C. This temperature is sufficient to permit diffusion of interstitial impurities C, N, O in the bulk material. The material is thus homogenized, thanks to the dissolution of the inclusions. At the same time, a liner of titanium placed close to the material to be purified sublimates. Its vapor deposits at the niobium surface and acts as a getter for the interstitial impurities which diffuse here. After a few hours of heat treatment, the bulk niobium is purified to a large extent. This technique is now capable of improving the niobium RRR by a factor of 10 (from 30 to 300 for "reactor grade" material, and from 200 to 2000 for "state-of-the-art" material). Niobium thermal conductivity can be considerably improved by this treatment, with a subsequent improvement of the cavity quench threshold. In spite of its drawbacks (cost, severe degradation of its mechanical properties), niobium post-purification is presently an interesting route to very high cavity performance.

5. THIN FILMS

Superconducting cavities internally coated with a thin superconducting film can also be envisaged. For a complete screening of the RF field by the superconductor, a minimum thickness of 10 times the penetration depth λ should be deposited. This corresponds to a thickness ranging between 0.5 and 2 μm and can be achieved by many deposition techniques. Considerable advantages can be expected from such "thin film" cavities:

- a cheap substrate can be used, with a subsequent cavity cost saving;

- a good heat conductor can be chosen for the substrate, giving a good cavity thermal stability.
  These two criteria can be met by using copper as the substrate.

- materials unavailable in bulk form can be deposited in thin films, with potentially interesting superconducting properties.

Nb/Cu thin films

A number of superconducting materials have been investigated for RF applications. So far, the most successful is niobium, sputter-deposited on a copper substrate. The deposition method, pioneered at CERN [11], is DC magnetron sputtering. Here, the copper cavity plays the combined roles of substrate and vacuum vessel, and the niobium target (cathode) is introduced at the centre of the vessel. The method is well suited to large cavities, working at low frequency, and at 4.2 K. In this case, it provides an appreciable cost saving in supply material. In addition, the RRR of the deposited material is around 30, close to the optimum value which minimizes the BCS contribution to the surface resistance. The archetype of this kind of application is LEP2 cavities. After a difficult start, this technology has now been transferred to industry, which provides cavities meeting the LEP2 specifications with a good success rate.

Nb(Ti)N thin films

Deposition of niobium-nitride films has also been attempted in various laboratories[12]. Here, the hope is to exploit the high critical temperature of this intermetallic compound (17 K) to lower the BCS contribution to the surface resistance, and to permit cavity operation at temperatures higher than 2 K (the goal is 4.2 K). The deposition method used so far is reactive sputtering, with a pure metallic target of Nb or NbTi alloy, and introduction of nitrogen in the sputtering gas.

So far, full success cannot be claimed. However, encouraging results have been obtained on samples, with low surface resistance at low field levels (400 mΩ at 4 GHz), and a BCS contribution much lower than for niobium (three times less at 4.2 K). Nitride accelerating cavities with competitive characteristics have not yet been produced.

Nb_3Sn layers

Thin films of Nb_3Sn have had more success (accelerating gradients as high as 15 MV/m, cavity Q-value higher than 10^{10} at low fields) [13], but the fabrication method (start from a pure niobium cavity, evaporate tin on its surface, and heat up until Nb_3Sn is formed to a few μm thickness) is probably expensive and does not lend itself to an easy industrialization. It might be interesting, however, in cases where cost is not a very important criterion, or to upgrade the performance of existing cavities.

YBCO

Theoretically, very low surface resistance could be expected from high T_c superconductors like YBCO. So far, the experimental results fall short of these expectations [14]. Sintered ceramics display a very large residual resistance. Moreover, R_{res} increases dramatically with increasing RF field. Thick coatings prepared by laser ablation, sputtering or other deposition techniques also display more or less the same behavior, whose cause can be found in the granular nature of the material. Epitaxially grown thin films have much smaller surface resistance, and their surface resistance is independent of the amplitude of the RF field. Their superconducting characteristics could make them attractive for accelerator applications. Unfortunately, they require a monocrystalline substrate, and so far have only been produced on flat samples of very restricted area.

Granular superconductivity

Most of the high T_c superconductor films and all the sputtered films have in common a very unfortunate feature: they display an increase of surface resistance with increasing RF field. This behavior limits severely the accelerating gradient obtainable from “thin film” or “high T_c " cavities. The disease seems to be worse for materials with higher T_c, or, more precisely, with smaller coherence length. The cause of this misfortune can be traced back to the granular nature of this kind of superconducting materials. The RF dissipation of granular superconductors has been investigated by many authors [15]. Here, the superconductor is modelled as an array of superconducting grains separated by weak links which, in the present case, can be structure defects like grain boundaries. The granularity is more pronounced if the grains are smaller, the boundaries more resistive or the coherence length smaller. If the current induced by the RF field through the weak link stays below its critical current, the weak link behaves as a resistively shunted inductance; in the opposite case, the weak link can be modelled as a pure resistor and the power dissipation of the array rises abruptly. These models provide guidelines to produce better, less granular superconducting films. They indicate that a weak link is a defect as large or larger than the coherence length. Since ξ_0 is small for high T_c materials, the challenge is to make perfect films, with a minimum number of structure defects.

6. CONCLUSION

In view of the arguments discussed above, one can understand why superconductors for magnet applications and superconductors for cavities are so different. The only requirement they have in common is a high critical temperature. Whereas superconductors for magnets need a high H_{c2}, and thus a small coherence length, materials for cavities need a low surface resistance and a large superheating field H_{sh}. In order to ensure an effective flux pinning, the former needs to be a dirty superconductor, the latter a clean one. Table 3 summarizes the requirements put on superconducting materials for RF and DC applications.

Overall, niobium seems to be an excellent compromise between the conflicting requirements imposed on a superconductor suitable for RF cavities, and its supremacy will probably last for a long time. In view of their superior critical fields and temperatures, compounds like NbN, Nb_3Sn or even YBCO are attractive substitutes to niobium, but it will be necessary to produce them in a very pure and perfect way in order to avoid problems related to the low coherence length of these materials.

Table 3
Main requirements of SC materials for RF and DC applications

Regime	H_c	H_{c2}	j_c	R_s	T_c	ξ	λ	Material
DC	-	large	large	-	high	small (pinning is needed)	large (bulk current)	NbTi Nb_3Sn
RF	large	-	-	small	high (R_s depends on T/T_c)	large (to give little sensitivity to structure defects)	small $R_s \propto \lambda^3$	Nb Nb(Ti)N ? Nb_3Sn? YBCO?
	....					....		
	Primary requirements					Consequences		

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REFERENCES

[1] H. Piel, “Superconductivity in Particle Accelerators”, CAS, Hamburg, 1988, ed. S. Turner, CERN 89-04 (1989).

J.H. Hinken, “Superconducting electronics”, Springer Verlag

[2] C.C. Mattis and J. Bardeen, Phys. Rev. 111 412 (1958)

[3] J. Pierce, “Superconducting microwave resonators” in “Methods of Experimental Physics” vol. 2, ed. R. Coleman, Acad. Press, N.Y. (1974)

M. Tigner and H. Padamsee in “Physics of high energy particle accelerators”, SLAC summer school (1982), ed. M. Month, AIP Conf. Proc. 105, 801 (1983)

J. Halbritter, J. Appl. Phys. 42 82 (1971)

P. Kneisel, O. Stolz, J. Halbritter, Proc. of the Appl. Superconductivity Conf. IEEE, New York (1972) p 657

[4] P. Schmüser, these proceedings

[5] T. Yogi and M. Mercereau, Phys. Rev. Lett. 39 826 (1977)

J. Halbritter, 1st Workshop on RF Superconductivity, Karlsruhe, KfK 3019 (1980)

[6] W. Weingarten, Particle World 1 93 (1990)

G. Müller, Proc. 3rd Workshop on RF Superconductivity, Argonne (1987) ANL-PHY-88-1 p 331

[7] H. Padamsee, 1st Workshop on RF Superconductivity, Karlsruhe, KfK 3019 (1980)

H. Padamsee, IEEE T. Mag. 19 1322 (1983)

[8] K.K. Schulze, Journal of Metals, May 1981, p33

[9] M. Hörmann, J. of Less Common Metals 139 1 (1988)

[10] T. Haas, Surf. Sci. 5 345 (1966)

E. Fromm, Vacuum 19 191 (1969)

H. Farrell, Surf. Sci. 38 31 (1973)

[11] G. Arnolds-Mayer and W. Weingarten, IEEE T. Mag. 23 1620 (1987)

C. Benvenuti et al., IEEE T. Mag. 21 153 (1985)

[12] C. Benvenuti et al., 5th Workshop on RF Superconductivity, DESY, Hamburg, ed. D. Proch, pp 189 and 518 (1991)

R. di Leo, A. Nigro, G. Nobile and R. Vaglio, J. Low Temp. Phys. 78 41 (1990)

P. Bosland et al, 6th Workshop on RF Superconductivity, CEBAF, USA, ed. R. Sundelin, (1993) p. 1028

[13] J.J. Hanak et al. RCA Rev. 25 342 (1964)

M. Peiniger and H. Piel, IEEE T. NS 32 3612 (1985)

R. Röth and P. Kneisel, 7th Workshop on RF Superconductivity, Gif sur Yvette, ed. B. Bonin (1995)

[14] G. Müller, this CERN course.

[15] L. Cohen, I.R. Gray, A. Porch and J.R. Waldram, J. Phys. F: Metals Physics, 17 L179 (1987)

A.M. Portis, IEEE T. Mag. 25 2390 (1989)

O.G. Vendyk et al., Zh. Tekh. Fiz. 59 107 (1989)

B. Bonin and H. Safa, Superc. Sci. Technol. 4 257 (1991)

C. Attanasio, L. Maritato and R. Vaglio, Phys. Rev. B 43 6128 (1991)