

# DEGRADATION OF SUPERCONDUCTING RF CAVITY PERFORMANCES BY EXTRINSIC PROPERTIES

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## Abstract

The intrinsic surface resistance of superconductors  $R_{BCS}(T, f, B)$  is degraded by extrinsic effects which actually limit applications, e.g., by rf breakdown or severe heat load. Examples for extrinsic effects are, rf residual losses  $R_{res}$  increasing with rf electric field  $E_p$  or rf magnetic field  $B_p$  or heating  $\Delta T$  of the cavity surface relative to the He-bath. With the now available material qualities and surface preparations, gross local defects are rare and global degradations show up. Aside of the rf residual losses  $R_{res}$  related to the Nb-Nb<sub>2</sub>O<sub>5</sub> interface, extrinsic field dependencies are encountered reaching from Q-slope, i.e.  $\delta R \propto R_{BCS}(T, f) (1 + \gamma(B_{rf}/B_c)^2 + \eta(B_{rf}/B_c)^4 +)$  with  $\gamma > \eta$  as the begin of a Taylor series and  $B_c(0) \approx 0.2$  T, to Q-drop, i.e.  $\delta R^E \propto \exp(-c/E_p)$  without field emission, and for Nb films or heavily corroded Nb, to  $R_{hys} \propto \omega B_{rf}/j_{cJ}$  with  $j_{cJ}$  the critical current density of weak links. For carefully prepared and oxidized Nb  $R_{res}(T, f) \geq 2 (f/\text{GHz})^2 n\Omega$  have been attained. In bulk Nb cavities immersed in He a Q-slope with  $\gamma (T < 2.17\text{K}=T_\lambda) \lesssim 0.2 - 3$  has been reached turning above  $T_\lambda$  to  $\gamma \gtrsim 10 - 20$  confirming  $\Delta T$  heating by the Kapitza thermal boundary resistance as main cause. Thermal interface resistances in sputtered Nb cavities yield, e.g.  $\gamma(T) \approx 20 - 90$ , simulating an exponential  $R(B)$ -increase via the exponential  $R_{BCS}(T + \Delta T, f, B)$  increase. The Q-drop is explained by interfacial tunnel exchange with Nb<sub>2</sub>O<sub>5</sub>. Internal surfaces, i.e., weak links, as they occur by oxidation along grain boundaries or at other defects, e.g., in cold worked Nb, yield hysteresis losses  $R_{hys}$  at rather low fields. Because of  $j_{cJ}(T \lesssim T_c/2) \approx \text{const}$ ,  $R_{res}(T \leq T_c/2)$  and  $R_{hys}(T \leq T_c/2)$  are T-independent for Nb, like the interface losses  $\delta R \propto \exp(-c/E_p)$ , whereas the Q-slope replicates via  $\gamma(T)R_{BCS}(T)$  the BCS surface resistance in its T-dependence for  $R_{BCS}(T + \Delta T) > R_{res} + R_{hys} + R^E$ . All observations, up to date, can be explained in this frame work, showing the importance of detailed analysis of  $R(T, f, B)$ -dependencies and pointing toward locally piled up rf losses transferred to interfaces and then to He being the key for further improvements of peak fields and for the reduction of rf losses.

## INTRODUCTION

The now available excellent Nb qualities free of gross defects, with resistance ratios  $RRR \geq 100$ , with surface treatments reaching from buffered chemical polishing (BCP), to electropolishing (EP) followed by high pressure ( $\geq 80\text{bar}$ ) water rinsing (HPR) [1] has yielded rf breakdown fields above  $B_{c1}(T \geq 1.6 \text{ K}) \leq 0.17$  T, corresponding to surface peak fields  $E_p > 70\text{MV/m}$  [2]. Those excellent results have been reached despite global deteriorations related, e.g., to oxidation or heat resistances discussed in Sects. 2 and 3.

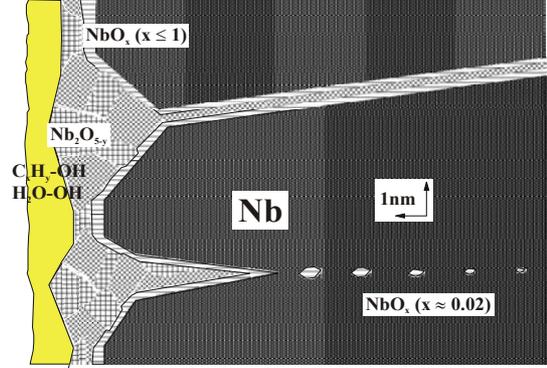


Fig. 1: Nb surface with crack corrosion by oxidation by Nb<sub>2</sub>O<sub>5</sub> volume expansion (factor 3). Nb<sub>2</sub>O<sub>5-y</sub>-NbO<sub>x</sub> weak links/segregates (y, x < 1) extend up to depths between 0.01 – 10  $\mu\text{m}$  for good – bad Nb quality and weak - strong oxidation [4b]. For sputtered Nb the oxidation proceeds preferably along already existing island boundaries due to growth. Embedded in the adsorbate layer of H<sub>2</sub>O/C<sub>x</sub>H<sub>y</sub>OH ( $\geq 2$  nm) being chemisorbed by hydrogen bonds to NbO<sub>x</sub>(OH)<sub>y</sub>, adsorbate covered dust is found. This dust yields dynamically enhanced field emission (EFE [5b]).

The consequences of oxidation sketched in Fig. 1 are summarized in Sect. 2 material science wise, whereas in Sect. 3 the surface resistances  $R(T, \omega, B)$  due to intrinsic and extrinsic effects are discussed as function of temperature T, frequency  $\omega$  and B<sub>rf</sub> fields. These discussions focus on newly clarified aspects, like enforced oxidation along grain boundaries or dislocations and on different Nb<sub>2</sub>O<sub>5-y</sub> qualities, mentioned shortly in [3].

The amount and size of NbO<sub>x</sub> cluster depend on nucleation and treatment, as discussed in Section 2, especially sizes will grow by segregation with 90 - 150 °C baking. Those NbO<sub>x</sub> cluster with their depressed order parameter  $\Delta^*$  yield localized states easily driven out of the thermal equilibrium causing the peak at  $B_{max} \approx 1-2\text{mT}$  in  $R_{BCS}(T, \omega, B)$  and rf induced superconductivity stabilizing those regions, as discussed in Sect. 3 – 5.

The different Nb<sub>2</sub>O<sub>5-y</sub> qualities and thicknesses cause exponentially increasing electric interface losses  $R^E \propto \exp(-c/E)$  being nonanalytical, in contrast to surface resistance increases by heating with  $\delta R^E \propto \gamma^* B^2$  in lowest order in B.

Bulk Nb with wall thickness  $d \approx 2\text{mm}$  submersed in LHe show a jump in  $\gamma^*(T)$  at  $T_\lambda \approx 2.17$  K by about a factor 5 – 10 proving the dominance of  $\Delta T$  heating by the Kapitza resistance  $R_K$  over the diffusive Nb thermal conductivity. Sputtered Nb shows enhanced  $\gamma \gtrsim 20$  values depending on treatment. All  $\gamma$  enhancements are likely caused by locally enhanced losses  $R_{res}$  being transferred by negligible diffusive broadening to interfaces by ballistic phonons or quasiparticles, discussed in Section 3 – 5.

With this basis, extrinsic and intrinsic low and high rf field surface resistances are analyzed in Sect. 4 and consequences and ways to improve the rf cavities will be discussed.

## CHEMISTRY AND MORPHOLOGY OF THE Nb-Nb<sub>2</sub>O<sub>5</sub>-ADSORBATE SYSTEM

In Fig. 1, a typical Nb surface is shown being homogenized and cleaned by heating in UHV or in a Ti-enclosure above 1000 – 1400°C followed by BCP or EP [1,2] removing about 40 - 100µm of the Nb surface containing defects by, e.g., machining nucleating enforced oxidation [3,4]. Starting from the vacuum side, by its strong hydrogen bonds the NbOOH surface is coated by 20 – 100 monolayers (ML) of H<sub>2</sub>O and hydrocarbons [5,6], where the last 2 ML cannot be removed by UHV annealing below about 400°C. Embedded in this soup, dust coated by water and hydrocarbons may cause surface flashover and enhanced field or secondary electron emission [5]. Dust is reduced drastically by intense HPR making so surface field strengths above 10MV/m possible [1,4a]. By the high energy per bond of 7 eV in the metallic interface oxide NbO<sub>x</sub>(x ≈ 1) and of 5eV in the dielectric Nb<sub>2</sub>O<sub>5</sub>, any Nb surface gets oxidized instantaneously at pressures above 10<sup>-10</sup> Torr below about 10<sup>3</sup> K to thicknesses up to 1 – 3 nm Nb<sub>2</sub>O<sub>5</sub> by the Cabrera-Mott process [4b]. There O<sup>-</sup> is driven by the potential through the Nb<sub>2</sub>O<sub>5-y</sub> oxidizing the metallic NbO<sub>x</sub> to Nb<sub>2</sub>O<sub>5</sub> by a factor above three volume expansion, therewith the Nb is strained yielding cracks and NbO<sub>x</sub> cluster injection into Nb. Hence the Nb and its surface layers, e.g., Nb<sub>2</sub>O<sub>5</sub> are compressed shifting all clumsy ions (PO<sup>-2</sup>,...) out of O-octahedra network making up the surface as known from the compact part of the double layer at metal-electrolyte interfaces. Nb<sub>2</sub>O<sub>5-y</sub> is nanocrystalline by its rigid, edge connected NbO-octahedra blocks with  $\phi = E_c - E_F \approx 1\text{eV}$  as difference of Fermi energy  $E_F$  and conduction band  $E_c$  which in turn get side connected via crystallographic shear planes (CS) with  $\phi \approx 0.1\text{eV}$ , where oxygen vacancies occur easily yielding for 300 K surface oxides with  $n_L \approx 10^{19-20}/\text{cm}^3$  localized states at  $E_F$  [4c]. The tunnel transport through Nb<sub>2</sub>O<sub>5-y</sub> barriers follow CS channels showing up most clearly for barrier width  $d \geq 1.5\text{nm}$  via resonant tunneling [4c], being normal conducting for  $z_L \geq 0.3\text{nm}$ . The states  $n_L(z \leq 2\text{nm})$  are in fast ( $< 10^{-10}\text{sec}$ ) interface tunnel exchange (ITE) with Nb conduction electrons of density  $n_m$  as sketched in Fig. 2. For the conduction electrons ITE yield surface scattering described by a surface mean free path  $l_s$  and normal conducting  $n_L(z_L)$  yield rf residual losses  $R_{\text{res}}$  and tunnel leakage currents. The actual exchange with Nb<sub>2</sub>O<sub>5-y</sub>-states under influence of an rf electric field  $E^\perp(t)$  correspond to dielectric interface losses  $R^E$  [7].

The unperturbed bulk Nb has a  $T_c = 9.25\text{K}$  and an average energy gap  $\Delta_0(0) = 1.56\text{meV}$ , where 2 At % O degrades  $T_c$  down to 7 K and  $\Delta_0$  down by 20 to 30% [4]. The 2 – 3 ML thick metallic NbO<sub>x</sub> (x ≈ 1) – layer between Nb<sub>2</sub>O<sub>5-y</sub> and Nb has a reduced energy gap  $\Delta^* < \Delta_0$ . Inside the Nb below 600 K Nb<sub>2</sub>O<sub>5-y</sub>-weak links or NbO<sub>x</sub> (0.2 ≥ x 0.02) precipitate (Fig. 1) out of the Nb-O lattice gas [4a]. By the enhanced volume of NbO<sub>x</sub>-precipitates and of Nb<sub>2</sub>O<sub>5-y</sub>-interfaces the surrounding Nb lattice is compressed enhancing  $\Delta^* > \Delta_0$  by up to 10% whereas at NbO<sub>x</sub> itself  $\Delta^* < \Delta_0$  is depressed by 10 to 20% [4]. The intermingled enhanced and depressed  $\Delta^*$  regions of size below about 5nm  $\ll \xi_F$  smear the BCS density of state and depress the average energy gap to  $\Delta < \Delta_0$ . For defective Nb, e.g. RRR ≈ 50 or by coldworking, the Nb<sub>2</sub>O<sub>5</sub>

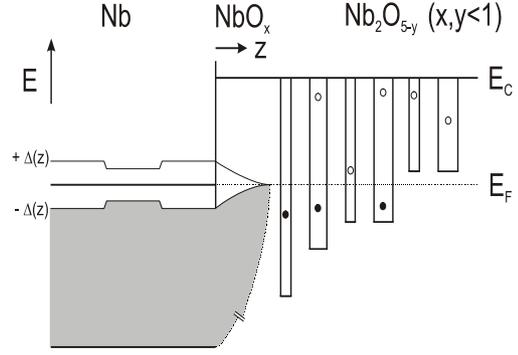


Fig. 2: Band structure at Nb-NbO<sub>x</sub>-Nb<sub>2</sub>O<sub>5-y</sub> interfaces with  $E_c - E_F \approx 0.1 - 1\text{eV}$  as barrier heights for tunneling along crystallographic shear planes ( $\sim 0.1\text{eV}$ ) or of Nb<sub>2</sub>O<sub>5-y</sub> crystallites ( $\sim 1\text{eV}$ ). Added is the superconducting energy gap  $\Delta^*(z)$  being reduced in NbO<sub>x</sub> clusters or interfaces. By their volume expansion those clusters locally enhance  $T_c^*$  and  $\Delta^* > \Delta_0$  in adjacent Nb by the uniaxial strain yielding a smeared BCS DOS.

filled weak links extend between  $w \approx 0.2 - 1\mu\text{m}$  deep into Nb [3], in contrast to RRR > 100 well annealed Nb with  $w < 0.1\mu\text{m}$  sketched in Fig. 1.

Like the Nb-O-system below 600K, hydrogen precipitates to Nb-H below about 130 K preferably to form weak links ( $w \approx 1\mu\text{m}$ ) nucleating close to the surface because of the NbH volume expansion [8]. NbH<sub>x</sub>-precipitates are removed to larger depth by lattice compression by NbO<sub>x</sub> precipitates in the surface layer. Better is UHV annealing above 800°C, where H<sub>2</sub> is evaporated, and then sealing-off the surface by Nb<sub>2</sub>O<sub>5-y</sub> to prevent H-pick-up.

## INTERACTION OF RF-FIELDS WITH Nb CAVITY SURFACES

The rf fields at metallic surfaces can be separated in rotational free component  $\text{rot } E \equiv 0$ , namely  $E^\perp$  giving rise to an electric field surface impedance  $Z^E = R^E - i\omega\mu_0\lambda^E$  and in a divergence free component  $\text{div } E \equiv 0$ , namely  $H^\parallel$  giving rise to rf shielding currents and magnetic surface impedance  $Z^H = R^H + i\omega\lambda^H$ . Whereas the dielectric surface impedance  $Z^E$  of clean metals is negligible because of  $\lambda^E \leq 0.1\text{nm}$  [7], Nb-Nb<sub>2</sub>O<sub>5-y</sub> by ITE cause a measurable  $Z^E$  being discussed in Sect. 3.1. By a penetration depth  $\lambda^H (T < T_c/2, f < 100\text{GHz}) \geq 40\text{nm}$  and  $\lambda^H (T > T_c) > \mu\text{m}/\sqrt{f/\text{GHz}}$  the shielding current cause the dominant impedance  $Z^H$  in superconducting and normal conducting rf cavities.

### Electric Surface Impedance $Z^E$

Whereas the intrinsic  $Z^E$  of metals is negligible small [7], the localized states  $n_L$  of Nb<sub>2</sub>O<sub>5-y</sub>, at the Fermi energy  $E_F$  (Fig. 2) enhance by interface tunnel exchange (ITE) with the high DOS of Nb  $n_m (E_F)$   $Z^E$  by several orders of magnitude. Quantitatively, the exchange is governed by a tunnel rate (Fig. 2)

$$1/\tau(z_L, \varepsilon) = 1/\tau(0, \varepsilon) \exp(-2\kappa z_L) \quad (1.1)$$

with  $\hbar\kappa = \sqrt{2m(E_c - E_F)} = \sqrt{2m\phi}$  with  $n_m \propto 1/\tau(0, \varepsilon)$ . This exchange defines a distance  $z^*(\omega)$  up to which the localized states are in equilibrium

with the DOS of the metal for a frequency  $\omega\tau(z^*) = 1 = \omega\tau(0) \exp 2Z^*(\omega)\kappa$ .

According to [7]  $Z^E$  is proportional to  $z^*(\omega) n_L(z^*)$ , i.e. linearly increasing with  $n_m$ ,  $n_L$  and  $z^*(\omega)$ , i.e. exponentially with lowered barrier height  $\phi$ . The latter is quite small along crystallographic shear planes with  $\phi \approx 0.1$  eV which house the localized states  $n_L \approx 10^{19-20}/\text{cm}^3$  [4]. For normal conducting Nb estimates gave with  $\varepsilon = \varepsilon_r \varepsilon_0$  and  $\varepsilon_r(\text{Nb}_2\text{O}_{5-y}) \approx 10 - 20$  at the interface [7] with  $z^* \approx 1 - 3$  nm < d the  $\text{Nb}_2\text{O}_5$  thickness

$$R^E(\text{Nb} - \text{Nb}_2\text{O}_{5-y}) \approx 10^{-4} \Omega / \varepsilon_r^2 \approx 10^{-5} - 10^{-7} \Omega \quad (1.2)$$

at about 1 GHz. In the superconducting state an energy gap  $|\varepsilon| < \Delta$  in  $n_m(\varepsilon)$  opens blocking ITE. With  $2\kappa\Delta\varepsilon_r/e\beta^* = c$  for fields higher than  $E_{rf} = \varepsilon_r\Delta/e\beta^* \min\{z_E^*(\omega), d\}$  ITE sets in where  $e z_E \beta^* E_{rf}^2/\varepsilon_r$  overcomes the energy gap  $\Delta$ . The static, geometric field enhancement factor  $\beta^* < 10$  is smaller than the dynamic field enhancement factor  $\beta \approx 10^2 - 10^3$  [5b]. Then  $R^E$  writes:

$$R^E = \omega\mu_0 d^* e^{-c/E}, E^\perp > E_{rf}^\circ \quad (1.3)$$

i.e. only states between  $z_E^\perp$  and  $z^*(\omega)$  cause ITE rf losses approaching for  $E_{rf}^\perp \approx 10^9$  V/m Eq. (1.2) with  $d^* \propto n_m n_L$ . Hence  $R^E$  shows an onset at  $E_{rf}$  increasing exponentially like  $\exp(-c/E_{rf})$  with  $E^\perp$ .

The small exponent  $c$  in Eq. (1.3) is fitted by  $E^{\perp n}$  starting with  $4 \leq n \leq 16$  with  $n$  increasing by surface smoothness and reduced  $n_L$ , i.e. not by a Taylor series [4a]. ITE in Eq. (1.3) develops out of standard field emission [7] by substituting  $\phi = E_c - E_F \approx \text{eV}$  and  $\beta \geq 10^2$  by  $\Delta \approx \text{meV}$  and  $\beta^* \ll \beta$  yielding as ratio between FE- and ITE-exponents  $\phi/\Delta \approx 10^3$ .

### Intrinsic Magnetic Surface Impedance $Z^H$

The BCS-theory [9] is an excellent tool to obtain averaged material parameters over a  $2\lambda = 80$  nm Nb surface layer as shown in [4a,b] with agreement experiment/theory above 6 orders in magnitude in  $R(T)$ . The lengths describing  $Z^H$  of Nb are: the London penetration depth  $\lambda_L$ , the dimension of Cooper pairs  $\xi_F$  and the electron mean free path  $l$ .  $l$  is reduced to  $\lambda_L/\sin 30^\circ \approx 3\lambda_L$  by inelastic surface scattering via ITE [7] by trajectories with  $\theta \approx 30^\circ$  relative to the surface enhancing with  $1/l = 1/l_0 + 1/l_s$

$$\lambda(T, l) \approx \lambda_L(T) f(\gamma) \sqrt{1 + \xi_F/l} \quad \text{with } \gamma = \lambda_L / \xi_F \quad (2.1)$$

with  $\lambda(0) f(\gamma) = 38$  nm by non-local effects [9].  $\lambda(T) > \lambda_L(T)$  is only weakly depending on temperature below  $T_c/2$ , whereas above 5K strong and preparation dependent  $\lambda(T)$  increases have been reported [4b]. Especially the  $\lambda(T_c)$  jump [3] being related to stretches of  $\text{NbO}_{0.02}$ -clusters with  $T_c \approx 6-7$  K are worth mentioning [3] being indicated in Fig. 1, extending as weak links up to  $\approx 10\mu\text{m}$  deep into cold worked Nb.

In contrast to  $\lambda(T)$ , the surface resistance  $R(T, f, B)$  has been studied intensively in the last year because of its direct relevance to superconducting rf accelerator performance [4a]. Whereas  $\lambda(T, l)$  of Eq. (2.1) are approximated very well by a superconducting momentum transport carrier density, the surface resistance needs a complex computer code [9] for the matrix elements depending sensitively on the BCS density of states close to the Fermi energy  $\varepsilon = E - E_F$

$$N(\varepsilon) = N_F(0) \cdot \varepsilon / \sqrt{\varepsilon^2 - \Delta^2} \quad (2.2)$$

and on momenta of in- and outgoing quasiparticles. Due to the BCS singularity  $R$  is enhanced yielding [9]

$$R_{\text{BCS}}(T, \omega) \approx c\omega^2 N(\Delta) (\Delta + \hbar) \\ [f(\Delta) - f(\Delta + \hbar\omega)] M^2 \approx c^* \omega^2 \Delta/kT \sqrt{2\hbar\omega\Delta} \\ \exp(-\Delta/kT); (T \leq T_c/2, \hbar\omega \ll \Delta) \quad (2.3)$$

Small  $\text{NbO}_x$  ( $x < 0.1$ ) clusters of sizes below  $5 \text{ nm} < \xi_F$  show a depressed order parameter  $\Delta^* < \Delta_0$  compensated in part by proximity to bulk Nb, whereas their strain onto the surrounding enhances  $\Delta^* > \Delta_0$ . Those  $\text{NbO}_x$ -cluster smear the BCS square root singularity by  $\delta\Delta^*$  yielding a reduction of the mean  $R_{\text{BCS}}$  in Eqs. (2.3) by a factor 2 at 1.5 GHz

$$R_{\text{BCS}}^*(T, f) \propto (\hbar\omega)^2 \exp(-\Delta/kT) / \sqrt{\Delta_0 \delta\Delta^*} kT < R_{\text{BCS}} \\ (T, f < 10\text{GHz}) \quad (2.4)$$

Clearly, those cluster reduce also the mean free path  $l$ , which is already reduced to  $l_s \approx 3\lambda_L$  by inelastic surface scattering at  $n_L$ -sites of  $\text{Nb}_2\text{O}_{5-y}$  of Fig. 2.

At  $\text{NbO}_x/\text{Nb}_2\text{O}_{5-y}$  interface layers the energy transferred to quasiparticles is effectively transferred to phonons via localized states  $n_L$ . This is different to  $\text{NbO}_x$ -cluster depicted in Fig. 1, where quasiparticles are weakly localized by  $\Delta^* < \Delta$  yielding enhanced losses by  $\Delta^* < \Delta$  with an electron-phonon coupling like in bulk Nb at his temperature. This electron phonon coupling is weak as shown by the inelastic scattering time for  $\hbar\omega \ll \Delta$  excitations

$$\tau_{\text{in}} \approx 1.15 \cdot 10^{-10} (T_c/T)^3 \text{ sec} \quad (2.5)$$

which has to be compared to the quasi particle absorption rate  $\tau_{\text{qp}}$  [4a]

$$\frac{1}{\tau_{\text{qp}}} = \frac{B_{rf}^2}{\hbar 2 \cdot \mu_0} \frac{1}{n_c \hbar \omega} \cdot \frac{R}{X} \quad (2.6)$$

where for Nb with  $n_m = 6.3 \cdot 10^{22}/\text{eVcm}^3$  as density of states,  $\hbar\omega \approx 10^{-5}$  eV,  $B_{rf} \approx 1\text{mT}$  and  $X/R \approx 200$   $\tau_{\text{ab}} \approx 10^{-8}$  sec is obtained comparable to  $\tau_{\text{in}}$ . For  $B_{rf} \gtrsim 1\text{mT}$  this bottleneck enhances the local electronic temperature  $T^* > T$  at  $\text{NbO}_x$ -clusters and  $\text{NbO}_x$  surface layers and, thus, according to Eq. (2.3) by  $\exp(-\Delta^*/kT^*)$ , the absorption growth first with  $B$ . In contrast, adjacent Nb with the mean energy gap  $\Delta$ , with the absorption given by  $\exp(-\Delta/kT)$ , the absorbed quanta easily dissipate somewhere in the Nb-wall because of  $\tau_{\text{in}} v_F \approx 2$  nm. But in  $\Delta^* < \Delta$ -regions confined states overheat by  $1/\tau_{\text{qp}} > 1/\tau_{\text{in}}$  holding locally. The overheating of  $\Delta^* < \Delta$  regions in a second step for  $B > B_{\text{max}}$  yields rf losses decrease with  $T^* \rightarrow \infty$ , where according to [4a]

$$R^* B_{rf}^2 \approx \text{const}; B > B_{\text{max}} \quad (2.7)$$

holds for energy independent  $\tau_{in}$ . Above about  $10B_{max}$   $R_{BCS} \propto \exp(-\Delta/kT)$  holds where thermal equilibrium is guaranteed because of  $v_F\tau_{in} \gg \lambda$  for quasiparticles with energies  $|\epsilon| \geq \Delta$ . This dynamic stabilization of regions with  $\Delta^* < \Delta$  corresponds to rf stabilization and rf induced superconductivity in those regions [10]. It should be mentioned that this rf stabilization has been observed also in the transition of  $NbO_x$  layers to the normal conducting state [3b].

### Nonlinearities in $R(T,B)$ for Homogeneous Nb

Intrinsic nonlinearities, like R-increases due to heating, can be cast in a power series [4a].

$$R(T,f,B) = R(T,f, \approx 10mT)$$

$$[1 + \gamma^* (B/0.2T)^2 + \eta (B/0.2T)^4 + \dots] \quad (3.1)$$

where as scaling field for Nb  $B_c(0) \approx 0.2$  T is used substituting  $B_c(T)$ . For intrinsic BCS nonlinearities  $\gamma^*_{BCS} < 0.1$  is estimated as upper limit, whereas heating yields larger  $\gamma^*(T)$  values given in the one dimensional case by

$$\gamma^*(T_0) = \Delta T / \partial R(T) / \partial T / R(T_0) \approx$$

$$R_{BCS}(T_0) (0.2T/B_c(T_0))^2 2\Delta/kT_0^2 \left[ \sum \frac{d_i}{\kappa_i} + R_k(T) \right] \quad (3.2)$$

Here  $d_{Nb}$  and  $\kappa_{Nb}$  are thickness and thermal resistance of Nb, for sputtered Nb/Cu the interface resistance has to be added and, at least, the Kapitza resistance  $R_k(T)$  toward the He-bath has to overcome. The heat transfer by phonons is given by their scattering at quasiparticles and phonons which in RRR  $> 100$  Nb is with  $l_{ph}(T < 4K, < 4GHz) \approx 2.6cm$  [4a] very scarce and, hence, yields for homogeneous Nb walls ( $\leq 5$  nm) ballistic heat transfer from the rf interaction volume to e.g. the Nb/ $Nb_2O_{5-y}$ /OH/ $H_2O$ -He-interface. Grain boundaries may scatter phonons but always the Kapitza resistance has to be overcome, yielding  $\gamma^*(T) \geq 0.2$  increasing at  $T_\lambda = 2.17$  K to  $\gamma^* > 10$  by film boiling, as shown in Fig. 3.

### Residual rf Magnetic Losses $R^H_{res}$ and $R_{hys}$

As discussed in [10a] the  $Nb_2O_{5-y}$ -interface with normal conducting states  $n_l(z \geq 0.3$  nm) yields inelastic surface scattering and coherent momentum transfer to the lattice staying finite for  $T \rightarrow 0$  with the ITE  $\tau(z^*)$  of Eq. (1.1) which yield [4a,b]

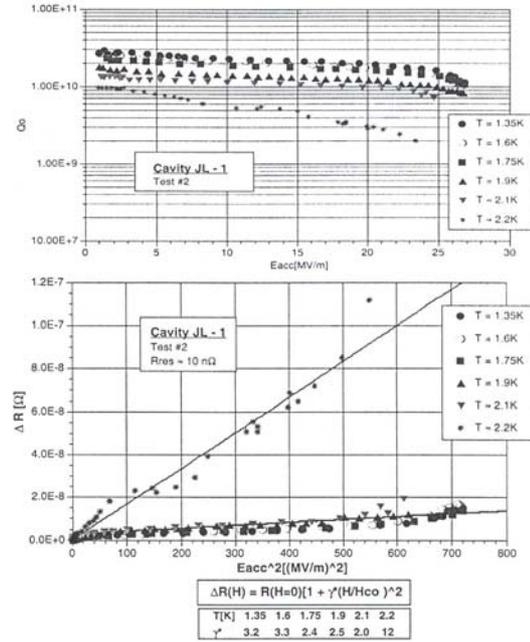


Fig. 3:  $R(T,B)$  results [12] show that for  $R_{res} \approx 2.6$  n $\Omega$   $B_p \approx 180mT > B_{c1}$  (1.6 K) = 160 mT is obtained with  $\gamma^* = 0.25$  whereas after 2 years  $R_{res} \approx 10n\Omega$  yielded  $B_p \approx 120$  mT at 1.6 K, where  $R(T,H) B_{crit}(T)^2 \approx const$  holds.  $\gamma^*(T) (B_p(T)/0.2 T)^2$  shows a pronounced  $\gamma^*(T)$  dependence [12b] with a jump at  $T_\lambda$  to  $\gamma^* \geq 12$ . The surface with  $R_{res} = 2.6$  n $\Omega$  shows a slight  $R^E \sim \exp(-c/E_p)$  degradation above  $E^0 \approx 72MV/m$  whereas  $R_{res} = 10n\Omega$  is accompanied by ITE and  $R^E$  (Eq. (1.3) above  $E^0 \approx 45$  MV/m [12].

$$R_{res}(T,f) < 4 \cdot 10^{-9} \Omega (f / GHz)^2 \quad (4.1)$$

The weak links filled with  $Nb_2O_{5-y}$  show the Josephson penetration depth  $\sqrt{\lambda_J(T,H)} \approx \hbar / 2e\mu_0 \lambda_J(T) j_{cJ}(T)$ . For weak links in a mean distance  $a_J$  with  $R_{bl}$  as leakage current resistance rf residual losses are obtained [10]

$$R_{res}^{WL}(T,f) \approx \frac{(2\mu_0 \omega \lambda_J(T))^2}{1 + (\omega \tau_J(T))^2} \frac{\lambda_J(T)^3}{a_J R_{bl}} \quad (4.2)$$

$$\text{with } 1/\tau_J = \frac{2e}{\hbar j_{cJ}(T) R_{bl}}$$

as Josephson frequency. In weak links extending deep into Nb  $w \gg \lambda_J$ , for  $B_{rf} > B_{c1J} \approx B_{c1} \lambda_L/\lambda_J$  Josephson fluxons penetrate causing hysteresis losses with the areal factor  $2\lambda/a_J$  [11]

$$R_{hys}(T,\omega,B) \approx 4\pi/3\mu_0 \omega B/j_{cJ} 2\lambda/a_J \quad (4.3)$$

For Nb  $\lambda(T \leq T_c/2) \approx \lambda(0)$  and  $j_{cJ}(T \leq T_c/2) \approx j_{cJ}(0)$  holds and, hence, below  $T_c/2$  the rf residual losses show  $R_{res}(T \leq T_c) = const$  and  $R_{hys}(T \leq T_c/2) = const$ .

## EXPERIMENTAL RESULTS

Experimentally, eigenfrequency changes  $\Delta f/f_0$  and rf losses, e.g. the decay time  $\tau = Q_0/\omega$  are the observables in

rf measurements, being related to the surface impedances by [10]:

$$\frac{\Delta\omega}{\omega_o} = \frac{i \oint ds ZH^{\parallel 2}}{2\omega_o \mu_o \int dVH^{\parallel 2}} + \frac{\epsilon_o / \mu_o \oint ds - Z^E E^{\perp 2}}{2i\omega_o \mu_o \int dVH^{\parallel 2}} =$$

$$i \frac{Z^H}{2G^H} + i \frac{Z^E}{2G^E} \text{ for } \{H^{\parallel}, E^{\perp}\} \ll \{H_c, 10^8 V/m\}$$

For TEM modes  $G^H = G^E$  holds, being weakened in TM-modes to  $G^H \leq G^E$  approaching  $G^E \rightarrow \infty$  for TE-modes. In our analysis of the  $TM_{010}$ -mode we assume  $G^H = G^E$  with  $G^H \cong 290 \Omega$  and peak field ratios  $B_p/E_p = 2.5 \text{ mT/MV/m}$  and  $E_p/E_{acc} \cong 1.8$

In [4a,b,9] the surface resistances  $R(T, \approx 10 \text{ mT}) \cong R_{BCS}(T) + R_{res}(T)$  have been quantified by the BCS theory, resulting in material parameters of Nb in 80 nm ( $\geq 2\lambda$ ) surface layers. At 1.5 GHz by annealing Nb at  $T \approx 100^\circ\text{C}$   $R_{BCS}$  decreases by a factor 2 for RRR > 100 Nb and 1.5 for RRR  $\approx 30$  Nb to the identical lower level [4a], whereas the already degraded  $\Delta/kT_c \approx 1.85 < 2.05$  does not degrade further. Related to this  $R_{BCS}(10\text{mT})$ -decrease is an increase of the  $R(B \approx \text{mT})$ -peak.

In Figs. 3 above  $E_p \approx 10 \text{ MV/m}$  field dependencies are fitted by  $\gamma^*(B/B_c)^2$ . In the Nb cavity with the highest  $B_p$  obtained to date,  $\gamma^*$  (1.6 K) = 0.25,  $R_{res} \approx 2\text{n}\Omega$  and  $B_p \approx 185 \text{ mT} > B_{c1}$  (1.6 K)  $\approx 168 \text{ mT}$  has been achieved [12a], degrading to  $R_{res} \approx 10\text{n}\Omega$  and  $B_p \approx 120\text{mT}$  by BCP after two years, where  $\gamma^*$  degrades to  $\gamma^*$  ( $T < T_\lambda$ )  $\approx 3$  jumping to  $\gamma^* \approx 12$  above  $T_\lambda$  shown in Fig. 3.  $\gamma^*$  ( $T < T_\lambda$ )  $\approx 2$  and  $E_p$ -onsets at 40MV/m are standard as obvious in [4a, 7, 12b, 13] and Figs. 3 - 5. In Fig. 4  $R(B)$ -dependencies and in Table 1 fit results are shown obtained in the  $TE_{011}$  and  $TM_{010}$ -modes of one cavity before and after UHV baking. The exponential  $R^E$  fits for TM modes to Eq. (1.3) are excellent being discussed in Sect. 5. Spun cavities show a very rough surface and, hence onset fields  $E_p^o \approx 20 \text{ MV/m}$  in Fig. 5, which shifts to higher fields with BCP, whereas  $c$  increases. The  $c$  increases of Fig. 5 are fitted in [4a,13] by the expansion of  $\delta R^E \propto E^{2n}$  starting with  $n \geq 4$  showing that this effect cannot be cast into a Taylor series. In Fig. 4b also such  $E^{2n}$  fits are shown with much larger errors than  $\exp(-c/E)$ -fits in Fig. 4.

For the identification of this  $R^E$  component  $R(T,f,B) \approx (R_{res}^H + R_{BCS}^H(T_o, f) + R^E(E^\perp)) (1 + \gamma^*(B/0.2T)^2 +)$  of Eq. (3.2) has to be used, where the geometry factors are not correct for a non-linear interaction mechanisms i.e. local measurements are more appropriate. For Fig. 5 those are depicted as temperature maps in Fig. 6, showing no pronounced bad spot but critical  $R^E(E^\perp)$  - heating on a ring with  $E \approx E_{crit}$  in this  $TM_{010}$ -mode, which change by surface smoothing to higher  $E_{crit}$  moving closer to the equator where  $R^E$  enhances  $B^2 R_{BCS}(T + \Delta T)$  via  $\Delta T$  yielding breakdown on a ring with coarse spots. The smoothing of the rough, spinned surface by successive BCP [13] reaches  $E_{crit} \approx 30 \text{ MeV/m}$  by successive higher onset field and higher  $n$ -values. Further  $E_{crit}$ -enhancements take advantage of EP yielding  $E_{crit} \geq 40 \text{ MeV/m}$  for onset-fields  $E_{rf} \approx 30 \text{ MeV/m}$ .

Whereas in Nb cavities using bulk Nb degradations with  $\gamma^*(T < T_\lambda) \leq 3$  are typical - see, e.g., Figs. 3 - 6 or [4a, 12,13], sputtered Nb or heavily corroded Nb shows  $\delta R \propto \omega B/j_{cJ}$  of Eq. (4.3) at  $T \lesssim 2 \text{ K}$  depicted in Fig. 7. As worked out in [11] those hysteresis losses are due to Jo-

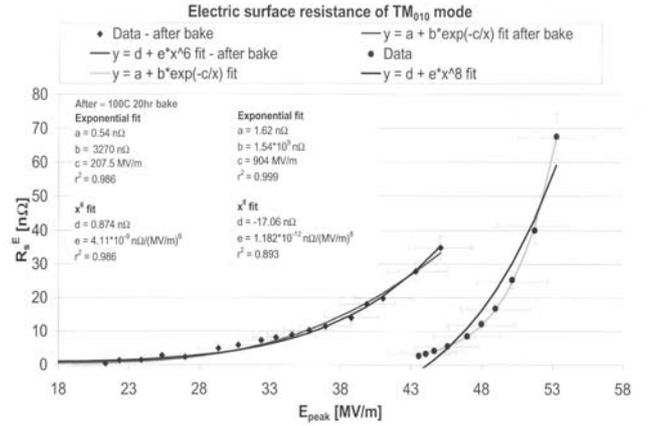
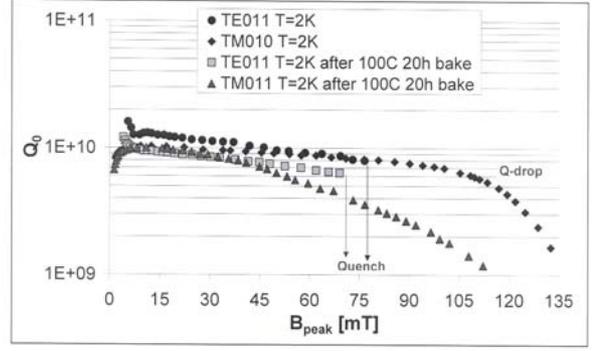


Fig. 4: a)  $Q$  versus  $B_p$  in  $TM_{010}$  ( $= 1.47 \text{ GHz}$ ) and  $TE_{010}$  ( $= 2.83 \text{ GHz}$ ) modes without and with  $100^\circ\text{C}$  baking for 20 hr [14]. The  $Q$  ( $B \approx 10\text{mT}$ ) drop in the  $TE_{010}$  mode, like the low quench fields  $B_p \approx 10\text{mT}$  are indicative for defects, which corrode faster as shown by the strong  $R_{res}$  increase by baking in Table 1.

Fig. 4b) Whereas the  $TE_{011}$  modes show no sign of  $R^E \propto \exp(-c/E_p)$  losses the  $TM_{010}$ -modes allow with Eq. (3.1) to deduce  $R^E(E) = R(T_o B)/(1 + \gamma^*(T_o)(B_p/0.2T)^2) - R(T_o \approx 15\text{mT})$  which fits to  $\exp(-c/E)$  excellently, in lowest order ( $\sim B^2$ ) approximation [14].

Table 1: Fit constants to the data represented in Fig. 4 of modes in a multimode cavity containing defects causing low field  $Q(B)$  decrease and early breakdown in the TE-mode [14]. Also the strong degradation of  $R_{res}$  and  $\gamma^*$  by baking links to defects promoting corrosion.

	Before baking		After baking	
	$TM_{010}$	$TE_{011}$	$TM_{010}$	$TE_{011}$
$\gamma$ @ 2.425 K	13	38	20	39
$\gamma$ @ 2.2 K	15	34	26	62
$\gamma$ @ 2 K	1.7	4.6	8.4	4.6
$R_{res}$ [nΩ]	17	13	22	29

Josephson fluxon penetration along weak links. At higher fields and  $R_{BCS}(T) \gtrsim R_{res}$  the  $R_{hys} \propto \omega B/j_{cJ}$  - increase is negligible compared to the  $\delta R \propto \gamma^*(B/B_c)^2$  increases shown in Fig. 8 with  $\gamma^* \approx 20 - 90$ , as being discussed below.

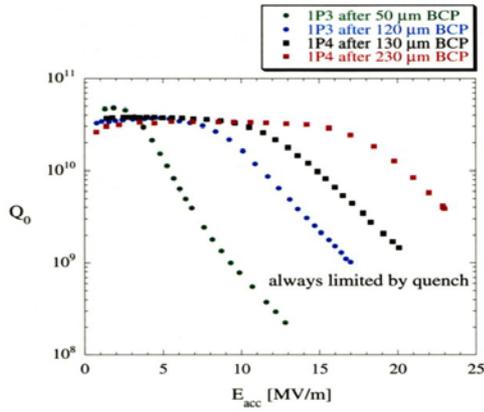


Fig. 5:  $R(2K, B)$  of a Nb  $TM_{010}$ -mode cavity at 1.3 GHz spun of RRR  $\approx 30$  sheath (P2 and P3). The onset fields  $E_p^0 = 6.3; 13.5; 19; 29$  MV/m correspond to  $\delta R^E \propto E^{1n}$  with  $n = 4; 6; 8; 10$  after successive BCP smoothing up to 230  $\mu m$ . Similar changes with BCP-smoothing are reported in [4a].

## DISCUSSION

As mentioned in the introduction, [4a, 13] are not repeated in detail here, but new facts related to 100°C baking and prebreakdown and rf breakdown are elaborated. The BCS surface resistance  $R_{BCS}(T, f, \approx 10B_{max})$  – see Sect. 2 – shows  $\Delta/kT_c \approx 1.85$ , i.e. 10% below intrinsic values and reduced (surface) mean free paths  $l_s$  [4a] by oxidation sketched in Fig. 1. With 100°C baking ( $> 10h$ )  $R_{BCS}(T, \sim 1.5GHz, \approx 10mT)$  is reduced to a saturation value, e.g., by a factor 2 for high quality BCP cleaned Nb. By baking the  $R_{BCS}(T, f, B_{max})$  peak grows [4a, 15]. All those baking observation are explained by the growth of  $NbO_x$  clusters in size yielding enhanced smearing of the BCS density of states (Eq. (2.2)) reducing  $R_{BCS}(< 10 GHz)$  and enhancing the  $R_{BCS}$ -peak, yielding for  $B \geq B_{max}$  rf induced superconductivity [10], stabilizing those  $NbO_x$ -regions up to  $T_c$  and  $B_c$  of the bulk [3b].

Rf residual losses  $R_{res}(T, f) \approx 2n\Omega (f/GHz)^2$  are due momentum transfer by surface scattering being enhanced by localized states  $n_L$  in  $Nb_2O_5$  and by the leakage current via those normal conducting  $n_L$ -states (Eq. (4.2)). Hence  $R_{res}$  grows by oxidation – see Table 1 or Fig. 4 and [4,13] – the more, the more defects exist in Nb acting as nucleation for crack corrosion. For extended weak links  $w \gg \lambda_J$  (Eq. (4.2)) Josephson fluxon are created for fields above  $B_{c1J}$  causing hysteresis losses  $R_{hys} \propto \omega B/j_c$  of Eq. (4.3). Those are observed not only for sputtered Nb films in Fig. 7 but for heavily oxidized Nb also [16]. Aside of the magnetic residual losses  $R_{res}^B \propto f^\alpha$  ( $\alpha \approx 1-2$ ) also standard dielectric losses of  $Nb_2O_5$  or by interface tunnel exchange of conduction electrons  $n_m$  with localized states are found – see Eqs. (1.1) - (1.3). Those losses exist in TM-modes only and explain their higher rf residual losses, e.g. in Table 1. But with fields  $E > E_{rf}$  of Eq. (4.3) exponentially increasing losses with  $c = 2\kappa\Delta\epsilon_r/e\phi^*$  occur, where the local electric field of Fig. 6 has to be used initiating via  $B^2R_{BCS}(T+\Delta T)$  and  $B > 0.7B_p$  the rf breakdown. Shown in Fig. 4b is excellent agreement with theory in contrast to  $E^{2n}$  ( $n \geq 4$ ) fits. The increase of onset

fields  $E^0$  in Fig. 5 are related to smoothing of the surface decreasing  $\beta^*$  shifting the  $R_{BCS}$ -breakdown to the equator, as will be analyzed below.

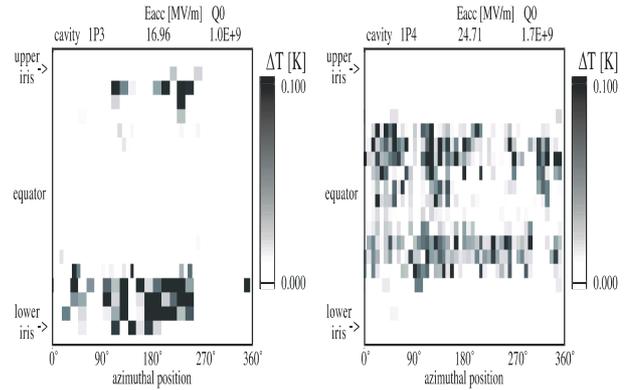


Fig. 6: Heat maps at different field levels for spun Nb cavities P3 and P4 of reactor grade Nb (RRR  $\approx 30$ ) at field just below the quench at  $B_{crit}$  showing in this field region an  $E^{\perp}-Q_0$ -drop with  $6 \leq n \leq 12$  in Fig 5 [4a]. The maximum temperature rise was 0.4 K in order to display also areas with lower heating. The rotational symmetric heatings, which do not show a pronounced spot typical for impurities or stripes for field emission, shifts with increasing  $E_{rf}$  away from  $E_{max}$  towards  $B_{max}$  adding there to the temperature dependent losses  $R^H(T) B^2$ . For example for the  $E_{acc} \approx 17MV/m$  breakdown the heat pulse develops at  $E_p \approx 33MV/m$  and  $B \approx 50mT$  and for  $E_{acc} \approx 25MV/m$  at  $E_p \approx 40 MV/m$  but  $B_p \approx 100 mT$ .

The field dependences in Figs. 3 – 8 always contain for  $B \gtrsim 10 mT$  an  $\gamma^*(T) (B/0.2T)^{-2}$ -increase. Remarkable is the jump of  $\gamma^*(T_\lambda)$  in Figs. 3 and Table 1 by factors around 4 [4a, 13] proving the dominance of the Kapitza boundary resistance  $R_K$  over the thermal conductivity through Nb and proving that the BCS like  $B^2$  increase is heating. The dominance of  $R_K$  is in line with ballistic heat transfer through 2 – 3 mm bulk Nb due the long phonon (26mm) and inelastic electron ( $\sim 2mm$ ) mean free paths [13] mentioned in Sect. 3.2. The  $\gamma^*(T_\lambda)$  jump indicate also that Nb grain boundaries are not effectively scattering phonons for  $T \approx T_\lambda$ .

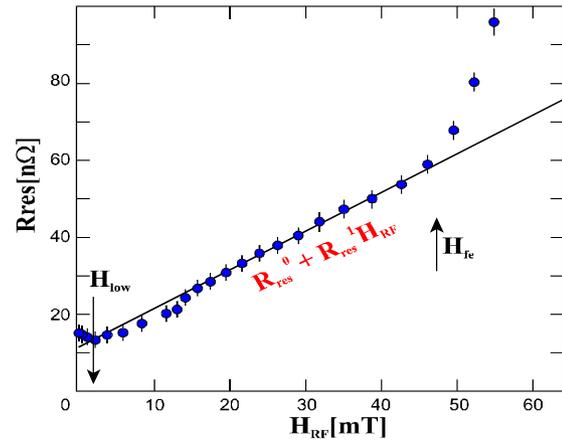


Fig. 7: Dependence of the residual resistance  $R(T \leq 2K, B)$  on  $B_{rf}$  for a typical film on oxidized Cu with  $a_s \approx 0.1 \mu m$ . The solid line represents the linear fit used to define  $R_{res}^0$  and  $R_{res}^1 \propto R_{hys} \propto \omega B/j_{cJ}$  – see Eq. (4.3) and [16]

The other remarkable results shown in Fig. 3 and [12] are: a factor 4 higher  $R_{res}$  values relate to a factor 10 larger

$\gamma^*(T < T_\lambda)$  values and in Fig. 8 [16,17] for Nb films sputtered onto Cu  $\gamma^*$  is enhanced to values of about 60 decreasing to about 20 at 4.2 K. Whereas  $\gamma^*(T < 2K) \approx 0.25$  [12a] is in line with measured Kapitza resistances [14],  $\gamma^* \approx 2 - 90$  can neither be explained by any boundary resistance Nb/He or Nb/Cu measured to date. But the enhanced  $R_{res}$  accompanied by a  $\gamma^*$  increase by a factor 10 or the locally enhanced losses  $R_{hys}$  by weak links in Fig. 7 accompanied by  $\gamma^* \approx 90 - 16$  offer a plausible explanation by locally confined rf losses being transferred with negligible diffusive smearing to the Nb/He or Nb/Cu interface. Hence  $\gamma^*$  in Eq. (3.2) has to be multiplied by the energy pile-up, e.g., for Fig. 7 with hysteresis losses  $R_{hys}$  and  $R_{res}$  confined in weak links of  $d(Nb_2O_5) \approx 2nm$  width in distances  $a_j \approx 0.2\mu m$  enhances  $R_{Nb/Cu}$  by a pile-up factor  $r_k \approx 2nm/a_j \approx 100$  confirmed by  $\gamma^* \approx 16 - 60$  in Fig. 8. The temperature dependence of  $\gamma^*(T)$  [16] is in line with  $R_{Nb/Cu} \propto T^{-4}$  in Eq. (3.2) and the fact that in Fig. 8 no  $\gamma^*(T_\lambda)$  jump occurs, confirming our model of dominant interface resistances  $R_k$  adjacent to the origin of rf losses and that the interface Nb/Cu and Cu yields a diffusive heat transfer with  $r_k$  diminishing to  $r_k \approx 1$  at the Cu/He interface.

In the case of typical, natural oxidation with some weak links depicted in Fig. 1 yielding  $\gamma^*(T < T_\lambda) \approx 2$  a pile-up factor  $r_k \approx 1nm/a_j \approx 10$  is line with the factor 10 enhancement of  $\gamma^*(T < T_\lambda)$  as compared to  $\gamma^*(T < T_\lambda) \approx 0.25$  [12]. This concept of energy pile-up  $r_k$  transferred ballistically to thermal interfaces resistances is obvious also in Tabel 1, where the TE-mode limitation by a defect shows  $\gamma^*(T < T_\lambda)$  being a factor 3 larger than in the TM mode of the same cavity. Also the fact that  $\gamma^*(T < T_\lambda)$  is enhanced by those defects as compared to TM modes prove the validity of the energy pile-up factor  $r_k$ . In [14] the  $\gamma^*(T_\lambda)$  jump is explained semiquantitatively.

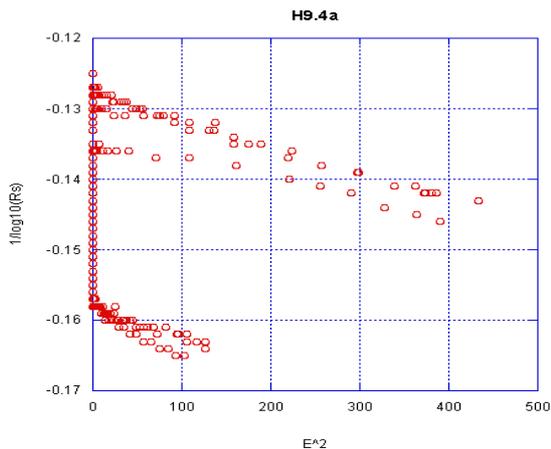


Fig. 8: Plot of  $1/\log R(T,E)$  versus  $E^2$  of a  $3\mu m$  Nb film on Cu. The straight lines for  $R(T,E) > R_{res} + R_{hys}$  prove  $1/\ln R(T + \Delta T, B) \propto (T + \Delta T)/\Delta$  with  $\Delta T \propto E^2$ . Oxidized Cu with  $a_j \leq 0.1\mu m$  shows an  $E^2$  decrease with  $\gamma^*(2K) \approx 55$  decaying to  $\gamma^*(4.2K) \approx 20$  [16,17].

The breakdown in Figs. 3 - 8 is due to thermal runaway via  $R_{BCS}(T)$  needing higher orders in  $\Delta T$  than  $\gamma^*$  alone in Eq. (3.1). But  $\gamma^*(T < T_\lambda) > 0.2$  acts as signature for defects, as shown in Fig. 4, by the defect induced breakdown in the TE-mode. In the TM-modes in Fig. 3 - 5 the  $\exp(-c/E_p)$  triggers the rf breakdown. As shown in the heat maps in Fig. 6 belonging to Fig. 5 the  $\exp(-c/E)$  increases

causes heating amplifying  $R_{BCS}(T)$  triggering rf breakdown. This amplification needs higher order, as in Fig. 8 for  $\gamma^* > 10$ . The combined action of  $\exp(-c/E)$  amplifying  $R_{BCS}(T + \Delta T, B)$  is obvious in Fig. 6 showing rf breakdown at  $E \approx E_p/3 > 40$  MV/m,  $B \approx B_p > 70$  mT and  $E_{acc} \approx 25$  MV/m. The crucial influence of enhanced  $\gamma^*$  by  $r_k > 10$  is obvious in Fig. 4 by the  $B_{crit}$  being reduced by 20% by  $\gamma^* \approx 8$  after baking or in sputter cavities by the low  $E_{acc} \approx 15 - 25$  MeV/m decreasing with  $\gamma^*$  increases [16].

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