

On Extrinsic - Weak Link - Effects in the Surface Impedance of Cuprate - and Classical - Superconductors

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Abstract:

The reported rf surface impedance of superconducting cuprates continues to drop with two years of improving material quality. As leading indicator for quality the weak link critical current $j_{cJ}(T \approx 0, B \approx 0)$ has grown from 10^2 to above 10^6 A/cm² and thus the Josephson penetration depth $\lambda_J \propto 1/\sqrt{j_{cJ}}$ and the excess normal (leakage) tunnel current j_{bl} are shrinking. This j_{cJ} -growth has now saturated, whereas the rf residual surface impedance Z_{res} is still shrinking with material improvements. This shows clearly that Z_{res} is an extrinsic property. We present evidence that Z_{res} is due to the large leakage current j_{bl} and the small j_{cJ} of weak links where the latter destroys the intrinsic shielding from a λ_I -thin seam λ_J deep into the bulk. This causes rf residual losses $R_{res} \approx (\omega\mu_0)^2 \lambda_J^3 \sigma_{bl}/2$. R_{res} stays finite at $T \approx 0$ by $\sigma_{bl}(T \rightarrow 0) \approx \sigma_{bl} (\propto j_{bl})$ being amplified by $(\lambda_J/\lambda_I)^3 > 10^3$ as a weighting factor. With slow crystal growth weak links are improved and their density is reduced so that R_{res} -values better than R_{Nb} (4.2 K) are now obtainable. For Classical superconductors the Josephson coupled weak links are due to crack corrosion or grain boundaries. The grain boundaries occur in large densities in sputter deposition. These weak links cause an rf residual surface impedance which is minimal for minimal weak link boundary resistances being proportional to the leakage resistance $\propto 1/\sigma_{bl}$. Thus, like for YBCO, Z_{res} is minimal for minimal extrapolated resistivity $\rho(T \rightarrow 0)$.

The $j_{cJ}(T, B)$ -values explain λ_{res} and R_{res} quantitatively and in temperature $\propto (a + T^n)$; $n \approx 1$, $T < T_C/2$ and $\propto (b + H^n)$, $n \approx 1$; $H > H_{c1J}$ in field dependence. Here

H_{c1J} is the field where flux enters into weak links as Josephson fluxons having negligible viscous losses, but act by enhancing the penetration depth.

1. INTRODUCTION

Cuprate superconductors show penetration depths $\lambda(T)$ and surface resistances $R(T)$, which because of their magnitude ($10^1 - 10^6$ above BCS) and temperature dependencies ($\propto (T/T_c)^m$, $m \approx 1$, $T < T_c/2$)¹ well above expectations, have been related to, e. g., an energy gap with nodes² or intrinsic normal carriers³. The recently observed field dependencies⁴ ($\propto H^n$, $n \approx 1$, $H \geq 0.1$ Oe) or dc resistance dependence⁵ $R_{res} \propto \rho^2(0)$ ask for new explanations not covered by Ref. 2 or 3. In this note a comparative discussion between the different explanations are not given. Instead, evidence is presented that all "residual rf effects" in: magnitude-, frequency-, temperature-, dc resistance-, and field- dependence can be related to "weak links", as outlined in Ref. 4. Here "weak link" stands for planar defects being weakly superconducting only, i. e., being crossed by a reduced Josephson current j_{cJ} where the reduction is compensated by a normal, leakage current j_{bl} . The weak Josephson coupling yields a long Josephson penetration depth $\lambda_J > 1 \mu\text{m}$ causing the destruction of rf shielding deep into the superconductor. In lowest order approximation in a two fluid model this destruction causes rf residual losses given by⁴

$$R_{res} = \mu R_J = \mu (\mu_0 \omega)^2 \lambda_J^3 \sigma_{bl} / 2 \quad (1.1)$$

with $\mu \leq 0.1$ as geometrical factor describing the effective areal density of weak links and σ_{bl} the normal conductivity across weak links, e. g., caused by leakage currents or fluxoids discussed in Section 2. Crucial in (1.1) is the finite $\sigma_{bl} (T \rightarrow 0) \propto j_{bl} \approx \text{const}$ and the λ_J^3 amplification, being typical for any two fluid model. So, $R_{res} \approx \text{const.}$ dominates over the intrinsic BCS surface resistance $R_I(T, \omega) \propto \exp(-\Delta/kT)$ ($T < T_c/2$):

$$R_{eff} = (1 - \mu) R_I + \mu R_J \quad (1.2)$$

For the effective penetration depth a linear averaging holds in contrast to $R_J \propto \lambda_J^3$:

$$\lambda_{eff} = (1 - \mu) \lambda_I + \mu \lambda_J \quad (1.3)$$

and thus λ_{eff} is much closer to an intrinsic λ_I ⁶.

In Ref. 4 the physics of the rf residual surface impedance is worked out in detail. Here those formulas are elaborated for comparison with experiments. Thus in Section 2 weak links and their parameters are introduced. For example it is shown that, $j_{cJ} \propto 1/R_{bn}^{2n}$ ($n \geq 1$) holds with R_{bn} as weak link grain boundary resistance⁷ being proportional to $\rho(0)$ which yields $\lambda_J \propto 1/\sqrt{j_c} \propto R_{bn}^n$, $\lambda_J \propto \rho(0)$ and $R_{res} \propto \rho^2(0)$.

On the other hand $j_c(T) \propto (1 - T/T_c)^2$ yields $\lambda_J(T) - \lambda_J(0) \simeq \delta\lambda_J \propto T$ and $\delta R_J(T) \propto T$ for $T < T_c/2$. The penetration of fluxoids at H_{C1J} in the weak links causes λ_J and R_J to increase linearly with H where details depend on the specific fluxon dynamics. All these phenomena summarized in Section 2 have been observed experimentally, see Section 3.

2. WEAK LINKS

Weak links as planar defects with reduced normal conductivity, can be described by an enhanced grain boundary resistance $R_{bn}(\Omega\text{cm}^2)$ and a reduced critical Josephson current j_{cJ} in the superconducting state. In YBCO these are usually separated^{1, 5-17} into two main categories according to their experimental appearance⁴: "Intergrain ("J") weak links" include extrinsic grain surfaces occurring often in granular material, e. g., by sintering. "Intragrain ("G") weak links" occur typically in epitaxial films as small (or large angle) grain boundaries. For the classification in Table 1 the weak link critical current is used as leading indicator. This j_{cJ}/G in cuprates is well below expectations using the normal state grain boundary resistance R_{bn} :⁷⁻⁹

$$j_c R_{bn} = \frac{1}{c} \frac{\pi \Delta(T)}{2e} \tanh \frac{\Delta(T)}{2kT}; \quad c \propto R_{bn}^{2n} \quad (T < T_c; n \geq 1) \quad (2.1)$$

holding for S-I-S (superconductor-insulator-superconductor) junctions, whereas for S-N-S (S-normal conductor-S) holds ($t = T/T_c$):

$$j_c(T) \simeq j_c(0)(1 - t)^m \quad (T_c/4 \leq T < T_c; 1 \leq m \leq 2) \quad (2.2)$$

The transition from S-N-S (2.2) to S-I-S (2.1) behaviour shifts to higher temperatures for better junctions, i. e., higher j_c and smaller $c = j_{bn}/j_c$ - values see Table 1. The j_c -reduction, i. e., $c > 1$, is compensated by a "quasi normal leakage current" j_{bl} or leakage grain boundary resistance R_{bl} :

$$j_{bl} \simeq j_{bn} - j_c = j_{bn} \left(1 - \frac{1}{c}\right); \quad R_{bn} = R_{bl} \left(1 - \frac{1}{c}\right) \quad (2.3)$$

This normal, leakage current defines the residual losses via σ_{bl} in Eq. (1.1) by

$$\begin{aligned} \frac{1}{\sigma_{bl}} &= \left(R_{bl} - R_{bn} |H| / \left(H(T) c^2 u(T) \right) \right) / 2\lambda_1(T) \\ &= \left(R_{bn} / \left(1 - \frac{1}{c}\right) - R_{bn} |H| / H(T) c^2 u(T) \right) / 2\lambda_1(T) \end{aligned} \quad (2.4)$$

In Eq. (2.4) the first term $\rho_{bl} = R_{bl}/2\lambda_1(T)$ describes a normal resistance for the shielding current destroyed along a weak link deep ($\sim \lambda_J$) into the YBCO in a $2\lambda_1 = 2\lambda_{BCS}(T)$ wide seam. This term corresponds to a resistively shunted Josephson junc-

Table I: Parameters characterizing weak links and bulk, intrinsic YBCO at $T = 0$ assembled in Refs. 4 and 9 from experiments. The abbreviations "J" or "G" or "I" are subscripts added to R_b , j_c , λ_J , H_{c1} or H_{c2} as needed for clarity. c is the ratio of the normal weak link current and supercurrent and d the estimated insulator thickness, see references 4 and 9. The critical currents $j_{cJ/G}$ cited are Josephson critical currents which together with pinning yield the actually measured critical current $j_c > j_{cJ/G}$. The fluxoids entering at H_{c1} are Josephson fluxons for intergrain weak links turning to a more Abrikosov - like fluxon for the "G" - and "I" - system.

Weak link	abbr.	$R_b/\Omega\text{cm}^2$	$j_c/\text{A}/\text{cm}^2$	$\lambda_J/\mu\text{m}$	H_{c1}/Oe	$H_{c2}/100\text{ Oe}$	c	d/nm
insulator		∞	0	∞	-	-	-	> 6
intergrain	J	$\geq 10^{-6}$	$\sim 10^2$	~ 30	≈ 1	1	$\approx 10^2 \cdot 10^3$	≈ 2
intragrain	G	$\approx 5 \cdot 10^{-8}$	$\geq 10^4 - 10^6$	≈ 1	≈ 100	> 100	≈ 10	≈ 1
intrinsic	I	0	$\leq 2 \cdot 10^8$	0.14	≥ 1000	> 1000	1	0

tion (RSJ) analyzed in Ref. 10. The second term in (2.4) describes the residual losses by fluxon ($\propto |H|/H_{c2}$) motion along weak links having a negative sign due to the electric field being induced by this fluxon motion. The resistance is given by $R_{bn}/2\lambda_I(T)$ being much larger than $\rho(T)$ of YBCO crystallites. In Section 3.4 the fluxon dynamics are discussed for the surface impedance, especially the difference between Abrikosov fluxons ($u \approx 1$) and Josephson fluxons being highly correlated described by $u(T) \gg 1$. Such fluxons may exist as residual flux H_{res} , already, or may penetrate above H_{c1J} with the characteristic nucleation frequency $f_N \approx 10^{11}$ Hz⁴. The grain boundary resistance R_{bn} in Eq. (2.4) can be measured by $\rho(T)$ of strips more narrow than the grain size t_G , so that no parallel shunting occurs⁷. Then $R_{bn} \approx t_G \rho(0)$ holds⁹.

The superconducting critical Josephson current (2.1) is related to a penetration depth:

$$\lambda_J(T, H) = \sqrt{\hbar/2e j_c(T) 2\lambda_I(T)} > \lambda_I \quad (2.5)$$

describing the reduced supercurrent shielding at weak links as compared to the intrinsic magnetic field penetration depth $\lambda_I \approx 150$ nm. Because weak links have small flux entry fields H_{c1J} , fluxons enter and enhance the Josephson penetration depth for $|H| > H_{c1J}$ by a mean reduction of shielding by $j_{cJ}(T, H)$ (Eq. (2.5)) as summarized in Ref. 4. This flux entry field is for Abrikosov fluxons with $H_{c1} = \Phi_0 / \ln \lambda_I / \xi_{G1} / \mu_0 \lambda_I^2$ ($\Phi_0 =$ flux quantum, $\xi_{G1} =$ Ginzburg Landau coherence length) quite large. Because of $H_{c1} \propto 1/\lambda^2$, H_{c1} is already reduced for Josephson fluxons by $\lambda_J/\lambda_I \approx 10$ (Eq. 2.5) to

$$H_{c1J} \approx \frac{\Phi_0}{\mu_0 \lambda_J^2(T)} \ln \frac{\lambda_J}{\xi_{G1}} \quad \text{and even more to}$$

$$H_{c1H} \approx \frac{\Phi_0}{\mu_0 \lambda_H^2} \ln \frac{\lambda_H}{t_G} \quad (2.6)$$

for Hyper fluxons³⁰, being extended over many grains of size $t_G \ll \lambda_I$. Then the penetration depth

$$\frac{1}{\lambda_H^2} = \frac{4e j_C t_G \mu_0}{\hbar} \quad (2.7)$$

is enhanced by the new length scale t_G substituting λ_I or ξ_{G1} . It should be mentioned that the meaning of H_{c2} in this context of Josephson or Hyper fluxons is the instability against a "normal state" in this intergranular matter. But a super current may be still carried across weak links by narrow microbridges above H_{c2J} .

3. COMPARISON WITH EXPERIMENT

The rf field dependencies have been modelled, discussed and compared with experiments in Ref. 4. This model, also summarized in Section 2, and new experimental information on $\rho(0)$, λ_{res} , R_{res} and their ω -, T- and H-dependencies are analyzed in the following. For this analysis we use Eqs. (1.2) and (1.3) referring for a more quantitative treatment to Refs. 10 and 17. In these equations the areal ratios μ enter: for intergrain weak links cristallite sizes t_J around $10 \mu\text{m}$ are typical with $\lambda_I = 0.15 \mu\text{m}$ yielding $\mu_J \approx 1 \%$. For intragrain weak links occuring, e. g. in epitaxial \hat{c} -axis films, subgrain or island sizes around $t_G \approx 1 \mu\text{m}$ ^{11, 18} are typical, yielding $\mu_G \approx 10 \%$. For \hat{a} -axis films $t_G \approx 0.2 \mu\text{m} \approx \lambda_I$ are found¹⁷ asking for the more refined treatment carried out in Ref. 17. For single crystals or melt-textured growth¹⁹ $t_G \approx 40 \mu\text{m}$ yields $\mu_G \approx 0.4 \%$.

3.1 Parameter dependencies of the weak link surface impedance Z_J

In Section 2 in Eqs. (2.1)-(2.5) the dependencies of Z_J on: "weak link quality" R_{bn} ($\propto \rho(0)$), temperature $t = T/T_c$ and magnetic field H/H_c are given implicitly. Explicitly we get for the temperature dependence $j_{cJ/G} \propto (1-t)^m$ ($1 \leq m \leq 2$) for the intermediate temperature range $T_c/4 < T < T_c$, where most surface impedance measurements have been analyzed (Eqs. (3.1)-(3.4)). Starting with Eq. (2.5) one obtains for the T-dependence:

$$\begin{aligned} \lambda_{J/G}(T) &\propto 1 / \sqrt{j_{cJ/G}(T) \lambda_I(T)} \propto R_{bn}^n / \left[(1-t)^{\frac{m}{2}} \sqrt{\lambda_I(T)} \right] \\ &\propto R_{bn}^n \left(1 + m/2 t \right) / \sqrt{\lambda_I(T)} \end{aligned} \quad (3.1)$$

$$\begin{aligned} R_{J/G}(T) &\propto \lambda_{J/G}^3 \sigma_{bl} \propto R_{bn}^n / \left[\left(1 - 1/c(T) \right) \lambda_I(T) \left(1 - t \right)^{\frac{3m}{2}} \sqrt{\lambda_I^3(T)} \right] \\ &\propto R_{bn}^{2n} \left(1 + 3m t/2 \right) / \left[\left(1 - 1/c(T) \right) \sqrt{\lambda_I^5(T)} \right] \end{aligned} \quad (3.2)$$

for $T_c/4 \leq T < T_c$. These equations contain a linear increase with t by $j_{cJ}(T)$, but via $H_c(T) \approx H_c(0) (1-t^2)$ a t^2 increase occurs in Z_J by 4:

$$\lambda_{J/G}(T, H^*) \approx \lambda_{J/G} \left(T, H_{c1J/G} \right) \left(1 + H^*/2H_{c2J/G}(T) \right) \quad (3.3)$$

and by reactively oscillating fluxons in a pinning potential as described by 20, 21:

$$\lambda_p(T, H^*) \approx \lambda(0, H^*) / (1 - t^2)^u(T) \quad (3.4)$$

The dependencies of $R_{J/G}(T, H)$ on field $H > H_{C1J/G}$ are in first order given by:

$$R_{J/G}(T, H^*) \approx (\omega\mu_0)^2 \lambda_{J/G}^3(T, H_{C1J/G}) \left(1 + \frac{3}{2} \frac{H^*}{H_{C2J/G}} \right) \left[\sigma_{bl} + \sigma_{bn} \frac{H^*}{H_{C2J/G}} \right] / 2 \quad (3.5)$$

The fluxons oscillating in a pinning potential may be of external origin $H^* \approx H_{dc}$ or $\approx H_{rf}$ or due to frozen - in flux $H^* \approx H_{res}$, which may consist of vortex - antivortex pairs, created at defects 21. This latter term is the -low frequency- real part of the fluxon viscosity of Eq. (2.4) 23. It should be mentioned that for films of thickness t_F smaller than λ_J or λ_G (Table 1), an effective, reduced impedance Z^* is to be introduced:

$$Z^* \approx (R_{J/G} + i\omega\mu_0\lambda_{J/G}) / \coth t_F/2 \lambda_{J/G} \quad (3.6)$$

This changes the dependencies of the weak link impedance and, e. g., for $t_F \gg \lambda_{J/G}$ in Eqs. (3.1)-(3.5) λ is substituted by $t_F/2$.

One word about the critical currents. In Eq. (3.1) and (3.2) $j_{cJ/G}$ is the Josephson critical current and not a transport critical current. In general, the pinning critical current $j_{cp}(T, H)$ is larger than j_{cJ} or $j_{cG}(T, H)$. For long Josephson junctions $t_G > \lambda_G$ pinning causes $j_c > j_{cG}$. Thus for \hat{c} -axis epitaxial films with $t_G \geq 1 \mu m$ 18 $j_c \approx 10^7 A/cm^2$ may correspond to $j_{cG} \approx 10^6 A/cm^2$ and thus $j_{cp}(T, H)$ dominates the T and H dependence of j_c . This is in contrast to \hat{a} -axis films with $t_G \approx 0.2 \mu m \approx \lambda_I$ where weak links and pinning sites are never spatially separated. For such films $j_c \approx j_{cG}(T, H) \leq 10^6 A/cm^2$ is a good approximation and found experimentally 17.

3.2 Dependence of Z_{res} on R_{bn}

For standard sintered YBCO with $\mu_J \approx 1\%$ and $\lambda_J/\lambda_I \geq 2 \cdot 10^2$ intrinsic properties may show up in λ_{eff} very close to T_c , only. That intergrain weak links dominate below T_c is shown for granular YBCO 1 by the large quasilinear T-increase with

$$\Delta\lambda_{0.5} = \left[\lambda(T_c/2) - \lambda(0) \right] / \lambda(0) \approx 0.2 - 0.3 \quad (3.7)$$

This results from Eq. (3.1), but is much larger than the BCS value $\Delta\lambda_{0.5} \leq 5\%$ 6. In the surface resistance given by Eq. (1.1), $\mu_J (\lambda_J/\lambda_I)^3 > 8 \cdot 10^4$ makes any identification of intrinsic properties impossible, where our extrinsic two fluid $R_{eff} \propto \lambda_J^3$ -dependence actually has been observed experimentally 12. Quantitatively, Eq. (1.2) yields for granular YBCO:

$$R_{\text{effJ}} = R_{\text{res}} \approx \mu_J R_J \leq 10^{-4} \Omega (\ell/\text{GHz})^2 \quad (3.8)$$

which is in line with observations 1,4,12. The orientation dependence of R_{res} in Ref. 14 is described by $R_{\text{res}} \propto 1/\sqrt{j_{cJ}^3}$ with the measured j_{cJ} anisotropy of the textured YBCO. This shows, that these residual losses are not dependent on $\mu\sigma_{bl}$. This was proven by an identical H-field dependence 14 in both orientations, indicating that these losses are due to \hat{a} - \hat{b} -plane weak links. In summary, rf residual losses due to intergrain weak links are dominated by the weighting factor λ_J^3 because the normal leakage current is always maximum $\sigma_{bl} \approx \sigma_{bn}$ in Eq. (3.2).

For intragrain weak links, smaller $(\lambda_G/\lambda_I) \approx 10$ values allow the identification of the intrinsic temperature dependence of the penetration depth λ_I (T) for material with small μ_G , as discussed in Section 3.3. For epitaxial films with large angle intragrain boundaries, $\lambda_J \approx 5 \mu\text{m}$ seems typical 11. Then with $\mu_G \approx 10 \%$, the weak link λ_J and λ_I are roughly in line with $\lambda_{\text{eff}}(0) \approx 0.4 \mu\text{m}$ reported in Ref. 13. But the residual losses $R_{\text{eff}} \propto \mu_G(\lambda_G/\lambda_I)^3 \approx 10^2$ are still dominated by intragrain weak links (Eq. (3.2)):

$$R_{\text{effG}} = R_{\text{res}} \approx \mu_G R_G \leq 10^{-6} \Omega (\ell/\text{GHz})^2 \quad (3.9)$$

For this type of material the correlation of $R_{\text{res}} \propto (\lambda_G/\lambda_I)^3 \propto R_{bn}^2 \propto \rho^2(0)$ has recently been observed, 5, 14 by a reduction of large angle intragrain weak links 5 for epitaxial films with identical substrates MgO and identical dp/dT , i. e., identical μ . Further reductions, of R_{res} , e. g., by a reduction of μ_G and j_{bl} in single crystals, 16 are already underway guided by the $R_{bn} \propto \rho(0)$ -value.

3.3 Temperature dependence of Z_J

The temperature dependencies of $Z_{\text{eff}}(T)$ are not only defined by $\lambda_J(T)$, $\lambda_G(T)$ or $\lambda_I(T)$ or $H_c(T)$ but also by $\sigma_{bl}(T)$. For intergrain weak links, the low H_{c1J} -field smaller than 1 Oe (Table 1) 4 is also influencing the experiments (Eq. (2.4)), but the H-field is usually not stated in these publications. Thus the temperature dependencies $\delta Z \propto T^n$ ($n \approx 1$) 1 are not only given by $\delta \lambda_J \propto T/T_c$ (Eq. (3.1) and (3.2)) but also by $H_{cJ}(T) \propto (1 - t^2)$. Thus nothing quantitative can be said, aside from stating that $R_J(T) \gg R_I(T)$ holds and that in intergrain weak links λ_J and R_J dominate. Qualitatively, a $1/(1-t^2)$ -dependence suggests fluxons 20,21 and $\Delta\lambda_{0.5} > 0.1$ (Eq. (3.7)) suggests weak links.

For intragrain weak links, large H_{c1G} fields (Table 1) allow experimental results not influenced by the field dependence of Eq. (3.3) 4. Eq. (1.3) yields with (2.1) as

effective penetration depth for $T < T_c/2$ with Δ_I the intrinsic - and Δ_G the weak link energy gap:

$$\lambda_{\text{eff}} \approx (1 - \mu) \lambda_I(0) \left(1 + \exp(-\Delta_I/kT) \right) + \mu \lambda_J(0) \left(1 + \exp(-\Delta_G/kT) \right) \propto c + T^n, \quad (3.10)$$

where the $\exp(-\Delta/kT)$ -dependence with $6 \ 2\Delta/kT_c \approx 2.5$ yields a better fit than T^n for improved film quality. This is explained by the exponential dependence in $j_{cG}(T)$ (Eq. (2.1)) for S-I-S junctions ^{7, 8}. Using $j_{cG}(0) \approx 10^6$ A/cm², $\lambda_G(0) \approx 0.33$ μm is obtained, which fits quantitatively ⁶ for subgrain island sizes of $t_G \approx 1$ μm ¹⁸. At temperatures $T \geq T_c/2$, a BCS-dependence with $2\Delta_I/kT = 4.5$ was assigned ⁶ to be intrinsic. This two component model ⁶ for $\lambda(T)$ was confirmed by \hat{a} -axis films with $t_G \approx 0.2$ μm showing $2\Delta/kT_c \approx 2.5$ up to T_c , i. e., the higher density of weak links causes their dominance in $\lambda_{\text{eff}}(T)$ up to T_c with $\lambda_{\text{eff}}(0) \approx 280$ nm $\approx \lambda_G(0)$. It should be mentioned, that with increasing amounts of extended defects vortex-antivortex pairs are frozen in, causing $\lambda_p(T) \approx \lambda_p(0)/(1-t^2)$ ²¹, see Eq. 3.4 and Section 3.4.

Whereas the penetration depth $\lambda(T)$ for good \hat{c} -axis films already shows "BCS-type" behavior ⁶, the intrinsic $R_I(T)$ is much more difficult to observe in the surface resistance. It might be that the drop of $R(T)$ by more than 2 orders at 90 GHz for melt textured material with 40 μm as weak link separation ¹⁹ is already this intrinsic $R_I(T)$ ¹⁶. But all other results known to the author indicate that for weak links $R_J \propto \omega^2 \lambda_J^3(T, H) \sigma_{bl}(T, H)$ dominate. E. g., for \hat{c} -axis films, subgrain sizes of 1 μm , as deduced by STM, ¹⁸ can account for R_{res} quantitatively ^{5, 17}. For \hat{a} -axis films with 0.2 μm weak link distances, $j_c(T) \approx j_{cG}(T)$ holds. Using measured $j_{cG}(T)$ -values, $R_{\text{res}}(T)$ was fitted in magnitude and T-dependence ¹⁷.

For \hat{c} -axis films and single crystals weak links are well separated by distances in excess of 1 μm , i. e. large compared to λ_I . Thus $j_c(T)$ measured is dominated by pinning inside subgrains and not by $j_{cG}(T)$ of the weak links. Under these circumstances $R_{\text{res}}(T)$ -dependencies have been observed with a plateau around 40-70 K and decreases - or increases - below this plateau. These strange dependencies may reflect $\sigma_{bl}(T)$ dependences due to $j_c(T)$ or due to fluxon dynamics (Eq. (2.4)), as discussed in the next section.

3.4 Field dependence of Z_J

The magnetic field dependence of the surface impedance is complex, because different types of fluxoids are involved. Josephson fluxons exist in intergrain weak links, whereas the Abrikosov character grows for intragrain weak links to actual-

ly Abrikosov fluxons inside the grains. These differences correspond to different rf losses, being smallest for Josephson fluxoids where the "insulating core" cause negligible rf losses, if an rf current is driven through the fluxon ²² as described in Eq. (2.4) by the enhanced weak link resistance R_{bn} and correlation volume $\propto u(T)$. This corresponds to fluxon viscosity being 5 orders of magnitude smaller for a Josephson fluxon than for an Abrikosov fluxon ²²⁻²⁴. For example, the rf losses of Abrikosov fluxons are described by their viscosity:

$$R \approx R_{nc} |H| / H_{C2} \quad \text{with} \quad R_{nc} \propto \sqrt{\omega} \quad (3.11)$$

the normal skin effect surface resistance ³¹. In contrast, the rf losses of Josephson fluxons have a negligible viscous part (Eq. (2.4)) and are dominated by the leakage current as described in Eq. (3.5) by

$$\frac{d R_{J/G}}{d H} \approx \frac{3}{2} R_{J/G}(T, H_{C1J/G}) / H_{C2J/G} \quad (3.12)$$

This equation has been proven for the classical granular superconductors, namely, sputtered Nb and NbN discussed in Section 3.5, and seems to explain YBCO data also ¹⁴. There $H_{C1G} \approx 200$ Oe has been found which fits according to Eq. (2.6) $H_{C1G} \propto 1/\lambda_G^2$ with $\lambda_G/\lambda_1 \geq 3$, i. e., $j_{CG} \approx 10^6$ A/cm² - see Table 1.

But this is only part of the story: in an rf field correlated fluxons are hindered in their response by a pinning potential ¹⁹ and by an effective mass ²⁵. That is, they can perform a correlated motion only. This is most clearly shown for Josephson fluxons which can move only as a linear array. We summarize this in $u(T) \gg 1$ in Eqs. (2.4) (3.4) and (3.5). The pinning potential is described by an activation energy U_0 being proportional to $u(T)$ and to the correlation volume V_c , which is then, e. g., proportional to the length of the weak link. Because the "effective mass" of Josephson fluxoids also grows with V_c , an array of intergrain fluxons will not respond ²⁵ to an rf field, as found experimentally ²⁴. In addition, this response being proportional to $1/u(T) \propto 1/U_0(T, B)$ reflects the temperature dependence of $U_0(T, B)$. This $U_0(T, B)$ increases with T up to a maximum between 50-80 K, depending on B-field strength ¹⁹. Thus $u(T)$ has a similar dependence.

With the above introduction the weak link surface impedance $Z_{J/G}$ is discussed in the following. As obvious from $u(T) \gg 1$, fluxons in intergrain (intragrain?) weak links will not respond to an rf field. Thus the surface impedance increases with fluxon density mostly because $\lambda_{J/G}(T, H)$ increases, according to Eq. (3.3). As a consequence, $R_{J/G} \propto \lambda_{J/G}^3 \sigma_{bl}$ grows accordingly and this growth can be used to measure $H_{C1J/G}$ by the onset of the linear increase and $H_{C2J/G}$ by the slope given in Eq. (3.12). The increase of $\lambda_{J/G}(H)$ obviously depends on the fluxons in the weak

links only, and thus on their overall spatial distribution and orientation. The spatial distribution might explain why the losses in the zero field cooled case (ZFC) are larger than in the field cooled case (FC) ²⁶: In ZFC, fluxons are piled up at the surface causing larger $\lambda_{J/G}(H)$ - and thus larger $R_{J/G}$ -values. The activation energies $U_0 \approx 0.1$ eV observed ²⁶ at about 100 Oe fit to fluxons in intragrain weak links (Table 1) - see also Ref. 19.

For polycrystalline, isotropic material, an orientation dependence with H will not show up, in contrast to textured material ¹⁴. For textured samples, the above formula has been developed ⁴ for $H_{rf} \parallel H_{dc}$, i. e., the case when the dc field is parallel to the surface and to the weak links sampled by the rf shielding currents. Thus for dc fields perpendicular to the surface or perpendicular to the weak link planes, fluxons penetrate the weak links only in a small areal ratio. Thus, $\lambda_{J/G}(H)$ and $R_{J/G} \propto \lambda_{J/G}^3$ are not enhanced according the "H-field" but by a much smaller amount. Whereas these enhanced rf losses are due to Josephson fluxons enhancing the Josephson penetration depth, non-linear effects of frozen - in flux are governed by Abrikosov fluxons. This is due to their smaller correlation volume $\propto u(T)$, as has been confirmed experimentally ²⁴. In addition, Abrikosov fluxons yield $R(H_{dc\perp}) > R(H_{dc\parallel})$ because of "normal conducting cores" ending at the surface for the field H_{dc} perpendicular to the surface (Eq. (3.11)). This is in contrast to Josephson fluxons with $R(H_{dc\perp}) \leq R(H_{dc\parallel})$ because of reasons mentioned above.

Comparing $Z_{J/G}(H_{dc})$ and $Z_{J/G}(H_{rf})$ for H_{dc} parallel H_{rf} , $Z_{J/G}(H_{rf})$ will be larger for frequencies below the nucleation frequency ($\sim 10^{11}$ Hz). This is partly due to causes mentioned above for ZFC-FC: for rf fields the fluxons pile up at the surface. In addition, $R_{J/G}(H_{dc}) < R_{J/G}(H_{rf})$ holds, because fluxons have to be created and annihilated at the surface before they migrate into the interior. Qualitatively, $R(H_{dc}) < R(H_{rf})$ has been found ²⁷ experimentally. It should be mentioned that the increase in $\lambda(H_{rf})$ causes a decreasing eigenfrequency $\omega(H)$ and nonlinear effects generating odd harmonics ²⁸.

The typical case for rf residual losses of "epitaxial films" or "single crystals" holds for $H < H_{c1G} \approx 100$ Oe, i. e. no flux enters into the YBCO. Then only frozen - in flux or vortex - antivortex pairs exist and the losses are described by $R_{J/G} \propto (\lambda_{J/G})^3 \sigma_{bl}$. σ_{bl} is due to two mechanisms (Eq. (2.4)):

- The actual leakage current $j_{bl} \approx j_{bn} - j_c$ increasing with T as suggested by $j_c \propto (1-t)^m$.

- Oscillating fluxoids being governed by $1/u(T)H_{c2}(T)$ as T-dependence. Because $u(T) \propto U_0(T, B)$ increases with T with a maximum around 70 K, this loss mechanism will have a minimum around 70 K.

Thus the actual losses are given by a term increasing with T and one term decreasing below 50-70 K with T, where details depend very much on fluxons and their interaction and pinning. As indicated in Eq. (3.4), $\lambda(T)$ may increase similarly. With irradiation, the losses first decrease²⁹ due to enhanced pinning, i. e., u-increase. With further damage by irradiation the leakage current j_{bl} , and thus R_{res} , increases. Then T-dependencies by $j_c(T)$ and $u(T)$ are no longer visible and $R_{res}(T \leq T_c/2) \approx \text{const}$ holds²⁹. The nowadays "best YBCO" with $\rho(0) \leq 0$ show^{14, 29, 42} the mentioned T dependencies, i. e., the plateau around 70 K and the decrease of R_{res} for $T < 50-70$ K confirming the above model. But much more work is needed to pin down the proposed mechanisms and to reduce the known 0-loss from weak links reducing j_{cJ} adjacent to the surface.

3.5 Classical superconductors

For bulk, smooth superconductors the field penetration by fluxons is hindered by a surface barrier to fields above $H_{sh} > H_C$ ³². In order to form such an Abrikosov fluxon a macroscopic part of the superconductor has to be driven normal conducting yielding as nucleation time³² $f_N^{-1} < 10^{-6}$ sec. Aside from Abrikosov fluxons, Josephson fluxons exist in weak links with very much reduced flux entry field $H_{C1J} \ll H_{sh}$ and nucleation time 10^{-11} sec $\ll f_N^{-1}$ because of spatial current distribution and because of the "insulating cores" of Josephson type fluxons.

3.5.1 Surface impedance of Nb

In contrast to ideal surfaces assumed in Ref. 32, real metals are oxidized, showing by crack corrosion weak links filled with oxides. Such weak links have actually been identified, e. g., for Nb penetrating from the surface 0.1-1 μm deep into the superconductor³³. There the density, the leakage current and the depth of weak links increase with defect concentration of Nb, i. e., with $1/RRR$ and strain of Nb. For reactor grade Nb with the resistance ratio $RRR \approx 30$, $j_c \geq 10^5$ A/cm², $H_{C1J} \approx 1$ mT and $f_N \approx 10^9$ Hz have been found^{31, 33}, after handling in air (≈ 1 h) at room temperature. This oxidation of Nb can be reduced by a protective layer of Al or NbN³³.

Also sputtered Nb and NbN contain weak links. Those weak links are related to the columnar sputter growth with insulating Nb₂O₅ coating the columns, i. e., the

grain boundaries³⁴. For example, sputtered Nb with $RRR \approx 3$ has grain sizes of $t_G \approx 10$ nm where the oxides at the grain surfaces yield $R_{bn} \approx 2 \cdot 10^{-12} \Omega\text{cm}^2$ as grain boundary resistance. Sputtered NbN usually has $RRR \leq 1$ due to grain sizes below $t_G \approx 5$ nm and $R_{bn} \geq 3 \cdot 10^{-12} \Omega\text{cm}^2$ ³⁴, where t_G is roughly equal to the inelastic mean free paths at 300 K.

Returning to surface impedances, for bulk Nb with $RRR \approx 30$, UHV annealed at 1800 °C and oxidized in air at room temperature for several hours a nucleation frequency of $f_N \leq 10^9$ Hz has been found³¹. There, for $f > \text{GHz}$ negligible rf field dependence of R_{res} has been found below 20 mT whereas for $f \approx 10^8$ Hz $R_{res}(H_{rf})$ increases linearly beginning at $H_{C1J} \geq 1$ mT up to 60 mT. This increase, like $R_{res}(H_{rf} \approx 0)$, is described by Eqs. (3.5) and (3.12)⁴ with $H \approx H_{rf}$ as weighted average. At 4.2 K:

$$R_{res}(H_{C1J}, 4.2 \text{ K}) \approx R_{res}(0, 4.2 \text{ K}) = \mu (\omega \mu_0)^2 \lambda_J^3(4.2 \text{ K}, H_{C1J}) \sigma_{bl}/2 \approx 10^{-7} \Omega (\text{fGHz})^2 \quad (3.13)$$

holds for $\mu \approx 0.1$ as areal weak link ratio, i. e., weak links distances of the order of $2\lambda_l$ with $\lambda_l \approx 40$ nm as penetration depth in Nb³³, $\lambda_J(H_{C1J}, 4.2 \text{ K}) \approx 0.3 \mu\text{m} \approx 8\lambda_l$ and $R_{bl} \approx 100 R_{bn}$ as leakage resistance for very good Nb-Nb₂O₅ junctions. Usually Nb-Nb₂O₅-Nb junctions have much larger R_{bn}/R_{bl} ratios due to defective Nb₂O₅ at the counter electrode³⁵. For bulk Nb only a few weak links exist described by $\mu \approx 0.01$ and thus the observed³¹ $R_{res}(0.1 \text{ GHz}) \approx 2 \text{ n}\Omega$ at 1.4 K compares quite well with the estimate in Eq. (3.13). Also the flux entry field H_{C1J} (Eq. (2.6)) and H_{rf} field dependencies of Eqs. (3.5) and (3.12) fit well to the observations³¹.

This bulk Nb with $\mu \approx 0.01$ has nearly no weak links and thus dc field mostly penetrate as Abrikosov fluxons causing large additional rf losses proportional³¹ to $H\sqrt{\omega}$ described by Eq. (3.11) by the viscous losses of oscillating Abrikosov fluxons^{22, 23}. In contrast, sputtered Nb with $RRR \approx 3-15$ does not show such a dc field dependence^{36, 37}. This is due to the fact, that $RRR \approx 3-15$ correspond to weak link distances between 10 and 50 nm, i. e., the weak link distances are smaller than λ_l . Thus dc fields -like rf fields- penetrate as Josephson type fluxons with their small viscous losses²² and large $U(T)$ in Eq. (3.4). In contrast to bulk Nb (Eq. (3.11)) this yields $R_{res}(H_{dc} \leq \text{mT}) \approx R_{res}(0)$, as observed^{36, 37}.

In rf fields, the penetrating Josephson fluxons enhance R_{res} by a different mechanism, namely by enhancing the Josephson penetration depth $\lambda_J(T, H)$ yielding Eq. (3.5), (3.12) and

$$dR_{res}/dH_{rf\text{max}} \approx R_{res}(H_{C1J}, T) \cdot 3 / \left(2 H_{C2J}(T) \cdot 4 \right) \alpha \omega^2 \lambda_J^3(T) / \left(1 - (T/T_{CJ}^*)^2 \right) \quad (3.14)$$

Here the factor 1/4 has been introduced to take into account the time -and space-wise averaging by using the maximal field H_{max} of the rf cavity. Comparing

Eq. (3.14) with Ref. 37, $H_{C2J}(4.2\text{ K}) \approx 10\text{ mT}$ and with Ref. 36 $T_{CJ}^* \approx 5.1\text{ K}$ is obtained. The latter value fits well to a NbO_x ($x \approx 0.02-0.04$) proximity layer at weak link banks, as found by surface impedance measurements of bulk Nb also³². Taking $T_{CJ}^* \approx 5.1-6\text{ K}$, $H_{C2J}(4.2\text{ K}) \approx 10\text{ mT}$ yields $H_{C2J}(\text{OK}) \approx 33-20\text{ mT}$ fitting to $H_{C1J}(0) \approx 0.2\text{ mT} \geq H_{C1H}(0)$ (Eq.(2.6)). Because of $T_{CJ}^* \approx 5.1-6\text{ K}$, above $T_{CJ}^*/2 \approx 2.5\text{ K}$ the $\lambda_J^3(T/T_{CJ}^*) \propto 1/\sqrt{j_{CJ}^3(T)}$ dependence in $R_{\text{res}}(T)$ cannot be neglected. This may explain the published high intrinsic Δ/kT_C -value^{36, 39} which assumed $R_{\text{res}}(T) \approx \text{const.}$ Below $T_{CJ}^*/2$ the exponential T dependence via $\sigma_{bl}(T)$ in $R_{\text{res}}(T)$ may yield the small weak link energy gap Δ_J , as found for YBCO⁶.

Sputtered Nb forming strip line resonators show field dependencies^{14, 38, 39} of the surface resistance as described in Eq. (3.5), (3.12) or (3.14) indicative for weak links. But strip line resonators have at 0.5 GHz $R_{\text{res}} \geq 10^{-7}\ \Omega$ due to radiation losses as compared to $R_{\text{res}} < 10^{-8}\ \Omega$ in closed resonators³⁶. Thus, aside from the ω , H and T dependencies in agreement with Eq. (3.14), nothing quantitative can be stated, yet.

The above results on Josephson fluxons penetrating along weak links fit qualitatively and quantitatively the observed rf residual losses^{4, 14, 32, 33, 36-39} of Nb in magnitude, field-, temperature- and frequency dependence. Also the observed weak increase of $dR_{\text{res}}/dH_{\text{rf}}$ with H_{dc} ^{31, 36, 37} is in line with the model-see Sect. 3.4.

3.5.2 Surface impedance of NbN

Detailed results on NbN cavity surfaces exist as sputter deposited layer, only. By columnar growth with $\text{Nb}_2\text{O}_5\text{-NbN}_{0.5}\text{O}_{0.5}$ coatings $\text{RRR} \leq 1$ and grain sizes around 3 to 5 nm are typical^{38, 39}. The highest RRR obtained is 2.6 being indicative for $R_{\text{bn}} \geq 3 \cdot 10^{-12}\ \Omega\text{cm}^2$ ⁴⁰. The grain boundaries are degraded by $\text{NbN}_{0.5}\text{O}_{0.5}$ formation³⁴ with $T_{CJ}^* \approx 10\text{ K}$ ⁴¹. This small T_{CJ}^* together with the small grain size $t_G \approx 4\text{ nm} \ll \lambda_I \approx 50\text{ nm}$ causes (Eq. (2.6)) very small flux entry fields $H_{C1H} \propto j_C t_G < 1\text{ Oe}$ ³⁹ and also H_{C2H} will be reduced. To obtain the residual losses the large penetration depth $\lambda_H \propto 1/\sqrt{j(T)} t_G$ (Eq. (2.7)) samples many weak links uniformly, which yields:

$$R_{\text{res}}(T, H) = (\omega \mu_0)^2 \lambda_H^3(T, H_{C1H}) 0.1 \sigma(T) \left(1 + \frac{3}{2} \frac{H}{H_{C2H}} \right) / 2 \quad (3.15)$$

Here as leakage current 0.1σ of the normal current has been assumed. With $\rho \approx 10^{-4}\ \Omega\text{cm}$ and $\lambda_H(4.2\text{ K}, H_{C1H}) \approx 0.7\ \mu\text{m}$ one obtains:

$$R_{\text{res}}(4.2 \text{ K}, H) \approx 10^{-7} \Omega \left(1 + \frac{3}{2} \frac{H}{H_{\text{C2H}}} \right) (f/\text{GHz})^2$$

which agrees with Refs. 39 in magnitude and with Ref. 38 and 39 in field and frequency dependence. Due to the small $T_{\text{CJ}}^* \approx 10 \text{ K}$ values $R(T)$ - fits assuming $R_{\text{res}} \approx \text{const}$, i. e. neglecting $R_{\text{res}} \propto \lambda_{\text{J}}^3 (T/T_{\text{CJ}}^*)$, yield too large Δ/kT_{C} values 6, 39. Below 4.2 K detailed fits 39 reveal $\sigma_{\text{bl}}(T) \propto \exp(-\Delta_{\text{J}}/kT)$ with $\Delta_{\text{J}} < \Delta_{\text{I}}/2$.

To improve sputtered NbN for rf applications t_{G} and R_{bl} have to be enhanced drastically. This is achievable 40 as shown by $\text{RRR} \approx 2.6$ with $\lambda_{\text{eff}} \approx 100 \text{ nm}$, being a first step in this direction.

4. CONCLUSION

Intrinsic, bulk properties can be obtained by surface impedance measurements only for surfaces not serrated by weak links. Weak links, with their reduced current carrying capacity and thus enhanced weak link penetration depth λ_{w} , destroy the surface rf currents ($\sim \lambda_{\text{I}}$) deep into the bulk. Thus the penetration depth λ_{eff} is amplified by $\mu \lambda_{\text{w}}$ (μ areal ratio) and R_{eff} correspondingly by the normal skin effect.

In the superconducting state $\lambda_{\text{w}} = \lambda_{\text{J}} > 10 \lambda_{\text{I}}$ and $\sigma_{\text{bl}}(T \rightarrow 0) \approx \sigma_{\text{bn}}$ (normal conducting boundary conductance) holds for YBCO. Thus large rf residual losses are encountered in YBCO, which can be decreased by reducing λ_{J} and σ_{bl} by improving the material quality as indicated by the smallest R_{res} for the smallest $\rho(0) \leq 0$. But still: currents passing through weak links in series are dominated by the detrimental weak links. This is in contrast to NMR or ESR where an "atomic averaging" yields more intrinsic information.

The large Josephson penetration depth $\lambda_{\text{J}} > 10 \lambda_{\text{I}}$ yields a very small field $H_{\text{C1J}} \propto 1/\lambda_{\text{J}}^2$ where fluxons start to enter weak links. This yields strong field dependencies and nonlinearities at rather small fields. Only YBCO with $d\rho/dT \approx 0.5 \mu\Omega\text{cm/K}$ and $\rho_{100} \approx 50 \mu\Omega\text{cm/K}$, i. e., $\rho(0) < 0$, may show $H_{\text{C1G}} > 100 \text{ Oe}$ and small R_{res} 14, 42. For Nb or NbN weak links with their strongly degraded T_{CJ} - and Δ_{J} - values are detrimental for rf cavities, aside from their large density shown by $\text{RRR} < 10$. Also there smaller $\rho(0)$ -values, i. e., larger RRR values, yield better rf cavities.

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