THE FIELD-DEPENDENT SURFACE RESISTANCE OF DOPED NIOBIUM: NEW EXPERIMENTAL AND THEORETICAL RESULTS*

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Abstract

We present systematic work investigating how different doping and post-doping treatments affect the BCS surface resistance at 1.3 GHz and higher frequencies. We examine the field-dependent BCS resistance at many temperatures as well as the field-dependent residual resistance and use the results to reveal how impurity species and concentration levels affect the field-dependent RF properties. We further demonstrate the importance of thermal effects and their direct dependence on doping level. We use the tools of Density Functional Theory to work towards an *ab initio* model of electron overheating to theoretically confirm the impact of doping, create a full model that includes thermal effects to predict the field dependent resistance, and show that the predictions of the model agree with results from doped and non-doped cavities (*e.g.* the strength of the anti-Q-slope and the high-field Q slope). Finally, we use our experimental results to systematically assess and compare theories of the field-dependent BCS resistance, showing that the current theory on smearing of the density of states is incomplete.

INTRODUCTION

Nitrogen doping and infusion have been some of the most exciting developments in recent fundamental SRF research [1–4]. These treatments have been shown to increase the intrinsic quality factors of niobium SRF cavities at low fields and to cause a further field-dependent increase known as the "anti-Q-slope" or "positive Q-slope". Because of the project-driven nature of the field, much of this research has been focused on developing doping and infusion "recipes" that yield the highest performance. In the work presented here, we sought to perform systematic studies of infusion and doping treatments at 1.3 GHz and 2.6 GHz in an effort to improve the fundamental understanding of the physics of the positive Q-slope. Our studies also included the development of a full thermal model of the SRF cavity system, calculating the field-dependent surface resistance including the effects of quasiparticle overheating due to inefficient heat transfer. We also assessed several existing models of the positive Q-slope for their applicability to the conditions under which the phenomenon has been observed.

NITROGEN INFUSION TREATMENTS

In the context of the R&D program for the LCLS-II HE upgrade [5], the Cornell SRF group has been engaged in systematic fundamental research of impurity doping (~ 800° C) and infusion (~ 160° C) of niobium SRF cavities. Technical details of these recent Cornell experiments are presented elsewhere at this conference [6]; these results are summarized and analyzed here.

While early Cornell results of 160°C nitrogen infusion of niobium cavities as well as more recent testing of a niobium cavity with all-niobium flanges showed quite positive results [4, 6, 7], our more recent tests of cavities with niobium-titanium flanges suffered high residual resistance, likely linked to titanium contamination due to flange outgassing [6]. Attempts to ameliorate this contamination by light surface treatments like HF rinsing and cold VEP (vertical electropolishing) yielded cavities with improved performance, with removals on the order of 5 nm largely curing the contamination issue and causing a field-dependent decrease in the BCS surface resistance quite similar to that seen in other cavities with positive Q-slope. After the light removal some of these cavities indeed exhibited positive Q-slope, while others showed high residual resistance and as a result had no positive Q-slope; this was likely related to trapped flux issues and was not an intrinsic limitation in these cavities. Some of these cavities also received heavier removal on the order of 50 to 100 nm (including those that had previously suffered titanium contamination as well as those that had not); after this additional surface removal these cavities had high intrinsic quality factor but did not exhibit positive Q-slope or a field-dependent decrease in R_{BCS}. This suggests that the positive Q-slope effect seen in cavities that have been treated with "infusion" of nitrogen or other impurities is related to the physics happening in the very-near-surface layer, *i.e.* within the first 20 to 50 nm under the oxide layer.

What, then, does this tell us about the role of the various impurities introduced during doping and infusion in bringing about the positive Q-slope? Figure 1 shows a look inside the RF surface by way of SIMS (secondary ion mass spectrometry) measurements of impurity concentration in a representative cavity treated with nitrogen infusion. Very

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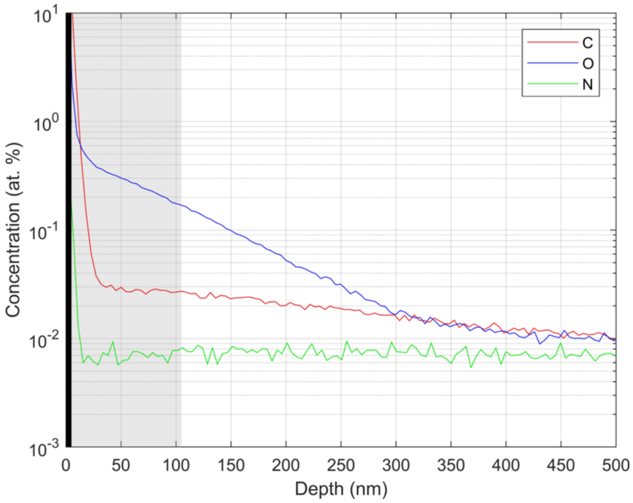

Figure 1: SIMS measurement of atomic concentration of impurities in a witness sample from a nitrogen infusion furnace run. Black bar indicates surface oxide; gray bar approximates RF penetration layer.

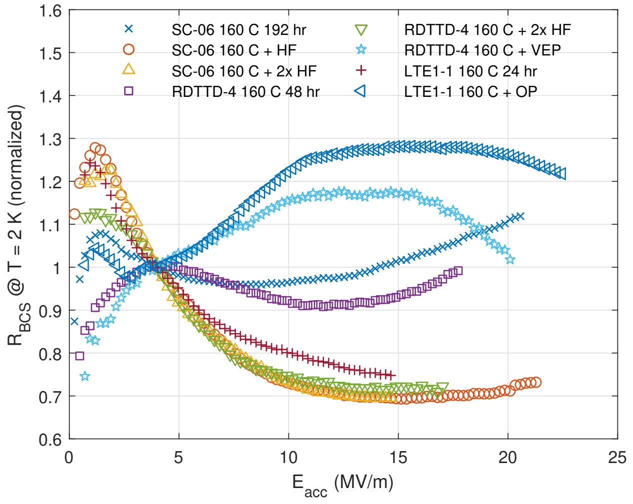

Figure 2: R_{BCS} taken for the 160 °C nitrogen infused cavities at 2 K. After infusion cavities received varying surface removal, including HF rinsing (5 nm), oxypolishing (54 nm), and light vertical electropolishing (100 nm). Peak field shown here does not indicate quench field.

close to the surface, within the first 10 to 20 nm, the carbon, oxygen, and nitrogen impurities exhibit "spikes" of greatly increased concentration. In particular, the nitrogen concentration very near the surface is very similar to the nitrogen concentration in high-temperature (*e.g.* 800 °C) nitrogen-doped niobium cavities [8]. The oxygen and carbon spikes are also dramatic but not uniformly so for nitrogen-infused cavities which show similar RF performance [6].

Because the cavities with more than 30 nm or so of chemical removal no longer exhibited a positive Q-slope, as shown in Fig. 2, it is reasonable to suppose that these concentration spikes are directly related to this field-dependent decrease in R_{BCS}. In addition, the R_{BCS} decrease is characteristically very similar for the uncontaminated cavity and the contaminated cavities after HF rinses, despite the differences in impurity concentration levels apart from nitrogen; the quantitative differences are likely accounted for by the change in RRR between the cavities. This is supported by the normalized field-dependent R_{BCS} as shown in Fig. 3. Moreover, because nitrogen is the element most thoroughly eliminated in the etches that removed the positive Q-slope behavior, the surface removal study suggests that indeed nitrogen is responsible for causing (or perhaps revealing as a more fundamental behavior) the field-dependent decrease in R_{BCS} seen in infused cavities, even by acting only in the first 10 to 20 nm of the RF surface. These results are far from conclusive, but they are certainly suggestive.

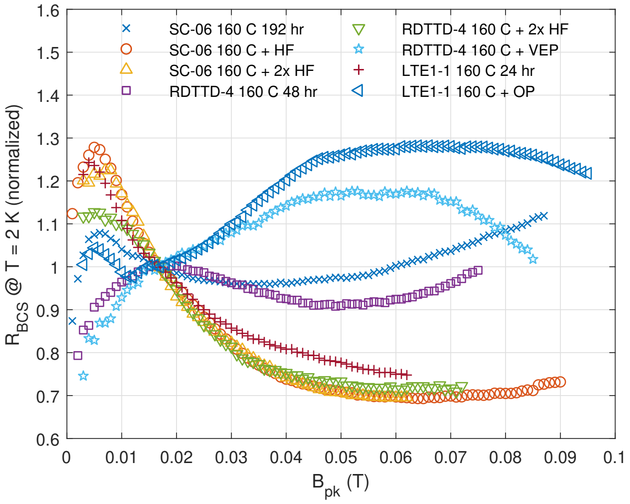

Figure 3: R_{BCS} results shown in Fig. 2, normalized at 4 MV/m.

With this hypothesis, we might next ask what possible effects a very thin layer of nitrogen impurities at a level of $< 0.1\%$ at. could have on a niobium surface. Prior experimental studies have linked surface nitrogen with suppression of niobium hydrides, which themselves have been linked to poor RF performance [9–12]. Recent DFT (density functional theory) calculations with our collaborators in the Center for Bright Beams find that interstitial nitrogen impurities near the surface of niobium suppress the formation of surface hydrides [13], consistent with the earlier experimental results. This is a promising lead for the continuing investigation into the fundamental source of the positive Q-slope.

HIGH-FREQUENCY SRF CAVITIES

Our group has also been actively engaged in studying high frequency (> 1.3 GHz) SRF cavities with modern treatments [14–17]. In general, newer surface treatments can offer greatly reduced RF dissipation and resulting cryogenic heat load; studying how these treatments behave at alternative frequencies will allow for finding more optimal frequencies for the treatments in terms of cryogenic efficiency

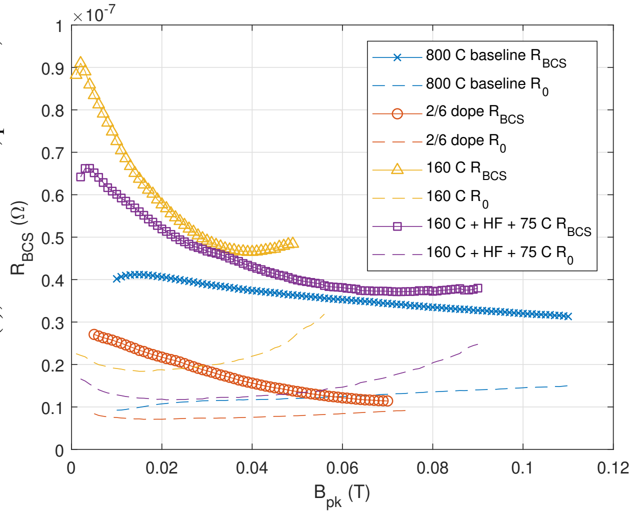

Figure 4: RF test results of several tests of 2.6 GHz cavity STE1-1. R_{BCS} taken at 2 K. Peak field shown here does not indicate quench field.

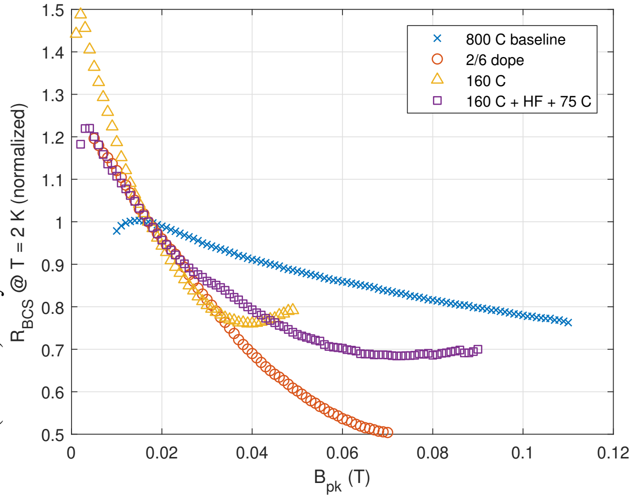

Figure 5: R_{BCS} results of the tests of 2.6 GHz cavity STE1-1, taken at 2 K and normalized at low field. Peak field shown here does not indicate quench field.

of particle acceleration. Our heaviest study at alternative frequencies for nitrogen doping and infusion has been at 2.6 GHz; Figure 4 shows the RF test results for a 2.6 GHz cavity with baseline treatment (800 °C + 6 µm VEP), 2/6 doping, 160 °C nitrogen infusion, and infusion plus HF rinsing and 6 hour 75 °C vacuum bake. All treatments except for the HF/75 °C were immediately preceded by a chemical reset of the surface by VEP. Figure 5 shows the R_{BCS} results at $T = 2$ K normalized at low field.

Remarkably, the 2.6 GHz cavity showed positive Q-slope even in the vacuum-baked baseline test, consistent with experiments elsewhere [18]. For the baseline, the cavity had an electron mean free path $\ell > 1000$ nm. 2/6 doping gave the best overall performance, with a very low R_{BCS} featuring a dramatic positive Q-slope, and $\ell = 47$ nm. The first test of the nitrogen infusion treatment had a strong field-dependent decrease in R_{BCS}, but both the BCS resistance and the residual resistance were quite high, the highest of any of the treatments at 2.6 GHz. This infusion run was done alongside one of the 1.3 GHz cavities which showed titanium contamination issues; it is possible that those issues also affected this cavity. It is also possible that the very high R_{BCS} was only related to the very short mean free path $\ell = 2.7$ nm. We performed an HF rinse as well as a 75 °C vacuum bake to try to cure potential contamination issues and investigate the possible effects of low-temperature baking for quench field increases (see [17] for more details); after these treatments, R_{BCS} and R_0 were reduced, suggesting that indeed this cavity had suffered from mild contamination during the infusion bake. The electron mean free path ℓ was increased to 4.8 nm.

Our studies of high frequency cavities will continue at 2.6 GHz as well as at 3.9 GHz and higher as we progress with our systematic studies of these surface resistance effects.

THERMAL MODELING OF SRF CAVITIES

During our early studies of nitrogen-doped niobium cavities, we found that thermal effects are quite important in controlling the magnitude of the positive Q-slope: quasiparticle overheating due to inefficient heat transfer from the quasiparticles in the RF layer out to the cryogenic bath can lead to a field-dependent increase in temperature of the quasiparticles [3]. This temperature increase in turn increases the BCS surface resistance, counterbalancing the field-dependent decrease due to the underlying positive Q-slope effect and "tuning" the overall field-dependent reduction in R_{BCS}. Our results indicated that this thermal effect was closely linked with the concentration of impurities in the RF layer, quantified by the electron mean free path, through the effect of impurities on the electron-phonon energy transfer rate.

In order to study these effects in infused cavities, which have material parameters that vary over the RF penetration layer, we developed a new framework for calculating the surface resistance of SRF cavities. Early results using this framework were published last year [16], showing good agreement between the model (using the Gurevich theory [19] as the underlying source of positive Q-slope) and experimental results of an infused cavity. Figure 6 reproduces these results.

The principle of the model is to calculate the surface resistance R_s given a model of the quasiparticle conductivity as a function of the quasiparticle temperature, local material properties, and field strength, and from this to calculate a flow of power per unit area dissipated by the RF field $P/A = H^2 R_s/2$. The framework can be used with any model of the quasiparticle conductivity, so long as it is a local (*i.e.* dirty-limit) model. We consider the electrons to be in thermal equilibrium with each other within the RF layer, though they may be at a temperature higher than that of the lattice.

The framework hinges on a self-consistent calculation of the heat flow through the cavity wall, considering the surface resistance model and a model of the niobium-helium system comprising the Nb thermal conductivity and the Nb-He Kapitza interface conductance [20]. Using these combined models the framework solves for the self-consistent surface resistance, the temperatures of the quasiparticles, lattice, outer cavity wall, and helium bath, and the power per unit area dissipated and transmitted thermally through the system: starting with a range of T_{lattice} guesses (T_{in}), the framework calculates the RF power dissipated and from this the value of T_{lattice} consistent with this power flow based on the thermal model (T_{out}). The self-consistent solution is where $T_{\text{in}} = T_{\text{out}}$.

Figures 7–9 demonstrate example inputs and intermediate results of calculations using the framework. Figure 10 shows the final results of these calculations for two different underlying positive Q-slope models; these results were previously shown in [21].

In order to complete this thermal model framework, we are working with our collaborators using DFT to establish a theoretical model of the electron-phonon heat transfer coefficient Y as a function of impurity concentration. As described in [13, 21], this work has yielded positive early results in approximating the electron mean free path for niobium with varying impurity concentrations, calculations that agree well with experimental results. This is promising, suggesting that the DFT calculations may indeed lead to a full theoretical model of Y. Until then, we plan to treat Y as a free parameter in our simulations, either as a fitting variable or as an empirical parameter based on our model presented in [3].

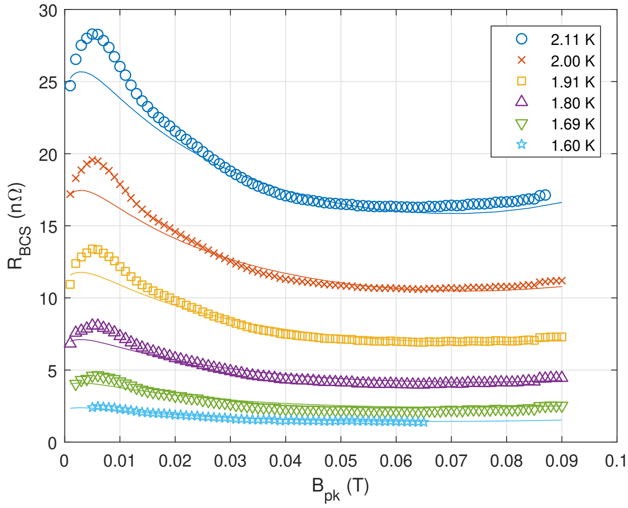

Figure 6: BCS surface resistance for a 96-hour nitrogen-infused 1.3 GHz cavity, with theoretical fits using the Gurevich positive Q-slope model, our electron overheating model, and our new thermal model. Previously published in [16].

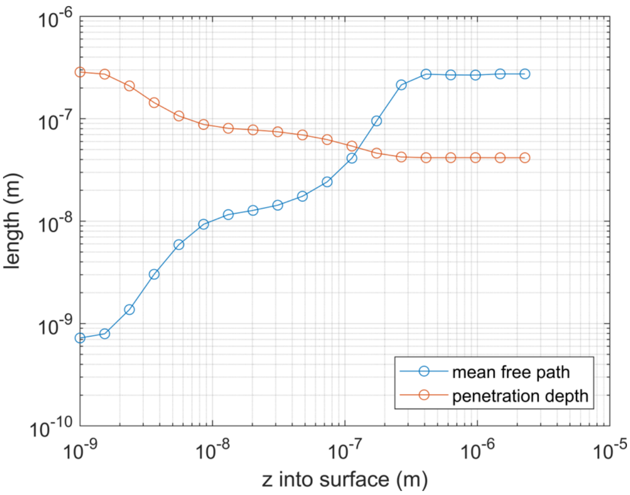

Figure 7: Example field-dependent material parameter input to thermal model framework.

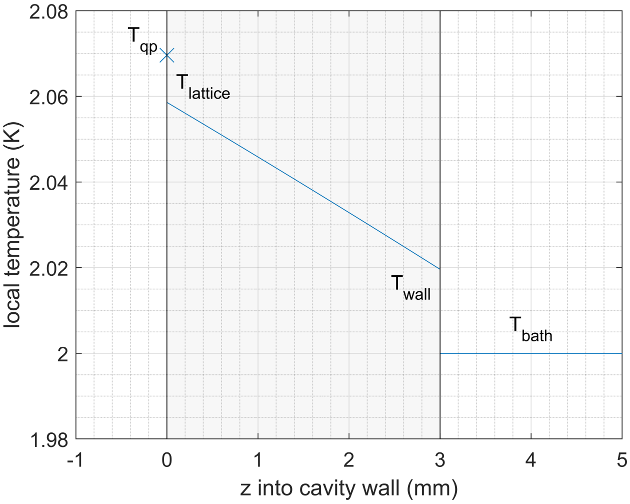

Figure 8: Example calculation results for the niobium-helium thermal system with quasiparticle overheating.

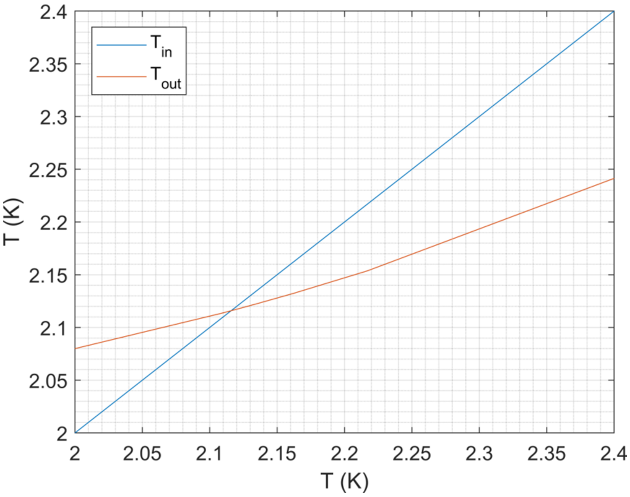

Figure 9: Example calculation results for the self-consistent power flow solution for the thermal model and surface resistance model.

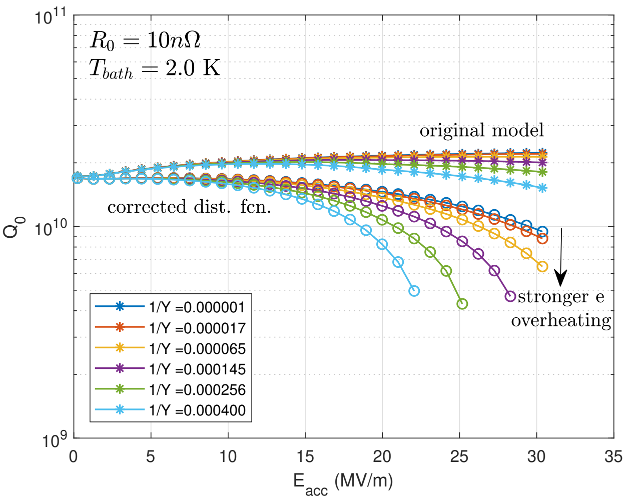

Figure 10: Example simulation output for a 1.3 GHz N-doped cavity, using the original Gurevich model of R_{BCS} as well as our version with corrected distribution function. Y is the electron-phonon heat transfer coefficient, here treated as a free parameter. Previously shown in [21].

ASSESSMENT OF ANTI-Q-SLOPE THEORIES

As previously reported [21], we have worked recently with our collaborators in the Center for Bright Beams to assess existing theories of the positive Q-slope and potentially develop alternative models. We present the results of our consideration for what we consider to be the three most relevant extant theories.

While we have previously found good agreement between the predictions of a 2014 theory involving surface-current-related "smearing" of the quasiparticle density of states as a method of reducing the BCS surface resistance with increasing surface field strength [3, 19], more recent experimental results at alternative frequencies greater or less than 1.3 GHz do not agree well with the theory [15, 22]. Moreover, this model predicts the opposite dependence of the positive Q-slope effect on cavity frequency: experiment shows an increasing strength of the field-dependent decrease in R_{BCS} [18], while calculations using the model predict the opposite dependence [23]. Deeper investigation of this theory found a potential physics error in an unjustified assumption about the quasiparticle distribution function which does not take into account additional quasiparticles driven out of equilibrium by the RF field. When replacing this distribution function with a simple model using detailed balance, the positive Q-slope prediction is replaced with a uniformly decreasing intrinsic quality factor. Figure 10 shows the results of calculations using the original model and the replacement distribution function, calculated with our new thermal model framework. We do note that this correction does not affect the "weak RF" case in the Gurevich model, where the relative strength of the RF field is too low to significantly change the quasiparticle distribution function. This means that other aspects of the model will be able to be tested with our forthcoming DC field dependence cavity [24].

We also considered a recent theory of a field-dependent decrease in the superconducting surface resistance that relies on a non-thermal quasiparticle distribution function to produce the field-dependent effects [21, 25]. Experimental results on stripline resonators have been reported to agree with the theory [26]. The field-dependent decrease arises from quantum mechanical perturbations in the distribution function that are most dramatic when $\hbar\omega \sim k_{\mathrm{B}}T$. Unfortunately, this condition is not met in doped niobium SRF cavities, where in energy units the temperature is nearly a factor of 50 greater than the frequency. Further, the field levels observed in the experiment are also substantially lower than those typically seen in SRF cavities with positive Q-slope. On top of this, the model makes no link to cavity doping or other surface treatments and how these treatments might bring about the field-dependent decrease in R_{BCS}. Because of these considerations, we find that this model may not be appropriate for describing the positive Q-slope observed in SRF cavities.

A third theory that we studied, in our understanding, considers nitrogen-doped niobium as a disordered composite of small pockets of material with poor superconducting properties (like T_{c}, H_{c}, or Δ); as the strength of the RF field increases, these pockets transition to the normal-conducting state and become proximity-coupled due to their small size [27]. This decreases the overall quasiparticle conductivity in a manner dependent on both field strength and frequency; in turn this decrease lowers the BCS surface resistance. The paper describing the model finds good agreement between the theory and recent SRF experiments exhibiting positive Q-slope, especially as it varies with frequency. However, our calculations using the model find that its prediction of the effects of the electron mean free path on the strength of the positive Q-slope are in strong disagreement with experiment: the model predicts that cleaner cavities (*i.e.* with longer electron mean free path) should exhibit stronger positive Q-slopes; this is in direct contradiction with experimental observations [3]. We also find that the model contains many finely tuned parameters and some physics assumptions that are not well justified; these findings are not conclusive but suggest that this theory is not appropriate for describing the positive Q-slope.

Our conclusions from this study of positive Q-slope models is that, indeed, no satisfying theory yet exists. The existing models are either not applicable in our estimation to the SRF cavity regimes where positive Q-slope has been observed or rely on potentially flawed calculations. Moving forward, our work in the Center for Bright Beams will look to develop a new model of the field-dependent surface resistance, with an eye towards using Floquet states in a DFT framework to describe the periodically driven quasiparticle system in SRF cavities.

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CONCLUSIONS

We have performed systematic studies on nitrogen-infused SRF cavities, finding no strong dependence in the positive Q-slope behavior on doping time but a correlation between chemical removal of surface spikes in nitrogen concentration and the presence or lack of the positive Q-slope. These findings point towards nitrogen above other impurities present in the RF surface as the cause of the positive Q-slope in infused cavities, either by fundamentally causing R_{BCS} to decrease with increasing RF field strength or by removing surface effects that counterbalance an intrinsic anti-Q-slope in niobium. We have studied the field-dependent BCS surface resistance in a 2.6 GHz niobium cavity under various treatment protocols, finding the best overall performance and strongest positive Q-slope in the 2/6-doped test. We have developed a full thermal model of SRF cavities which calculates the field-dependent surface resistance as influenced by the overheating of quasiparticles on the RF surface. Finally, we considered existing models of the positive Q-slope, finding that the current models are either incorrect or not relevant to the conditions under which the positive Q-slope has been observed. Future work will look towards developing a new model of the field-dependent surface resistance using Floquet theory.

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