Optimum multilayer coating of superconducting particle accelerator cavities and effects of thickness dependent material properties of thin films

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The maximum amplitude of the surface RF magnetic field \( B_0 \) is one of the key parameters of superconducting RF (SRF) cavities for particle accelerators.\(^1\)\(^2\) \( B_0 \) is proportional to the electric field on the cavity axis and its improvement leads to a reduction of the accelerator length necessary for getting a target energy. In the last several decades,\(^2\) SRF researchers have continuously pushed up the record value of \( B_0 \). The state-of-the-art Nb cavities can reach \( B_0 \sim 200 \text{ mT} \)\(^5\) which corresponds to the accelerating electric field \( \sim 50 \text{ MV m}^{-1} \) for Tesla shape cavities.\(^6\) However, further improvements are thought difficult as long as the present bulk Nb technology is used. This is because the corresponding screening current density at the surface \( J_0 \propto B_0 \) is already close to the deparing current density \( J_d \propto B_{sh} \propto B_0 \) at which the Meissner state becomes absolutely stable and vortex dissipation necessarily leads to strong \( Q \) degradations or quenches. Here \( B_{sh} \) and \( B_0 \) are the superheating field\(^7\)\(^8\)\(^9\)\(^10\) and the thermodynamic critical field, respectively.

Thus, to develop technologies beyond the bulk Nb cavity is of importance for even more improvement of the SRF cavity performance. Using an s-wave superconductor with a higher \( B_0 \) pushes up the theoretical field limit (e.g., \( B_{sh} \sim 400 \text{ mT} \) for \( \text{Nb}_3\text{Sn} \)\(^4\)\(^5\)\(^10\)) but cavities made from such alternative materials have not yet broken the record field of Nb cavity.\(^11\) This is probably because such materials are prone to have small lower critical fields \( B_{cl} \), resulting in the dissipative penetration of vortices at \( B_{cl} < B_0 < B_{sh} \) where the Meissner state is metastable and instead the vortex state is the stable state.

The multilayer structure\(^12\) shown in Fig. 1 has attracted much attention as it may solve this problem. The idea is to coat superconducting substrate (\( \Sigma \)) with a thin superconductor layer (\( S \)) separated by an insulator layer (\( I \)) to avoid catastrophic vortex dissipation and to achieve fields as high as \( B_{sh} (> B_{cl}) \). Then it has been recognized that there are appropriate material combinations and layers thicknesses:\(^13\) the penetration depth of the \( S \) layer \( \lambda_S \) must be larger than the sum of the insulator thickness \( d_I \) and the penetration depth of the substrate \( \lambda_S \). Then the screening current density in the \( S \) layer is suppressed by the counterflow generated by the substrate. This current suppression effect is pronounced as the thickness \( d \) decreases. On the other hand, to protect the substrate, a thickness \( d \sim \lambda \) is necessary. This observation\(^13\)

results in existence of the optimum thickness \( d_m \).

Theoretical calculations have shown that the field limit of multilayer structure \( B_{sh} \) can exceed the intrinsic \( B_{sh} \) when \( d \sim d_m \).\(^13\)\(^14\)\(^15\)\(^16\)

Today, various experiments for demonstrating the multilayer superiorities at \( d \sim d_m \) are ongoing (e.g., the third harmonic measurement of first flux penetration field,\(^17\)\(^18\)\(^19\) RF measurement by using the quadrupole resonator,\(^20\)\(^21\) mushroom-shaped cavities,\(^21\)\(^22\)\(^23\) etc). However, the theoretical calculations carried out so far had aimed for understanding of the general properties of multilayer structure and not for providing with predictions for a specific experiment. To extract predictions from the theory and compare them with experiments, the material parameters of one’s own films should be used. This is because superconducting properties of thin films are sensitive to the growth conditions, resulting in different \( J_d \), \( \lambda \), and then \( B_{sh} \) and \( d_m \).

In addition, it is known that superconducting properties of a thin film generally depends on its thicknesses below \( d \sim 100 \text{ nm} \).\(^24\) However, any theoretical calculation of \( B_{ML} \) has not yet taken into account the \( d \) dependences of superconducting properties of thin films. This effect also shifts \( B_{sh} \) and the optimum parameters, and should be taken into account for a comparison between the theory and experiments.

We revisit the field limit of a superconductor–insulator–superconductor multilayer structure for particle accelerator cavities (\( B_{sh} \)), taking into account thickness (\( d \))–dependent material properties of thin films. Resultant \( d \)-dependent thermodynamic critical field and penetration depth lead to the appearance of a peak in \( B_{sh}(d) \) which has been missed in the previous studies. The procedure shown in this note would be useful to evaluate \( B_{sh} \) based on properties of one’s own films.

Fig. 1. (Color online) The multilayer structure. A single superconducting layer \( S \) and a single insulator layer \( I \) are formed on a superconducting substrate \( \Sigma \). The parameters are summarized in Table I.
In this brief note, (1) we show an example of the way to obtain theoretical predictions by using real material parameters and (2) examine how $d$ dependences of superconducting properties affect the field limit and optimum parameters.

Let us consider a model shown in Fig. 1. We assume $T < T_c$, because the operating temperature of SRF cavities $T \lesssim 4 \, \text{K}$ is well below $T_c$ of $S$ layer material candidates (e.g., NbSn, MgB$_2$, NbN, NbTiN, etc). Since the nonlinear Meissner effect which makes $\lambda$ dependent on the screening current density is weak for $s$-wave superconductors at $T < T_c$, we use the London equation to calculate the field and current distributions. Then the screening current density at the surface of $S$ layer $J_d$ and the magnetic field at the $S$-$\Sigma$ interface $B_{\text{fl}}$ are given by

$$J_d = \gamma_1 \frac{B_0}{\mu_0 \lambda},$$

$$B_{\text{fl}} = \gamma_2 B_0,$$

where

$$\gamma_1 = \frac{\sinh \frac{d}{\lambda} + \frac{\lambda_0 + d_1}{\lambda} \cosh \frac{d}{\lambda}}{\cosh \frac{d}{\lambda} + \frac{\lambda_0 + d_1}{\lambda} \sinh \frac{d}{\lambda}},$$

$$\gamma_2 = \frac{1}{\cosh \frac{d}{\lambda} + \frac{\lambda_0 + d_1}{\lambda} \sinh \frac{d}{\lambda}}.$$

The Meissner state of the $S$ layer becomes absolutely unstable when $J_d$ reaches the depairing current density $J_B$. Then we find $B_0 = \mu_0 J_B / \gamma_1$ is the maximum field that the $S$ layer can withstand. On the other hand, breakdowns of the substrate are triggered when $B_{\text{fl}}$ reaches a threshold value $B_{\text{fl}}^{\text{th}}$, resulting in another limitation $B_0 = B_{\text{fl}}^{\text{th}}/\gamma_2$. Then the field limit of the multilayer structure is given by

$$B_{\text{ML}} = \min(\mu_0 \lambda J_d / \gamma_1, B_0 / \gamma_2).$$

Fortunately, we know $\mu_0 \lambda J_d$ at $T < T_c$: it corresponds with the superheating field for a semi-infinite superconductor at $T < T_c$ and is given by 0.84$B_c$ for a clean limit superconductor with the coherence length $\ll \lambda$.

$$\mu_0 \lambda J_d \simeq 0.84 B_c.$$  

Note here the parameters $\lambda$ and $J_d$ (or $B_c$) are affected by material properties of the $S$ layer through the BCS relations

$$\lambda = \sqrt{\frac{\hbar}{\pi \mu_0 \alpha k_B T_c}} = \frac{\rho}{\rho_{\text{ref}}} \frac{T_{\text{c,ref}}}{T_c} \lambda_{\text{ref}},$$

$$B_c = \sqrt{\frac{\mu_0 N}{\alpha k_B T_c}} = \sqrt{\frac{N}{N_{\text{ref}}} \frac{T_{c,\text{ref}}}{T_{\text{c,ref}}}} B_{c,\text{ref}}.$$  

Here $\rho$ is a normal resistivity, $N$ is a normal density of states, and $\alpha = \Delta/k_B T_c$. The parameters with the subscript ref represent some reference values.

For a concrete discussion on the effects of $d$-dependent material properties and for demonstrating how to use experimental data of real samples to extract theoretical predictions, we consider NbN–I–Nb multilayer structures in the following. The substrate is assumed a bulk cavity-grade Nb with $\lambda_0 = 40$ nm and $B_c = 180$ mT. For the NbN layer, we use the data reported by Semenov et al. as an example. Here we do not consider the origin of $d$-dependences of material properties and simply assume the superconducting properties of NbN films are well described by the BCS theory. Shown in Figs. 2(a), 2(b) are the $d$ dependences of $T_c$, normal resistivity $\rho$, and normal density of states $N$. There are a number of empirical fitting functions and models which explain the dependence of $T_c$ on $d$ (see, e.g., Ref. 28). Here we use the empirical fits$^{26,29}$ $T_c(d) = T_{c,\infty} \tanh(d/d_1)$, $\rho(d) = \rho_{\infty}(1 + d_2/d)$, and $N(d) = N_{\infty} \tanh(d/d_3)$, where the fitting parameters are given by $T_{c,\infty} = 14.9\, \text{K}$, $d_1 = 3.77 \, \text{nm}$, $\rho_{\infty} = 0.619 \, \rho_0 \, \Omega \, \text{m}$, $d_2 = 11.3 \, \text{nm}$, $N_{\infty} = 1.67 \times 10^{12} \, \text{cm}^{-3}$, and $d_3 = 5.29$ nm. Then we can evaluate $\lambda$ using Eq. (7) and $\Delta = 1.95k_B T_{c,\infty}$ $B_c$ can be calculated using Eq. (8) and $B_{c,\text{ref}} = 234$ mT at $T_{c,\text{ref}} = 15.5\, \text{K}$ and assuming $N_{\text{ref}} = N_{\infty}$. Shown in Fig. 2(c) are the $d$ dependences of $\lambda$ and $B_c$. As $d$ increases, $\lambda$ decreases down to $\approx 200$ nm. This comes from the increase of $T_c$ and the decrease of $\rho$. The value of $\lambda$ is consistent with previous studies. The increase of $B_c$ comes from the fact that both $T_c$ and $N$ increase with $d$.

Now we have all the parameters necessary to calculate $B_{\text{ML}}$. Shown in Fig. 3(a) is the contour map of $B_{\text{ML}}$ for different $d$ and $d_f$ calculated from Eq. (5). The maximum value $B_{\text{ML}} \approx 260$ mT is located at $d \sim 150$ nm and $d_f \lesssim 50$ nm. The contours extending to thick $d_f$ regions (upper left of the contour map) are not seen in the previous studies in which $\lambda$ and $B_c$ were assumed to be independent of $d$.

The solid curve shown in Fig. 3(b) is the cross Sect. of the contour map at $d_f = 280$ nm, different from the dashed curve calculated under the assumption of the thickness-independent $\lambda = \lambda(\infty)$ and $B_c = B_c(\infty)$. The peak at $d \approx 10$ nm comes from the following mechanism: (1) At $d \gtrsim 20$ nm, we have $\lambda \approx 200$ nm, which satisfies $\lambda < \lambda_{\text{ref}} + d_f$, at which the screening current density on the $S$ layer increases as $d$ decreases. Thus, the field limit decreases with $d$. (2) However, at $d \lesssim 20$ nm, $\lambda$ becomes to satisfy $\lambda > \lambda_{\text{ref}} + d_f$, at which the counterflow-induced current suppression effect is pronounced as $d$ decreases, enhancing the field limit. (3) For even smaller $d$ regions, the suppression of $B_c$ overwhelms the counterflow-induced current suppression effect and the field limit decreases with $d$.

The solid curve shown in Fig. 3(c) is the cross section at $d_f = 30$ nm, slightly shifted from the dashed curve calculated under the assumption of the thickness-independent $\lambda$ and $B_c$. At such thin $d_f$ regions in which $\lambda > \lambda_{\text{ref}} + d_f$, the counterflow-induced current suppression effect is always pronounced as $d$ decreases in contrast to thick $d_f$ cases seen in the above, and the field limit of the $S$ layer is enhanced as $d$ decreases. For $d \lesssim \lambda$, however, the $S$ layer is too thin to protect the substrate, resulting in breakdowns at the substrate.

**Table I. Summary of the parameters.**

| Bulk conductor $B_{\text{fl}}$ | Thickness, $d$: | London depth, $\lambda$: |
|-------------------------------|-----------------|------------------|
| Superconducting substrate $B_c$ | Empirical field limit, $B_{c,\text{ref}}$: |
| Empirical data of real samples | $\lambda_{\text{ref}}$: | 
| $\rho_{\infty}$: | $N_{\text{ref}}$: | 
| $d_1$: | $d_2$: | $d_3$: | $T_{c,\text{ref}}$: | $B_{c,\text{ref}}$: |
The optimum thickness is given by \(d = d_m\) at which the \(S\)-layer-limited \(B_{ML} = \mu_0 \lambda J_{d}\) equals the substrate-limited \(B_{ML} = B_{2f}/2\). If the condition \(\lambda > \lambda_S + d_I\) is satisfied and \(\lambda(d)\) and \(B_c(d)\) rapidly converge to constants at \(d \ll \lambda(\infty)\), the following analytical formula to find \(d_m\) is still useful (see Ref. 14 for \(d_I \ll \lambda_S\) and Ref. 16 for a finite \(d_I\)):

\[
ds_m = \frac{\lambda(\infty)}{\lambda(\infty) + \lambda_S + d_I} \left[ \frac{0.84 B_c(\infty)}{B_{2f}} \right]
\]

\[
+ \left( \frac{\lambda(\infty)}{\lambda(\infty) + \lambda_S + d_I} \right)^2 \frac{0.84 B_c(\infty)}{B_{2f}} \left[ \frac{\lambda(\infty) - \lambda_S - d_I}{\lambda(\infty) + \lambda_S + d_I} \right].
\]

This yields the optimum \(d\) of the dashed curve in Fig. 3(c), providing with a good approximation of the true \(d_m\).

For a more realistic evaluation of \(B_{ML}\) and the optimum thicknesses, we need to take into account the existence of defects on the surface; e.g., non-stoichiometric composition or impurities can locally suppress the depairing current density,32) topographic defects locally enhance the screening current density,33,34) etc. These effects can be expressed by replacing14,16,33)

\[
J_0 \to \eta J_0.
\]

Here \(\eta(0 < \eta \leq 1)\) is a phenomenological suppression factor. Shown in Fig. 3(d) is the effects of defects on the surface of NbN layer. The value of \(B_{ML}\) continuously changes from \(\eta = 1\) (ideal surface) to \(\eta = 0.9\). As \(\eta\) decreases, the optimum shifts to a smaller \(d\) and the maximum \(B_{ML}\) decreases.

Application of the above procedure to other materials data is straightforward. For NbTiN–I–Nb multilayer,35) we obtain \(B_{ML} \approx 220\) mT at \(d \approx 100\) nm and \(d_I \lesssim 30\) nm. Here we used \(\Delta = 1.86 k_B T_c\),36) the data of Ref. 37 for \(T_c(d)\) and \(\rho(d)\), the fitting function in Ref. 24, and the assumption of \(d\)-independent \(N = 1.17 \times 10^{27}\) m\(^{-1}\) for simplicity. It should be noted that the result is sensitive to material

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**Fig. 2.** (Color online) The thickness dependences of (a) \(T_c\), \(\rho\), and (b) \(N\) reported in Ref. 25. (c) \(\lambda\) and \(B_c\) calculated from Eqs. (7) and (8).

**Fig. 3.** (Color online) Achievable field by the NbN–I–Nb multilayer calculated by using the data shown in Fig. 2 and Eq. (5). (a) Contour map; (b), (c) cross section of the contour map at \(d_I = 280\) nm and 30 nm; (d) effects of defects at \(\eta = 0.9–1.0\).
parameters, then a quantitative evaluation of $B_{\text{ML}}$ needs material parameters of one’s own films. Another example, Nb–I–Nb multilayer, reaches $B_{\text{ML}} \approx 220$ mT at $d \approx 30$ nm and $d_f \lesssim 30$ nm. Here we used $\Delta = 1.9 k_B T_c$, the data of Ref. 29 for $T_c(d)$ and $\rho(d)$, the fitting function $T_c(d) = T_{c0}^d(1 - d^\alpha/d^*)$ (here $T_{c0}^d$ and $d^*$ are fitting parameters), and the assumption of $d$-independent $N = 5 \times 10^{12} \text{J}^{-1} \text{m}^{-3}$ for simplicity. In both the cases, we have the peak like Fig. 3(b) originating from the thickness dependent $\lambda$ and $B_c$.

We used the real experimental data that exhibits the thickness dependent $T_c$, $\rho$, and $N$ to evaluate $\lambda$ and $B_c$ (Fig. 2), and calculated $B_{\text{ML}}$ and the optimum thicknesses (Fig. 3). We found the effects of the thickness dependences are pronounced at thicker $d_f$ regions. When $d_f$ is large enough ($\lambda < \lambda_c + d_f$), we have a peak in $B_{\text{ML}}(d)$ which has been missed in the previous studies, shown in Fig. 3(b). Assuming constant $\lambda$ and $B_c$ is still valid at thin $d_f$ regions if $\lambda > \lambda_c + d_f$ is satisfied and $\lambda(d)$ and $B_c(d)$ rapidly converge at $d \ll \lambda(\infty)$. Yet, since material properties of a thin film are generally sensitive to growth conditions, those of one’s own films should be used to extract theoretical predictions. The procedure shown in this note might be useful to obtain $\lambda$ and $B_c$, and then $B_{\text{ML}}$, particularly when $\lambda$ and $B_c$ of one’s own films are not available.

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