

Analysis of Magnetic Vortex Dissipation in Sn-Segregated Boundaries in Nb₃Sn SRF Cavities

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We study mechanisms of vortex nucleation in Nb₃Sn SRF cavities using a combination of experimental, theoretical, and computational methods. Scanning transmission electron microscopy (STEM) image and energy dispersive spectroscopy (EDS) of some Nb₃Sn cavities show Sn segregation at grain boundaries in Nb₃Sn with Sn concentration as high as ~ 35 at.% and widths ~ 3 nm in chemical composition. Using ab initio calculations, we estimate the effect excess tin has on the local superconducting properties of the material. We model Sn segregation as a lowering of the local critical temperature. We then use time-dependent Ginzburg-Landau theory to understand the role of segregation on magnetic vortex nucleation. Our simulations indicate that the grain boundaries act as both nucleation sites for vortex penetration and pinning sites for vortices after nucleation. Depending on the magnitude of the applied field, vortices may remain pinned in the grain boundary or penetrate the grain itself. We estimate the superconducting losses due to vortices filling grain boundaries and compare with observed performance degradation with higher magnetic fields. We estimate that the quality factor may decrease by an order of magnitude (10^{10} to 10^9) at typical operating fields if 0.03% of the grain boundaries actively nucleate vortices. We additionally estimate the volume that would need to be filled with vortices to match experimental observations of cavity heating.

I. INTRODUCTION

Superconducting Radio-Frequency (SRF) cavities are used in accelerators to transfer energy to beams of charged particles. Induced magnetic fields are a fundamental limit to performance due to stability of the of the superconducting Meissner effect, i.e., perfect diamagnetism. For type-II materials, complete flux expulsion is thermodynamically stable up to a lower-critical field, H_{c1} , and a mixed state characterized by superconducting vortices is stable for fields up to an upper-critical field, H_{c2} . Thus, by limiting the fields on the walls of the SRF cavities, the superconductor can be kept in the flux-free Meissner state, so that surface dissipation is extremely small and quality factors $\sim 10^{10}$ can be achieved. For magnetic fields parallel to the cavity surface, the superconducting Meissner state can be maintained above the stability limit in a metastable state up to a limit (for an ideal surface) of the so-called superheating field H_{sh} [1]. H_{sh} has been studied extensively by the condensed matter community, primarily in the context of

Ginzburg-Landau theory at ideal interfaces[2–5]. Because high-power SRF cavities routinely operate in the metastable regime[6], there has been renewed interest by condensed matter community of the behavior of superconductors in large magnetic fields. Calculations extend results to the semi-classical theory of Eilenberger theory in both the clean[7] and dirty[8] limits and Time-Dependent Ginzburg Landau (TDGL) simulations that account for material[9] and spatial inhomogeneities[10–13]. In this paper, we explore the role of grain boundaries (GBs) in SRF cavity performance motivated by experimental observations of inhomogeneities in real-world SRF cavities. This study brings together the expertise of many areas of condensed matter and accelerator physics to explore fundamental physics of superconductors in extreme conditions and connect those results to real systems.

Recently there has been significant progress towards the employment of Nb₃Sn in SRF cavities as a higher performance alternative to the industry standard Nb for next generation particle accelerator applications [14, 15]. Nb₃Sn cavities are prepared with Nb₃Sn films ~ 2 μ m (nearly 20 penetration depths) thick coated on the surface of Nb cavities using the Sn vapor-diffusion process. Nb₃Sn is an intermetallic alloy with A15 crystal struc-

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ture; it is a promising material for next-generation SRF cavities in large part because of its large (predicted) superheating field (~ 400 mT). It also has a higher critical temperature ($T_c \sim 18$ K), making it possible for it to have a higher quality factor (Q_0 , another critical metric of cavity performance) at a given temperature compared to Nb ($T_c \sim 9$ K).

In practice, however, real world Nb_3Sn cavities quench well-below the theoretically predicted value. The maximum accelerating gradient that has been achieved within these cavities is about 22 MV/m, which corresponds to the surface magnetic field of ~ 90 mT. These cavities exhibit a high Q_0 of $\sim 10^{10}$ at 4.2 K [14, 16]; however, in some cavities, Q_0 begins to degrade significantly before the limiting quench field is reached, a phenomenon described as “Q-slope” [17].

Understanding the mechanism underlying the Q-slope phenomenon is an important question for cavity development. Several mechanisms have been proposed in terms of imperfections in the Nb_3Sn coatings [6, 18, 19] such as thin grains [20, 21] and Sn-deficient regions [22]. Another potential mechanism that may have detrimental effects on the performance of Nb_3Sn cavities is Sn segregation at grain boundaries [23]. In some Nb_3Sn coatings, tin concentrations as high as ~ 35 at.% have been observed in grain boundaries with the segregated zone extending by as much as ~ 3 nm, comparable to the coherence length of Nb_3Sn (~ 3 nm). Because of the inferior superconducting properties, magnetic flux may penetrate the segregated region, degrade Q_0 , and lead to premature quench.

In support of this hypothesis, witness samples coated with high-performance (quench at ~ 22 MV/m with $Q \sim 10^{10}$ at 4.4 K) Nb_3Sn cavities at Fermilab did not show Sn segregation at the grain boundaries in energy dispersive X-ray spectroscopy (EDS) and in scanning transmission electron microscopy (STEM). Similarly, a direct cutout from a high-performance Nb_3Sn cavity fabricated at Cornell did not show Sn segregation at grain boundaries within the detection limit of STEM-EDS. In contrast, Nb_3Sn cavities, which show Sn segregation at grain boundaries in witness samples that coated together with the cavities, displayed negative Q-slope for accelerating fields in the 5-15 MV/m range, see Fermilab cavity 1 and 2, Fig. 1. These experimental results, summarized in Fig. 1, suggest a potential link between Sn segregation at grain boundaries and cavity performance [23].

Experimentally, it is difficult to isolate the effects of Sn segregation at grain boundaries from other imperfections, such as Sn-deficient regions and surface roughness. To overcome these challenges, we use numerical tools to theoretically understand the role of segregation in grain boundaries for SRF cavity performance. We use density functional theory to estimate the effective T_c of the material within the segregation zone. Next, we use time-dependent Ginzburg-Landau simulations with spatial varying material properties motivated by the ab initio DFT calculations. Time-dependent Ginzburg-Landau theory allows us to conduct numerical experiments on

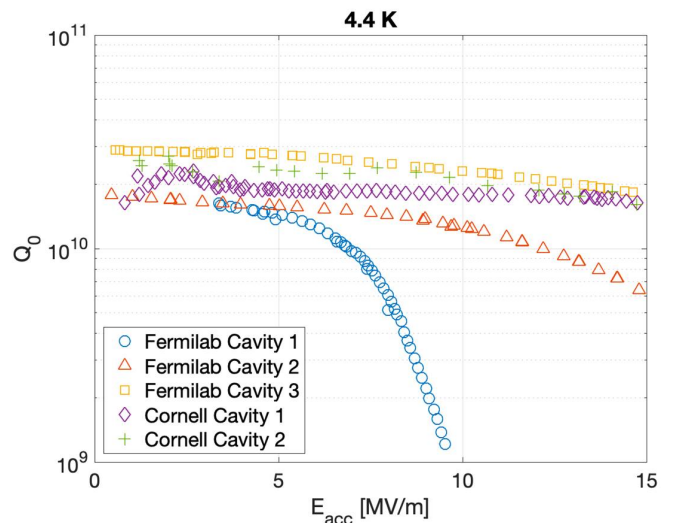


FIG. 1. Q vs E curve of various Nb_3Sn SRF cavities coated at Fermilab and Cornell. The GBs of a witness sample (Fermilab Cavity 3) and direct cutout (Cornell Cavity 2) of a high-performance cavity are characterized in STEM-EDS and showed no Sn segregation at GBs. On the other hand, witness samples of Fermilab Cavity 1 and 2, which show Q-slope starting at 8 MV/m, were found to have Sn segregation at GBs (reprint from [23]).

a mesoscale that probe the role of grain boundaries and segregated zones for vortex nucleation, pinning, and quenching. Finally, motivated by our TDGL simulations, we estimate power dissipated by vortex nucleation within segregated grain boundaries during an RF cycle and make quantitative comparisons to actual SRF cavities.

This paper is organized as follows: First, we present experimental images of defects in Nb_3Sn cavities in section II. We then report on first principles DFT calculations of superconducting properties for segregation zones in section III and time-dependent Ginzburg-Landau simulations of flux penetration in section IV. We estimate the effect on cavity performance in section V. Our numerical experiments isolate the effects of Sn-segregated grain boundaries from potentially confounding mechanisms. Our results indicate that the effects of Sn-segregated grain boundaries alone are consistent with observed behaviors. Specifically, grain boundaries may nucleate and then trap a limited number of vortices, leading to degradation in the cavity’s quality factor. We conclude by discussing the implications of these results for cavity development and further theoretical studies in section VI.

II. EXPERIMENTAL IMAGES OF Nb_3Sn DEFECTS

The high angle annular dark field (HAADF)-STEM image in Figure 2 displays a Nb_3Sn coating on Nb prepared by the Sn vapor diffusion process using coating

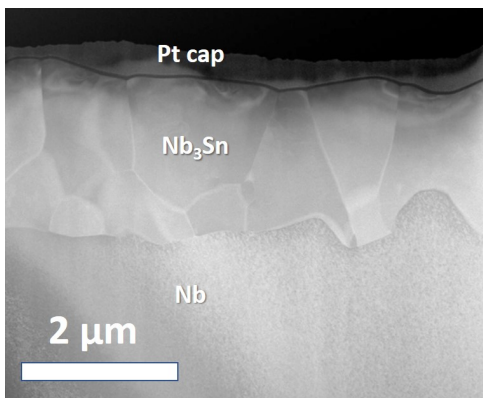


FIG. 2. HAADF-STEM image of a typical $\sim 2 \mu\text{m}$ thick Nb_3Sn coating on Nb prepared by Sn vapor-diffusion.

parameters from the early stage of the development of Nb_3Sn films at Fermilab [14]. EDS mapping is performed across a GB in a Nb_3Sn coating prepared by the same coating parameters and it reveals that Sn is segregated at the GB, Figure 3. A maximum concentration of Sn at the GB is $\sim 33 \text{ at.}\%$ and a width of the Sn segregated region is $\sim 3 \text{ nm}$. The Gibbsian interfacial excess of Sn is $\sim 16 \text{ atom/nm}^2$. Previous studies on analyses of structures of Sn-segregated GBs in Nb_3Sn employing HR-STEM and first-principle calculations indicated that most of the segregated Sn exist as Sn-antisite defects near the GBs rather than forming Sn-rich phases such as Nb_6Sn_5 or other non-equilibrium phases [23, 24].

Another GB from a witness sample of a high-performance cavity prepared at Fermilab is characterized by HR-STEM EDS, Fig. 4. It is noted that there is no Sn segregation at the GB within the detection limit of EDS ($\sim 1 \text{ at.}\%$). This may indicate that there could be a possible correlation between the Sn segregation at GBs and cavity performance. It has been reported that Sn segregation is caused by Sn diffusion via GBs due to high Sn supply and it can be controlled using carefully selected coating parameters [23].

Also, the dips are formed on the surface at GBs and HAADF-STEM image in Fig. 5 displays the geometry of a GB on the top surface. It has $\sim 80 \text{ nm}$ of depth and $\sim 420 \text{ nm}$ of width. The composition and surface roughness change at the GBs, possibly providing pathways for flux to penetrate through the imperfections. These experimental observations are the motivation for our *ab initio* and Ginzburg-Landau modeling to investigate the effect of the imperfections on the vortex penetration.

III. THE EFFECT OF TIN-RICH STOICHIOMETRY ON T_c

The presence of tin-rich stoichiometry near grain boundaries has been established experimentally, but because these regions are so small, it is difficult to directly probe their superconducting properties. We therefore

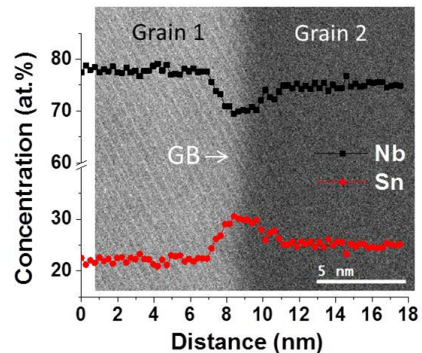


FIG. 3. The HAADF-STEM image and corresponding Nb and Sn concentration profiles across the GB between Grain 1 and Grain 2. Sn is segregated at the GB up to $\sim 33 \text{ at.}\%$ and the width of the Sn segregated region is $\sim 3 \text{ nm}$.

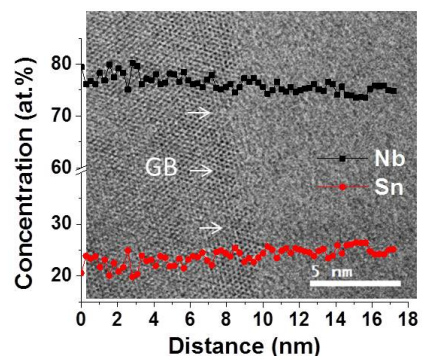


FIG. 4. BF-STEM and corresponding Nb and Sn concentration profiles across a GB from a witness sample of high-performance Nb_3Sn SRF cavity prepared at Fermilab.

consider *ab initio* T_c values calculated using Eliashberg theory [25] and Density Functional Theory (DFT) [26]. Ref. [27] presents such results obtained using a Wannier-based k-point sampling approach [28]. For the experimentally measured stoichiometry range of the A15 phase, the predicted T_c values are similar to or modestly higher than

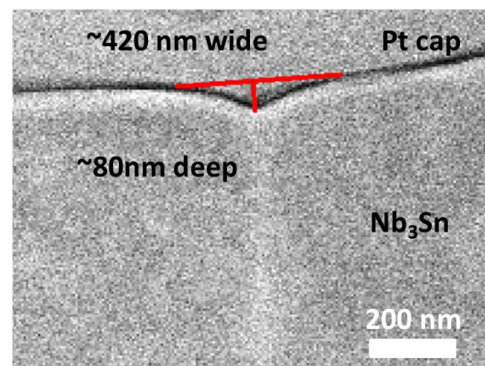


FIG. 5. HAADF-STEM image of the cross-section of the top surface of a Nb_3Sn GB.

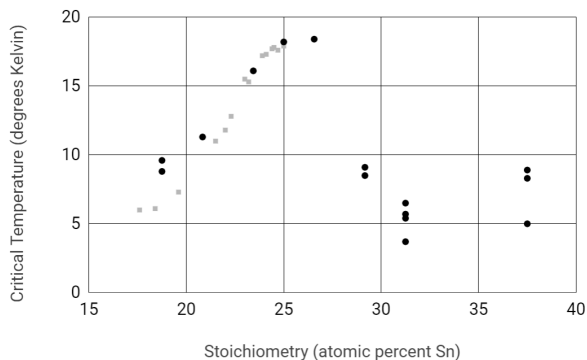


FIG. 6. Experimental T_c [29] (grey squares) and calculated T_c (black circles) for A15 Nb-Sn of different stoichiometries. For some stoichiometries, multiple possible defect configurations were considered. The calculated T_c reaches a minimum of about 5 Kelvin in the tin-rich regime.

experimental values, as described in Table 1. For experimentally inaccessible tin-rich stoichiometry, T_c values fall to a minimum of about 5K at 31.25% Sn stoichiometry (Fig. 6). This is well within the range that has been observed around grain boundaries.

TABLE I. Calculated vs. Measured T_c

Composition	Experimental T_c (K)	Calculated T_c (K)
18.75% Sn	6	9.2 [†]
20.83% Sn	9.5	11.3
23.44% Sn	16	16.1
25.00% Sn	18	18.2
31.25% Sn	n/a	5.3 [†]

[†] Averaged over multiple configurations.

IV. TIME-DEPENDENT GINZBURG-LANDAU SIMULATIONS OF VORTEX NUCLEATION

A. Introduction to Methods

Time-dependent Ginzburg-Landau (TDGL) theory is sophisticated enough to capture vortex dynamics without becoming too algebraically complicated and computationally expensive. We solve the TDGL equations using a finite element method implemented in FEniCS[30, 31]. We follow the finite element formulation described by Gao et. al[9, 32]. Note that in this formulation length is measured in units of the penetration depth λ .

This formulation reduces the full three-dimensional problem into a two-dimensional problem by assuming symmetry along the z-axis. The magnetic field points along the z-axis, fixing variations in the order parameter and magnetic vector potential to the x-y plane.

We consider the film geometry seen in Figure 7. We take a rectangular cross-section lying in the x-y plane. We enforce periodic boundary conditions on the left and right side. On the top and bottom, we enforce Dirichlet

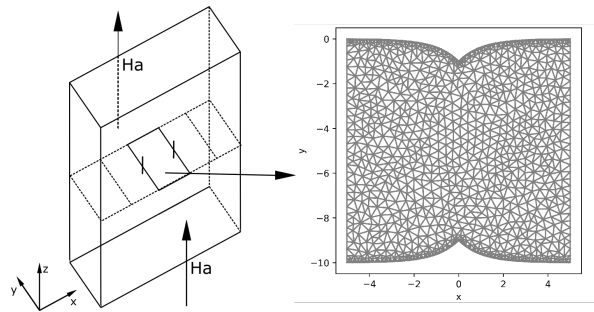


FIG. 7. The black square interior to the film geometry marks our domain of simulation. It lies perpendicular to the applied magnetic field H_a . We assume there are no variations in the direction of H_a so that we can simulate a 2D domain.

boundary conditions for the magnetic field and Neumann boundary conditions for the order parameter.

We model geometric defects by removing an exponential cut out from the top and bottom of the film. The region removed is given by $de^{-\frac{|x|}{w}}$ where d is the depth of the cut out and w determines the width. The depth and width are chosen to match experimentally observed geometries.

To capture Sn segregation we allow T_c to vary over the domain. This is done by varying the $\alpha \propto T/T_c - 1$ parameter as described in [9]. To mimic the distribution of material inhomogeneities shown in Figure 3, we introduce $\alpha' = (T/T_c - 1)/\alpha_{ref} \leq 1$ in the GB region $|x| \leq \xi/2$ and $\alpha_{ref} = -1$ elsewhere. The projection of this onto the mesh is shown in Figure 8.

The Ginzburg-Landau parameter for Nb₃Sn is $\kappa = \lambda/\xi \sim 26$, which is challenging to simulate because of the extreme separation in length scales that require a very refined mesh. However, the relevant physics for vortex nucleation are variations in material parameters on length scales comparable to the superconducting coherence length, ξ . Therefore, we have simulated a moderate type-II material ($\kappa = 4$) but scaled the width of the segregated region (i.e., depleted T_c) so that its dimensions relative to ξ are the same as that observed in Figure 3. Although these assumptions may affect quantitative details, we expect the qualitative results are the same for more extreme type-II materials.

B. Vortex Nucleation in Grain Boundaries

To simulate the nucleation of vortices into a grain boundary, we set the value of the magnetic field at the top and bottom boundary such that it is low enough that an array of vortices do not penetrate directly into the bulk, but large enough for vortices to enter into the grain boundary[9]. As we evolve time (assuming a constant applied field), two different behaviors are observed depending on the magnitude of the applied field. In the first scenario, vortices enter into the grain boundary at

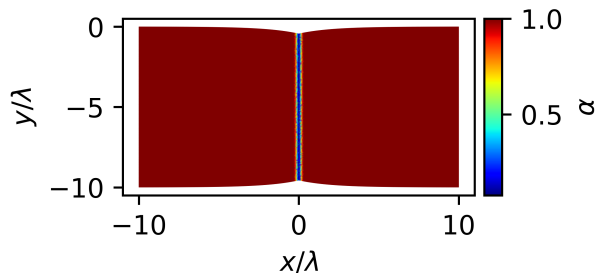


FIG. 8. Projection of $\alpha' = 1 - T/T_c$ onto a mesh, where $\alpha' = 0$ (i.e., $T \approx T_c$) in the region $|x|/\lambda \leq 0.125$ and $\alpha = 1$ elsewhere. The width of this region comes from experimental observations.

the geometric divot. With increasing field, the spacing between vortices decreases until it reaches critical levels. In the second scenario, vortices first fill the grain boundary, as in the first case, but then begin to penetrate into the grain from boundary.

Once a vortex has penetrated into the grain boundary, it is pushed from the surface, allowing more vortices to come in after it. Once space is available, another vortex penetrates. This continues until the vortices have filled the grain boundary, i.e. an optimal spacing between the vortices inside grain boundary has been achieved. This is illustrated by the sequence in Figure 9. Note that vortices are manifest as regions in which the order parameter is reduced to near zero at their center and exponentially decay radially outward to unity.

If the applied magnetic field is sufficiently high, vortices will also begin to penetrate into the bulk once the grain boundary has been filled, Figure 9, bottom. The field at which vortices penetrate from the grain boundary into the grain is dependent on the distribution of $\alpha' = 1 - T/T_c$. The shallower the slope of α' the lower the applied field needs to be to nucleate vortices into the grain from the grain boundary. These results are summarized in the phase diagram in Figure 10. Comparing with results from section III, for a segregated region with $T_c \sim 5K$ in a cavity operating at $T = 4.2K$ ($T/T_c \sim 1$), we observe a non-trivial region of the phase diagram that admits flux trapped at the grain boundary.

The value of the applied at which the vortices first leave the grain boundary and penetrate the bulk depends on the properties in the transition zone between the segregated and non-segregated region. If the transition from $\alpha' < 1$ (segregated region) to $\alpha' = 1$ (non-segregated region) is very sharp (as the blue solid curve in Figure 11), then vortices will be trapped in the grain boundary for larger fields. However, if the transition is more gradual (such as the orange dashed curve), then it is easier for vortices to escape the boundary and penetrate the

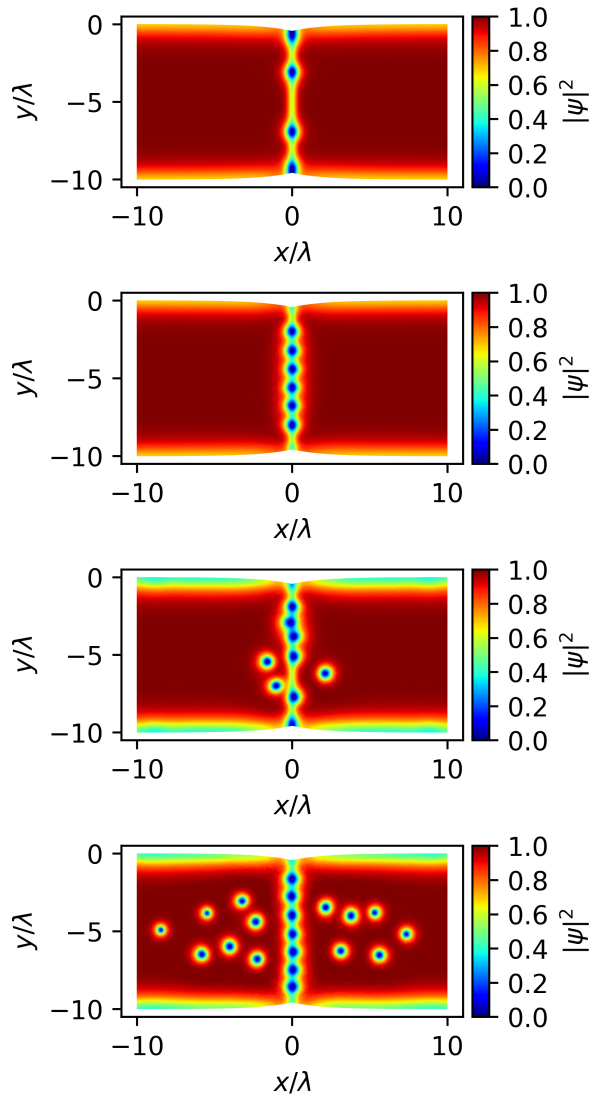


FIG. 9. Sequence of behaviors for increasingly large applied magnetic fields. An applied magnetic field is set as a boundary condition to the top and bottom of the film, with periodic boundary conditions on the right and left sides. At moderate applied magnetic fields, vortices penetrate only into the grain boundary from the geometric defect (top). At higher fields, a critical vortex spacing is reached (second pane) above which vortices begin to penetrate the bulk from the grain boundary (third pane). Finally, having entered the bulk, vortices disperse and fill the grain with an equilibrium distribution (bottom).

bulk. Figures [8, 9, 10] were generated with a very sharp interface.

V. ESTIMATES OF VORTEX DISSIPATION AT GRAIN BOUNDARIES

Inhomogeneous properties of superconductors have high impact on the performance of SRF cavities, affect-

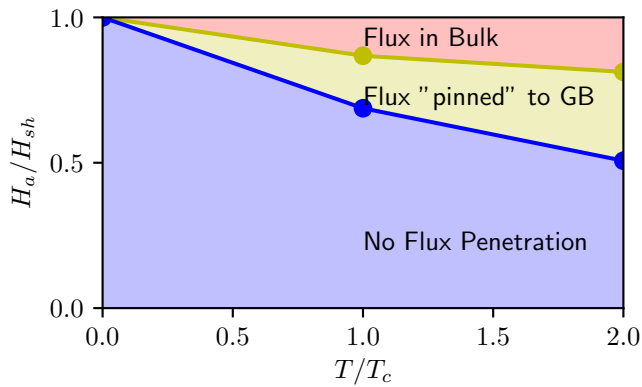


FIG. 10. Phase diagram of TDGL predictions for flux penetration in the presence of the grain boundary, interpolated from simulations at $\alpha' = -1, 0, 1$. At small applied fields, no flux penetrates (blues). At intermediate fields, flux penetrates but is constrained to the grain boundary (yellow). At sufficiently high fields, the flux penetrates from the grain boundary in to the bulk material (red).

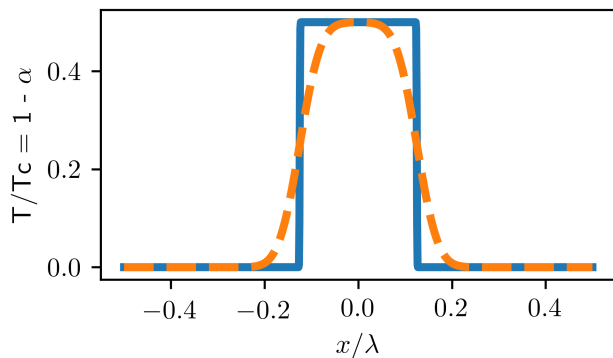


FIG. 11. Profiles of potential α' for the transition between the segregated and non-segregated regions. Sharp transitions (such as the blue solid curve) keep the vortices constrained to the boundary for larger applied fields. A more shallow transition (such as the orange dashed curve), however, allow vortices to escape into the grain more easily.

ing figures of merit such as the residual resistance due to trapped magnetic flux [33–35], and the superheating field [1, 2, 7, 36–38]. Grain boundaries well aligned with the surface magnetic field can become weak spots for the nucleation of superconducting vortices (see Fig. 12), and could be ultimately responsible for the quench of an SRF cavity. In this section, we discuss simple estimates for the power dissipation and heat diffusion due to nucleation of vortices at grain boundaries in Nb_3Sn cavities.

We start with an estimate for the power dissipated in an SRF cavity when a grain boundary is filled with n superconducting vortices. We assume of order $\mathcal{O}(n)$ vortex lines are annihilated once per cycle, their energy is lost, and the power dissipated per vortex line is simply the

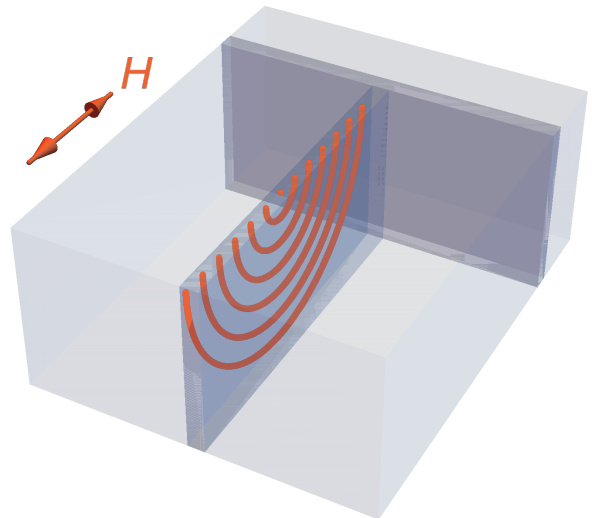


FIG. 12. Illustrating vortex nucleation (red lines) in a superconductor (light gray region) subject to a surface magnetic field H . Vortex entry starts at regions where superconductivity is weakest (dark gray regions, here representing grain boundaries).

drop in the energy of the outside magnetic field times the RF frequency. Our estimate gives a rough estimate for the actual power dissipated by vortices at grain boundaries, if the field reaches high enough values for them to enter.

The drop in magnetic energy when a vortex line of length D nucleates into the superconductor is given by:

$$\Delta E = \left| \int \frac{1}{2\mu} \left(B - \frac{\Phi_0 D}{V} \right)^2 dV - \int \frac{B^2}{2\mu} dV \right| \approx \frac{B\Phi_0 D}{\mu}, \quad (1)$$

where V is the volume, μ is the permeability of free space, Φ_0 is the fluxoid quantum, and λ is the penetration depth. Note that ΔE is also approximately the work done by the external magnetic field to push a vortex into the bulk of the superconductor: $W \approx f_L \cdot D \cdot \lambda = (\Phi_0 B / (\mu\lambda)) D \lambda = \Delta E$, where f_L is the Lorentz force per length. Thus, our calculation gives the vortex dissipation at grain boundaries assuming that the vortices do not leave the grain boundary as the external field drops and changes sign, as our simulations indicate, and as one would expect for vortices that enter a distance more than λ , past the surface nucleation barrier. The total energy drop for a grain boundary of linear size D with vortices spaced by λ (see Fig. 12) is $\Delta E_{GB} \approx \Delta E(D/\lambda) = B\Phi_0 D^2 / (\mu\lambda)$, and the power dissipated per grain boundary is given by

$$P_{GB} \equiv f \Delta E_{GB} = \frac{B\Phi_0 f D^2}{\mu \lambda}, \quad (2)$$

where f is the cavity frequency. For a 1.3GHz Nb₃Sn cavity with typical grain size of $D \approx 1\mu\text{m}$, we find $P_{GB} \approx 621\text{nW}$ at $B = 60\text{mT}$.

Note that our estimate rely on the assumption that vortices quickly fill the grain boundary before being annihilated during the RF cycle. If the vortex line relaxes slowly, the RF field might vanish and change sign before vortices had time to fill the grain boundary, and our assumption would not be valid. We now show that that is not the case — vortices move at extremely high speeds in the typical environment of Nb₃Sn SRF cavities.

Consider the one-dimensional motion of a “train” of N vortices moving through a grain boundary towards the superconductor interior (see Fig. 13). Assuming overdamped dynamics, the equations of motion for each vortex line i of velocity v_i read:

$$\begin{aligned} \eta v_1 &= f_L - f_{2,1}, \\ \eta v_2 &= f_{1,2} - f_{3,2}, \\ &\vdots \\ \eta v_{N-1} &= f_{N-2,N-1} - f_{N,N-1}, \\ \eta v_N &= f_{N-1,N}, \end{aligned}$$

where $f_L = \Phi_0 H_{rf} / \lambda$ is the Lorentz force per length at the surface, H_{rf} is the surface magnetic field, $\eta = \Phi_0^2 / (2\pi \xi^2 \rho_n)$ is the Bardeen-Stephen viscosity [39], ρ_n is the resistivity of the normal state and $f_{i,j}$ is the repulsion force from vortex i into vortex j . Thus,

$$\langle v \rangle \equiv \frac{1}{N} \sum_{i=1}^N v_i = \frac{f_L}{N\eta} = \frac{2\pi}{\mu\Phi_0} \frac{\rho_n \xi^2}{\lambda} \frac{B_{rf}}{N}. \quad (3)$$

For Nb₃Sn at $B_{rf} = 60\text{mT}$, we find $\langle v \rangle = 2.4\mu\text{m/ns}$ and $\langle v \rangle = 24\mu\text{m/ns}$ for ten vortices and one vortex, respectively. The average velocity of the vortex train is clearly high enough for vortices to quickly fill in the grain boundary during the RF cycle, but the numerical value should be taken with a grain of salt. The Bardeen-Stephen formula is not valid at these high speeds, which also exceed the pair-breaking limit of the superconducting condensate [40]. In recent work [41], Gurevich and Sheikzada consider the dynamics of Abrikosov-Josephson vortices to model the dissipation by vortices at grain boundaries.

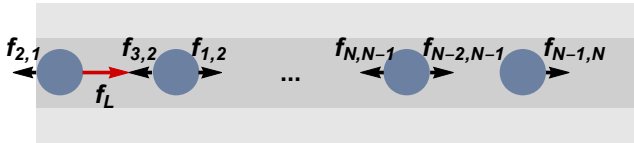


FIG. 13. Illustrating a “train” of N vortex lines (blue disks) moving through a grain boundary (dark gray) of a superconductor (light gray). The first vortex is subject to a surface Lorentz force from the RF field and each vortex is repelled by its nearest neighbors.

Grain boundary activation might be associated with the degradation of the quality factor Q of SRF cavities

at high fields. We now use our estimates to calculate the number of active grain boundaries needed to deplete Q by a certain amount.

The quality factor is given by $Q = GB_{rf}^2 / (2\mu^2 P)$, where P is the dissipated power per unit area and G is a geometry factor. We break up the total surface area s of the cavity into N blocks, so that $s = N s_{GB}$, where s_{GB} is the average area occupied by one grain boundary. Assume inactive and active blocks dissipate power P_1 and $P_1 + P_{GB}$, respectively, where by active block we mean a block with a grain boundary filled with vortices. For M active blocks,

$$\begin{aligned} Q &= \frac{GB_{rf}^2}{2\mu^2(NP_1 + MP_{GB}) / (N s_{GB})} \\ &= \frac{GB_{rf}^2 s_{GB}}{2\mu^2(P_1 + x P_{GB})}, \end{aligned} \quad (4)$$

where $x \equiv M/N$ is the ratio of active grain boundaries. In the absence of active grain boundaries, we assume $Q = Q_1$ is constant (i.e. $P_1 \sim B_{rf}^2$), so that

$$P_1 = \frac{GB_{rf}^2 s_{GB}}{2\mu^2 Q_1}. \quad (5)$$

Plugging Eq. (5) into Eq. (4) and solving for x , we find

$$x = \frac{G s_{GB} B_{rf}^2}{2\mu^2 P_{GB}} \left(\frac{1}{Q} - \frac{1}{Q_1} \right). \quad (6)$$

Figure 14 shows a plot of the percentage of active grain boundaries ($100x$, blue curve) as a function of B_{rf} corresponding to the artificial Q -slope profile shown in the yellow curve (using Nb₃Sn parameters with $Q_1 = 10^{10}$, $s_{GB} = 0.5\mu\text{m}^2$ and $G = 278\Omega$). Note that about 0.03% of the surface grain boundaries need be filled with vortices for Q to drop from 10^{10} to 10^9 for Nb₃Sn at about 66mT.

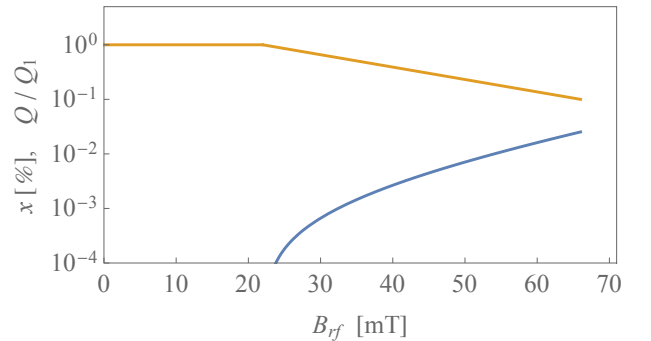


FIG. 14. Estimated percentage of active surface grain boundaries (blue curve) as a function of B_{rf} , corresponding to the quality factor profile displayed in the yellow curve for Nb₃Sn with $Q_1 = 10^{10}$.

We end this section with a simple model calculation of the steady-state thermal heating at a grain bound-

ary. Figure 15 illustrates our model for thermal diffusion near an active grain boundary. Light and dark gray regions represent cross sections of Nb and Nb₃Sn layers, respectively. The Nb surface is in contact with a low-temperature He bath. The Nb₃Sn surface is in contact with vacuum, and is subject to a parallel oscillating magnetic field. The grain boundary is represented by a blue rectangle of linear size D . Red arrows represent the direction for heat diffusion in our model. First, we assume one-dimensional heat diffusion away from the grain boundary up to a distance $r \approx D$. We expect the heat front to attain a semi-spherical shape for distances $r \gtrsim D$. We then assume three-dimensional heat diffusion away from a half-sphere of radius D for distances $D \leq r \leq R_1$. For $r \leq R_1$ we consider the Nb₃Sn thermal conductivity $\kappa = \kappa_{\text{Nb}_3\text{Sn}}$. At last, we assume three-dimensional heat diffusion away from a half-sphere of radius R_1 for distances $R_1 \leq r \leq R_2$, with κ given by the Nb thermal conductivity κ_{Nb} . Note that our assumptions are stronger when $R_2 \gg R_1$ ($R_2/R_1 \sim 10^3$ for typical Nb₃Sn/Nb SRF cavities).

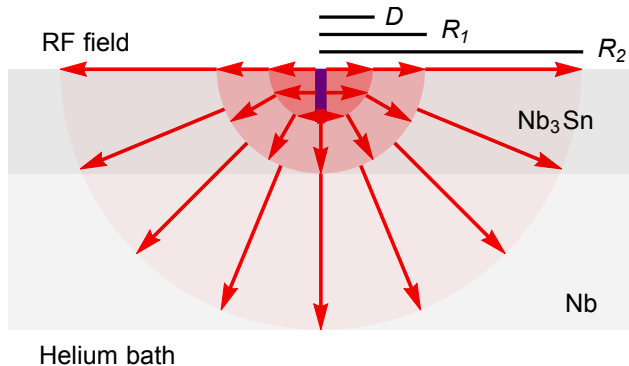


FIG. 15. Sketch of our model for heat diffusion near an active grain boundary (blue rectangle at the top center). The light and dark gray regions correspond to cross sections of Nb and Nb₃Sn. Red arrows represent the direction for heat diffusion. Heat fronts move along one dimension for distances smaller than the grain size D , and three dimensions otherwise.

The equilibrium temperature profile can then be cast from the stationary solutions of the heat equation in each region:

$$T(r) = \begin{cases} \zeta(r), & \text{for } 0 \leq r \leq D, \\ c/r + d, & \text{for } D \leq r \leq R_1, \\ e/r + f, & \text{for } R_1 \leq r \leq R_2, \end{cases} \quad (7)$$

where c , d , e and f are constants, and $\zeta(r)$ is the stationary solution of the one-dimensional heat equation (the Nb₃Sn thermal conductivity strongly varies with temperature in this region [42], which complicates the problem of finding an analytical solution for $\zeta(r)$) [43]. Note that we relax the definition of the coordinate r here, which should be interpreted as a lateral distance away from the grain boundary for $0 \leq r \leq D$, and a depth coordinate towards the Helium bath for distances $r > D$.

To calculate $\zeta(r)$, we use Fourier's law — $\dot{Q} = -\kappa dT/dr$, where \dot{Q} is the heat flux. The stationary solution of the heat equation can be found from the solution of $d\dot{Q}/dr = 0$, *i.e.*

$$-\kappa_{\text{Nb}_3\text{Sn}}(T) \frac{dT}{dr} = a, \quad (8)$$

where a is constant.

The thermal conductivity $\kappa_{\text{Nb}_3\text{Sn}}(T)$ in the superconducting layer has two important contributions: a phonon contribution and an electronic component (carried by superconducting quasiparticles). The low-temperature phonon thermal conductivity is strongly dependent on the morphology of the crystal [44]; in clean insulating crystals it is dominated by scattering off of grain boundaries and sample boundaries, and varies as T^3 . Scattering off of impurities can cut off the contribution of high-frequency phonons, or even resonantly cut off certain frequency bands. All of these mechanisms lead to a thermal conductivity that monotonically increases with temperature, so we avoid the complexity by using a constant phonon thermal conductivity k_1 , giving a lower bound for the conductivity and hence an upper limit to the heating. The electronic portion of the thermal conductivity k_2 in the normal metal at low temperatures is roughly independent of temperature, and is set by the electronic mean-free path. In the superconductor, it decreases exponentially as $\exp(-\Delta(0)/k_B T)$, as seen experimentally [42]. Using the BCS relation between the gap and the transition temperature, we therefore use

$$\kappa_{\text{Nb}_3\text{Sn}}(T) = k_1 + k_2 \exp(-1.76 T_c/T). \quad (9)$$

We use the normal electron thermal conductivity $k_2 = 2 \times e^{1.76} \text{ W/m}\cdot\text{K}$ from [42]. Because the electronic contribution is negligible at the operating temperature of the cavity, we set $k_1 = 10^{-2} \text{ W/m}\cdot\text{K}$ as the approximate total thermal conductivity of Nb₃Sn at 2K [42]. Both of these constants are dependent upon the preparation of the film, and also could vary from one region of the film to another as the growth conditions or the underlying Nb grain orientations vary.

Integration of Eq. (8) results in

$$\Pi(T) = -ar + b, \quad (10)$$

where b is constant, and

$$\Pi(T) = k_1 T + k_2 T e^{-1.76 T_c/T} + 1.76 k_2 T_c E_i \left(-1.76 \frac{T_c}{T} \right), \quad (11)$$

with $E_i(x) \equiv \int_{-x}^{\infty} [e^{-t}/t] dt$ denoting the exponential integral function. $\zeta(r)$ is then the solution of Eq. (10) for T . Note that our simple model assumes that the quasiparticles and the phonons remain at the same effective temperature (the inelastic electronic mean free path is small), and that both remain diffusive (the elastic phonon and

electron mean free paths are small). Violating either of these assumptions would likely lower the transport of energy away from the grain boundary, making the heating more dangerous.

We focus our attention on grain-boundary activation and ignore other sources of power dissipation (which could be added later using the principle of superposition). Thus, we assume that the heat flux is the power dissipated per grain boundary (P_{GB}) per unit area. We use Fourier's law to determine the coefficients a , c and e in Eqs. (7) and (10). For $0 \leq r \leq D$, $\dot{Q} = P_{GB}/D^2$, so that:

$$a = \frac{P_{GB}}{D^2}. \quad (12)$$

For $D \leq r \leq R_1$ ($R_1 \leq r \leq R_2$), $\dot{Q} = P_{GB}/2\pi r^2$, $\kappa = \kappa_{\text{Nb}_3\text{Sn}}$ (κ_{Nb}) and $dT/dr = -c/r^2$ ($-e/r^2$), so that:

$$c = \frac{P_{GB}}{2\pi\kappa_{\text{Nb}_3\text{Sn}}}, \quad e = \frac{P_{GB}}{2\pi\kappa_{\text{Nb}}}. \quad (13)$$

To find f , we use $T(R_2) = T_{\text{He}}$, where T_{He} is the temperature of the Helium bath:

$$f = T_{\text{He}} - \frac{P_{GB}}{2\pi\kappa_{\text{Nb}}R_2}. \quad (14)$$

To find d and b , we use the continuity of $T(r)$ at $r = R_1$ and $r = D$, respectively (we ignore the Kapitza resistance at the interface between Nb_3Sn and Nb). Thus,

$$c/R_1 + d = e/R_1 + f \quad \Rightarrow \quad d = \frac{e - c}{R_1} + f, \quad (15)$$

and

$$b = aD + \Pi \left(\frac{c}{D} + d \right). \quad (16)$$

Figure 16 shows temperature profiles (Eq. (7)) near an active grain boundary for $\text{Nb}_3\text{Sn}/\text{Nb}$ systems at $B = 60\text{mT}$. Note that the temperature of an active grain boundary increases to about 10K near the boundary surface for a Helium temperature of 2K. Although this increase in temperature is not large enough to drive Nb_3Sn into the normal state, it certainly has significant impact on the superconducting properties. Also, note that the temperature decays to nearly T_{He} as r approaches twice the grain size D , suggesting that heating due to grain-boundary activation is mostly localized.

A temperature rise of 10K at the grain boundary is over half of the critical temperature of the film, suggesting that larger grain boundaries or multiple nearby boundaries could raise the temperature high enough to quench the cavity. Cavities with tin-rich grain boundaries and more pristine grain boundaries show the same quench fields, suggesting that another mechanism controls the quench fields of existing Nb_3Sn cavities. If the excess dissipation in the cavities with tin-rich boundaries is due to vortex penetration (Fig. 14), one would expect

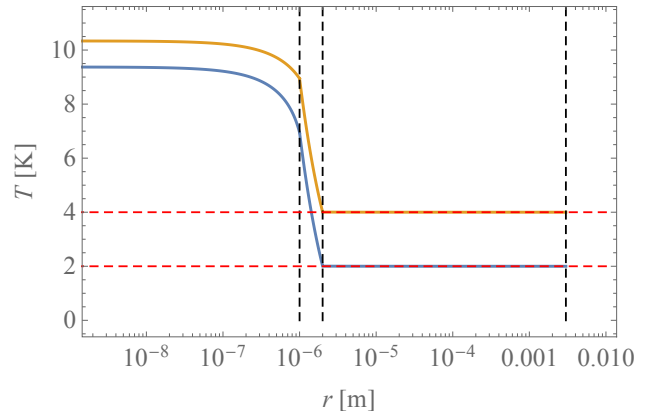


FIG. 16. Temperature as a function of distance from the grain boundary for the $\text{Nb}_3\text{Sn}/\text{Nb}$ system at a bath of 2K (blue) and 4K (yellow). Vertical dashed lines correspond to $D = 1\mu\text{m}$, $R_1 = 2\mu\text{m}$ and $R_2 = 3\text{mm}$, from left to right. Bottom and top red dashed lines correspond to $T_{\text{He}} = 2\text{K}$ and 4K, respectively. We have used temperature-dependent $\kappa_{\text{Nb}_3\text{Sn}}$ given by Eq. (9), $\kappa_{\text{Nb}} = 10\text{W}/\text{m}\cdot\text{K}$, and $P_{GB} = 621\text{nW}$ at $B = 60\text{mT}$, according to our previous estimates.

rare events with large or multiple grain boundaries would happen, suggesting that our grain-boundary heating estimate is unduly pessimistic. Alternatively, it remains possible that the grain boundaries have high superheating fields, and the excess dissipation has another explanation. In any case, our estimates suggest that vortex entry at grain boundaries should be expected for tin-rich boundaries well below the superheating field for a perfect crystal, and that the subsequent heat release should be important both as a contribution to the overall dissipation and as a quench mechanism for the cavity.

VI. CONCLUSION

In this work we have presented an interdisciplinary, multi-scale study of vortex nucleation in Sn-segregated grain boundaries and its subsequent effect on SRF performance. Scanning transmission electron microscopy images and energy dispersive spectroscopy show Sn concentration as high as ~ 35 at.% and widths $\sim 3\text{nm}$ in chemical composition in grain boundaries. We used density functional theory to estimate the effective critical temperature for the material in the segregation zone and find that the effective T_c can be reduced to as low as 5 K for Sn concentrations in excess of ~ 30 at.%. Next, we used these calculations as inputs into time-dependent Ginzburg-Landau simulations. These simulations demonstrate that grain boundaries can act as nucleation sites for magnetic vortices. The grain boundaries then act as a kind of pinning sites for vortices after nucleation. The pinning is non-traditional, however, as vortices are free to move vertically along the grain boundary, but are constrained

from moving laterally into the bulk. We have seen that for a range of applied fields, vortices may nucleate at but remain constrained to the grain boundary. These vortices will nucleate and annihilate once per RF cycle, and we estimate the superconducting losses of this phenomenon at the scale of SRF cavities. We have shown that as long as vortices do not penetrate the bulk grain, losses are localized near the grain boundary and will not lead to a global quench. However, the annihilation process each cycle will lead to a reduction in the quality factor that increases with larger applied fields, consistent with the experimentally observed Q-slope.

SRF cavities are an important application area that require multi-disciplinary talents to address. This study has leveraged the skills of accelerator physicists, material scientists, and condensed matter theorists with expertise across a range of scales to explore a question fundamental to the advancement of next-generation SRF material, Nb₃Sn. This study has presented evidence that segregation zones in grain boundaries play an important role in cavity performance. Understanding the mechanism behind the Q-slope will motivate new manufacturing protocols and help constrain the design space of future cavities.

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