

## Residual surface resistance of polycrystalline superconductors

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By modeling a superconducting polycrystalline film as a network of superconducting grains coupled via Josephson junctions, various authors have shown that rf losses at the grain boundaries can be the main source of the residual surface resistance in high- $T_c$  superconductors. The same model is extended here to include the effect of dc or rf applied fields and is then shown to be consistent with previous results on the surface impedance of low- $T_c$  superconducting polycrystalline films. The model also provides a satisfactory, though qualitative, description of recent results on the quality factor of niobium thin-film-coated rf cavities for particle accelerators.

### INTRODUCTION

Rf residual losses are known to be one of the main factors limiting the performances of superconducting cavities for particle accelerators,<sup>1</sup> and a clear understanding of the causes of such losses is therefore essential for further progress in this field.

The possible origins of residual terms in the surface impedance of superconductors have been widely discussed in the literature,<sup>2</sup> and include dissipation due to frozen-in magnetic flux, normal inclusions, phonon generation effects, and many others, but it is often difficult to identify the real mechanism operating in a specific situation.

In this Brief Report, following the simple model proposed by Hylton *et al.*<sup>3,4</sup> and, in a slightly different form, by Vendik *et al.*<sup>5</sup> for high- $T_c$  polycrystalline superconductors, we will show that losses at the grain boundaries can be the main source of the residual resistivity in both high- $T_c$  and low- $T_c$  polycrystalline superconducting films. Our results are indeed shown to be consistent with a model previously introduced to describe the surface losses in low- $T_c$  polycrystalline films<sup>6</sup> and to provide a satisfactory, though qualitative, description of recent results on niobium thin-film-coated rf cavities for particle accelerators,<sup>7-9</sup> including the effect of the rf field amplitude on the quality factor.

### THEORY

The surface impedance of a metal in the normal state and at low frequencies can be written as

$$Z_n = R_n + jX_n = \left( \frac{\mu_0 \omega}{2\sigma_n} \right)^{1/2} (1 + j), \quad (1)$$

where  $\sigma_n$  is the normal-state dc conductivity.

In the local dirty limit (short mean free path), we can obtain the theoretical surface impedance of a superconductor ( $Z_{\text{BCS}}$ ) by replacing  $\sigma_n$  in (1) with the BCS complex conductivity<sup>10</sup>  $\sigma_1 + j\sigma_2$ .

For  $T \leq \frac{1}{2}T_c$  and at low frequencies ( $\omega \ll 2\Delta/\hbar$ ,  $\Delta$  is the energy gap) it is  $\sigma_2 \gg \sigma_1$  and we get<sup>10</sup>

$$R_{\text{BCS}} = \frac{1}{2\lambda_1} \frac{\sigma_1}{\sigma_2^2}, \quad (2a)$$

$$X_{\text{BCS}} = \mu_0 \omega \lambda_1, \quad (2b)$$

where  $\lambda_1 = (\mu_0 \omega \sigma_2)^{-1/2}$  is the magnetic field penetration depth.

Since, in the limits considered, it is  $\sigma_1 \approx e^{-\Delta/kT}$  and  $\sigma_2 \approx 1/\omega$ , expression (2a) predicts that  $R_{\text{BCS}} \rightarrow 0$  for  $T \rightarrow 0$  with an exponential temperature dependence and  $R_{\text{BCS}} \approx \omega^2$  at low frequencies.

The experimental surface resistance  $R_s$  of superconductors is in general well described by the expression<sup>1</sup>

$$R_s = R_{\text{BCS}} + R_{\text{res}}, \quad (3)$$

where  $R_{\text{res}}$ , that describes the residual losses, is generally temperature independent and can exhibit different frequency dependences.<sup>2</sup>

For high- $T_c$  superconductors, due to the short coherence length, the local limit still applies, and Eqs. (2) and (3) are still valid, though an explicit expression for  $\sigma_1$  and  $\sigma_2$  is unknown.

If we are dealing with a polycrystalline superconductor it is clearly important to consider the contribution of the grain boundaries to the overall losses. One way is to describe the film as a network of superconducting grains coupled via a Josephson junction.<sup>3-5</sup> The basic element of such a network and the equivalent circuit for the admittances are represented in Fig. 1.

The element  $J$  is described by the Josephson current-phase relation:

$$I(t) = I_c \sin \left[ \frac{2e}{\hbar} \int_0^t V(t') dt' \right]. \quad (4)$$

The shunt resistance of the junction  $R_j$  is assumed to be constant.

For small rf signal amplitudes ( $I_{\text{rf}} \ll I_c$ ) the Josephson element can be well approximated by an inductance of value:<sup>11</sup>

$$L_j = \hbar/2eI_c F(I_{\text{dc}}), \quad (5a)$$

where

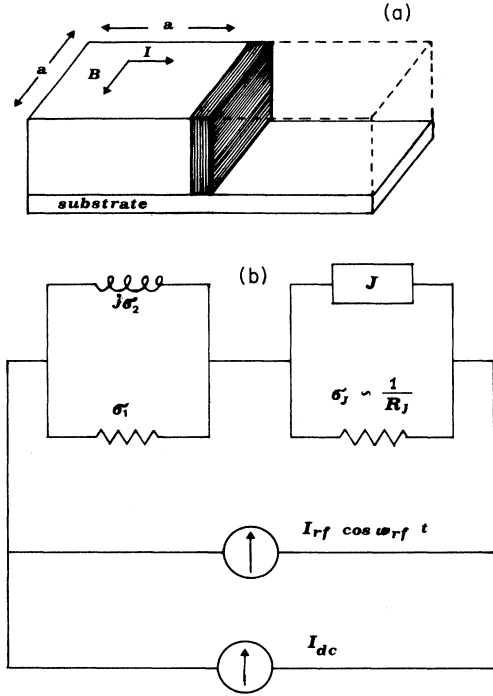


FIG. 1. (a) Basic element of the network of grains coupled by Josephson junctions used to describe a polycrystalline superconducting film. (b) The equivalent circuit for the same network.

$$F(I_{dc}) = [1 - (I_{dc}/I_c)^2]^{1/2}. \quad (5b)$$

If we further assume that the junction conductivity is dominated by the Josephson element ( $\hbar\omega/2eI_cR_jF \ll 1$ ), we are led to an effective conductivity  $\sigma_{j\text{eff}}$  of the grain boundary that, averaged over the grain dimension, can be written as<sup>3</sup>

$$\sigma_{j\text{eff}} = \{\mu_0\omega\lambda_j^2F^{-1}[(\hbar\omega/2eI_cR_jF) + j]\}^{-1}, \quad (6)$$

where  $\lambda_j = (\hbar/a2eJ_c\mu_0)^{1/2}$ ,  $a$  is the average grain dimension, and  $J_c$  is the critical Josephson current density.

The surface impedance  $Z_{sj}$  of the polycrystalline film is then obtained by replacing  $\sigma_n$  in Eq. (1) with the overall conductivity of the basic network element:

$$\sigma_{\text{eff}} = [\sigma_{j\text{eff}}^{-1} + (\sigma_1 + j\sigma_2)^{-1}]^{-1}. \quad (7)$$

In the same approximations made above we get

$$R_{sj} = \frac{1}{2\lambda_{\text{eff}}} \frac{\sigma_1 + \sigma_r}{\sigma_2^2}, \quad (8a)$$

$$X_{sj} = \mu_0\omega\lambda_{\text{eff}}, \quad (8b)$$

where we have introduced

$$\lambda_{\text{eff}} = (\lambda_1^2 + \lambda_j^2F^{-1})^{1/2}, \quad (9a)$$

$$\sigma_r = \mu_0\omega\lambda_j^2F^{-1}(\hbar\omega/2eI_cR_jF)\sigma_2^2. \quad (9b)$$

Assuming  $I_{dc} \ll I_c$  ( $F=1$ ) and neglecting  $\sigma_1$  in Eqs. (8), we exactly reproduce the results of Refs. 3 and 4. If we

further assume  $\lambda_j \gg \lambda_1$  the results of Ref. 5, in the low-frequency limit, are also reproduced.

We can now rewrite  $\sigma_r$  in a more convenient form remembering that, at  $T \leq \frac{1}{2}T_c$  and at low frequencies, it is  $\sigma_2/\sigma_n = \pi\Delta/\hbar\omega$ :<sup>10</sup>

$$\sigma_r/\sigma_n = \left[\frac{\lambda_j}{\lambda_1}\right]^2 \frac{\pi\Delta}{2eI_cR_j} F^{-2}, \quad (10)$$

where Eq. (5b) gives the explicit form for  $F(I_{dc})$ . In this way it is clear how, in the limits considered,  $\sigma_r$  is frequency and temperature independent (as long as we assume  $J_c$  and  $I_cR_j$  to be temperature independent as well).

In the more general case, i.e., removing the approximation  $I_{rf} \ll I_c$ , the Josephson element cannot be simply replaced by an inductance, but we can obtain  $\sigma_{j\text{eff}}$ , and then  $R_{sj}$ , for any value of  $I_{rf}$  by solving the circuit in Fig. 1(b) using the nonlinear equation (4).<sup>11</sup> However at low frequency (compared with the Josephson characteristic frequencies) the effect of a dc or an rf bias current is the same, as we have also checked numerically, so that, in this limit, we can just replace (or add)  $I_{rf}$  to  $I_{dc}$  in  $F$ . Finally for  $I_{dc}$  ( $I_{rf}$ )  $> I_c$  the Josephson junction is no more in the zero voltage state and much higher losses are present.<sup>11</sup>

## DISCUSSION

Equation (8a) predicts the presence of a "residual" (temperature independent for  $T \rightarrow 0$ ) component of the surface resistance for a polycrystalline superconductor, and this is essentially equivalent to add a residual term to the BCS surface impedance as in Eq. (3). The ratio  $\sigma_r/\sigma_1$  gives the relative weight of the residual term to the "BCS" term in the overall surface impedance.

Since  $\sigma_r$  is frequency independent and  $\sigma_2 \sim 1/\omega$ , the residual surface resistance in our model shows a quadratic frequency dependence as observed in high- $T_c$  (Ref. 12) and, in many cases, in low- $T_c$  superconductors.<sup>2</sup>

As clear from Eqs. (9) and (10), the residual term  $\sigma_r$  is large for small critical current densities ( $J_c$ ), small grain sizes ( $a$ ), or small  $I_cR_j$  products (related to the junction quality). As assumed in Refs. 3–5, for high- $T_c$  polycrystalline superconductors it is generally  $\sigma_r \gg \sigma_1$ , and in this limit, as discussed in the same references, the model introduced in the previous section, using reasonable parameters, describes very well the experimental results on the surface impedance of high- $T_c$  samples, including its temperature and frequency dependence.

As shown in the previous section,  $\sigma_r$  also depends on  $I_{dc}$  ( $I_{rf}$ ) [Eq. (10)] and increases as a function of an applied dc or rf field. This can account for the field amplitude dependence of the surface resistance observed in sintered pellets and polycrystalline films of the high- $T_c$  compounds<sup>12</sup> whereas single crystals and epitaxial films do not exhibit such dependence<sup>13</sup> (since, in this case,  $\sigma_r < \sigma_1$ ). An accurate fit of the experimental data would require the introduction of the statistical distribution of the junction parameters. Moreover the introduction of a capacitive term at the grain boundaries would improve

the model, accounting for the small hysteretic effects observed in some cases. However these tasks go beyond the scopes of this Brief Report.

It is now very relevant to observe that expression (8a) can be directly obtained starting from the BCS result [Eq. (2a)] by replacing the “intragrain” penetration depth  $\lambda_1$  with the effective magnetic field penetration depth  $\lambda_{\text{eff}}$ , and by adding a temperature and frequency independent contribution  $\sigma_r$  to the real part of the BCS conductivity  $\sigma_1$ . This is essentially what Broom and Wolf,<sup>6</sup> and others,<sup>14</sup> proposed to fit the data on the surface impedance of various polycrystalline films, obtained by Josephson effect measurements. Our model is consistent with those measurements and gives an explicit expression for  $\sigma_r$  [Eq. (10)].

We can finally prove that, also for low- $T_c$  polycrystalline materials, the grain boundaries may strongly influence the rf losses in the superconducting state even if

they have a negligible effect on the transport properties in the normal state.<sup>4</sup> As an example we can consider a “model” polycrystalline Nb film with the following parameters:

$$T_c = 9.2 \text{ K}, \quad \rho_n = 2\mu\Omega \text{ cm}, \quad a = 0.2 \mu\text{m},$$

$$I_c R_j = 2 \text{ mV}, \quad J_c(0) = 2.5 \times 10^7 \text{ A/cm}^2,$$

$\rho_n = 1/\sigma_n$  is the intragrain resistivity,  $a$  is the average grain size,  $I_c$  is the critical current of the Josephson junctions at the grain boundaries,  $R_j$  is the corresponding resistance, and  $J_c$  is the junctions maximum current density.

The values for  $T_c$ ,  $\rho_n$ , and  $a$  are typical of a niobium film obtained by standard sputtering techniques.  $I_c R_j$  has been chosen to be close to the ideal tunneling value  $\pi\Delta/2e$  ( $\Delta \approx 1.4 \text{ meV}$ ) and  $J_c$  has been set to a value only about twice less than the bulk critical current density.

The overall low-temperature dc resistivity of the film, including the contribution of the grain boundaries is<sup>4</sup>

$$\rho_0 = \rho_n + I_c R_j / J_c a. \quad (11)$$

In our case we get  $\rho_0 = 6\mu\Omega \text{ cm}$ , implying a residual resistivity ratio of 3.5. At  $T = 4.2 \text{ K}$ , with the same Nb parameters, at low frequencies and  $I_{\text{dc}}(I_{\text{rf}}) \ll I_c$ , we obtain [assuming  $J_c(4.2 \text{ K}) \approx J_c(0)$ ]

$$\lambda_1 = 50 \text{ nm}, \quad \lambda_j = 72 \text{ nm},$$

and, from Eqs. (9a) and (10),

$$\lambda_{\text{eff}} = 88 \text{ nm}, \quad \sigma_r / \sigma_n = 2.1.$$

(This value for the effective field penetration  $\lambda_{\text{eff}}$  is again appropriate for a sputtered Nb polycrystalline film.<sup>6</sup>)

If we set now, as an example,  $\omega/2\pi = 500 \text{ MHz}$ , we have, from the BCS theory (in the dirty limit) at  $T = 4.2 \text{ K}$ ,

$$\sigma_1 / \sigma_n = 1.04, \quad \sigma_2 / \sigma_n = 1960,$$

and we clearly see how, according to the present model, a niobium film with reasonable dc properties can have, due to the grain boundary losses, a quite large residual surface resistance ( $\sigma_r \gtrsim \sigma_1$  at 4.2 K and 500 MHz).

Inserting these data in Eq. (8a) we obtain, for our Nb film at 500 MHz,

$$R_{sj}(4.2 \text{ K}) = 90 \text{ n}\Omega.$$

At  $T = 1.2 \text{ K}$  we have  $\sigma_1 / \sigma_n \approx 0$  and

$$R_{sj}(1.2 \text{ K}) = 57 \text{ n}\Omega.$$

These values compare well with experimental data recently obtained at the same temperatures and frequency on Nb film sputter-coated rf cavities for particle accelerators.<sup>7-9</sup>

Equations (8a) and (10) also predict an increase of  $R_{sj}$  with  $I_{\text{dc}}(I_{\text{rf}})$ . Here, due to the rather strong grain coupling, a truly Josephson model may be inadequate so that the explicit form of the function  $F$  [Eq. (5b)] has to be taken as a qualitative indication. In Fig. 2(a) the dependence of the quality factor  $Q = \Gamma/R_s$  on  $I_{\text{dc}}(I_{\text{rf}})$  is re-

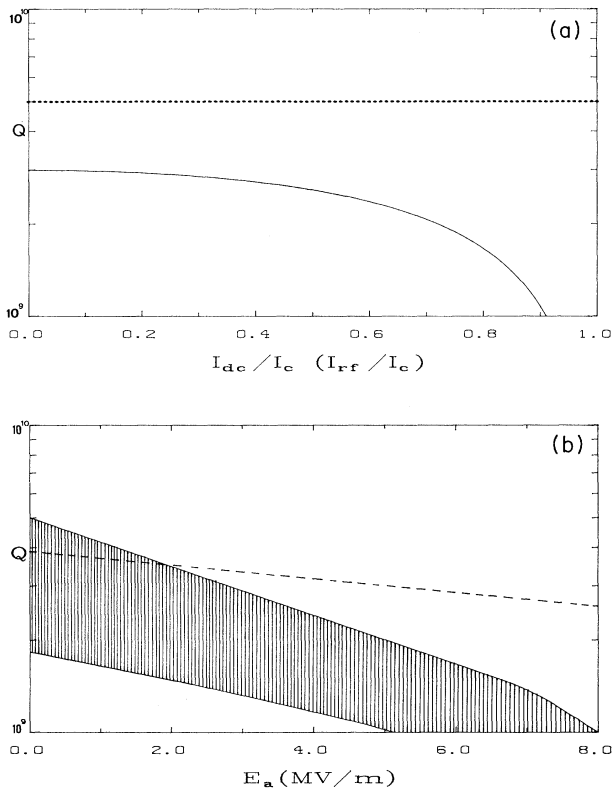


FIG. 2. (a) Quality factor ( $Q = \Gamma/R_s$ ) as a function of the applied dc (rf) current at  $T = 4.2 \text{ K}$  and  $\omega/2\pi = 500 \text{ MHz}$  for a hypothetical rf cavity sputter coated with a “model” Nb film (see text). The dashed line represents the predicted BCS result for the cavity, at the same temperature and frequency, without considering the effect of the grain boundaries. (b) Quality factor  $Q$  as a function of the maximum accelerating field  $E_a$  as experimentally observed in a set of polycrystalline Nb sputter-coated cavities at  $T = 4.2 \text{ K}$  and  $\omega/2\pi = 500 \text{ MHz}$  (hatched region) and for a bulk niobium cavity (dashed curve). The data are from Refs. 8 and 9.  $E_a \sim I_{\text{rf}}$ ; the relative amplitude of the horizontal scales in (a) and (b) has been set arbitrarily.

ported for a hypothetic rf cavity at  $T=4.2$  K and 500 MHz, sputter coated with our "model" Nb film, assuming  $\Gamma=270 \Omega$  as in Refs. 7–9. The quality factor  $Q$  for a niobium cavity with the same parameters and no grain boundaries ( $R_s=R_{BCS}$ ) is also reported in the figure for comparison.

The dependence of the quality factor  $Q$  as a function of the maximum accelerating field  $E_a$ , as experimentally observed in a large set of polycrystalline Nb sputter-coated cavities at the same temperature and frequency, is schematically reported in Fig. 2(b), hatched region (data from Refs. 8 and 9;  $E_a \sim B_{rf} \sim I_{rf}$ ).

The slope in the  $Q(E_a)$  curve, systematically observed in the experiments, is mainly attributable to the residual component of  $R_s$ , as verified by low-temperature measurements,<sup>15</sup> and it is much less pronounced in very high-quality films<sup>15</sup> (large grain sizes and intergrain critical currents) and in bulk niobium cavities [see, as an example, the dashed curve in Fig. 2(b), data from Refs. 8

and 9]. The qualitative agreement between the data and the results of our model strongly supports the idea that losses at the grain boundaries can be the main source of the residual resistivity in Nb polycrystalline superconducting film-coated cavities.

Finally, in the case of bulk Nb cavities, the fact that the observed losses must have a different physical origin than for polycrystalline thin film cavities, though the order of magnitude can be comparable, is also indicated by the different behavior exhibited when cooled down in the presence of a dc magnetic field.<sup>9</sup>

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