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of the RF surface resistance of nitrogen-doped
niobium SRF cavities with respect to existing
theoretical models*

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AN ANALYSIS OF THE TEMPERATURE AND FIELD DEPENDENCE OF THE RF SURFACE RESISTANCE OF NITROGEN-DOPED NIOBIUM SRF CAVITIES WITH RESPECT TO EXISTING THEORETICAL MODELS*

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Abstract

Recent progress with the reduction of rf surface resistance (R_s) of niobium SRF cavities via the use of high temperature surface doping by nitrogen has opened a new regime for energy efficient accelerator applications. For particular doping conditions one observes dramatic decreases in R_s with increasing surface magnetic fields. The observed variations as a function of temperature may be analyzed in the context of recent theoretical treatments in hopes of gaining insight into the underlying beneficial mechanism of the nitrogen treatment. Systematic data sets of Q_0 vs. E_{acc} vs. temperature acquired during the high Q_0 R&D work of the past year will be compared with theoretical model predictions.

INTRODUCTION

For many years the SRF community has informally assumed that the best performance to be obtained from SRF cavities was an rf surface with linear dissipation properties over the full dynamic range up until magnetic flux penetration phenomenon begins to create fundamental field limitations. The “best possible” performance was described by BCS losses produced by quasi-particle scattering according to the Mattis-Bardeen theory[1] for the surface resistance of a superconductor, and most conveniently calculated using the SRIMP code constructed by Halbritter[2] in the 1970’s.

This changed in 2012-2014 when reports of experimental observation of unprecedentedly low $R_s(B)$ appeared with distinctly *decreasing* dependence on surface magnetic field appeared[3-5], as did first theoretical predictions of such a phenomenon[6, 7]. A fresh attempt at developing a theoretical expectation for $R_s(B,T)$ was a simplified theory that sought to extend Mattis-Bardeen theory to higher fields[6]. This analysis addressed rf current induced pair breaking, but principally noted the effects of anisotropies in the distribution function of quasiparticles due to significant current flow, in the limit of thermal equilibrium. This latter constraint implies that the quasiparticle inelastic scattering time is short compared with the rf cycle. The anisotropic Fermi surface effectively induces a broadening of the peaks in the quasiparticle density of states without significant modification of the gap. This simplified theory, while limited in scope, perhaps provides some conceptual

guidance for understanding the rather surprising decrease in $R_s(B)$, which had previously been analyzed only for very thin superconducting films[8].

A more rigorous and general theoretical treatment has recently been proposed by Gurevich[9]. This treatment allows for non-equilibrium distribution functions and overheated quasiparticles.

It is interesting to examine the correspondence of these theoretical treatments with the field and temperature performance of some cavities that have received “nitrogen doping” to see if one may thereby gain insight into both mechanism and optimization strategies for best cavity performance.

DOPING NB

Evidence to date indicates that the beneficially “alloyed” N in Nb by thermal diffusion takes up residence interstitially with a concentration in the surface of ~0.1%[10, 11]. Analyses indicate that such N would first populate vacancies, then octahedral symmetry sites[12]. An expected effect of the interstitial nitrogen is lowering of the electron mean free path, l . It has been well recognized that a minimum in R_s with $l \sim 35$ nm is both experimentally observed and predicted by Mattis-Bardeen theory. Temperature dependent properties of the high- Q N-doped cavities have been consistent with $l = 7-12$ nm[13], significantly lower than would be the predicted optimum.

It has also been suggested that a beneficial role of the interstitial N is its ability to stabilize interstitial H, so that NbH nano-precipitates do not form. Previous work established that each N atom in an octahedral site will stabilize one H in a tetrahedral site[12, 14-16]. Creating conditions at elevated temperatures which give energetic preference to populating vacancies with N rather than O or multiple H, and adequate additional N distributed in octahedral sites to bind the available H during cavity cooldown (H diffusion length estimated at ~ 30 μm [17]) thus inhibiting the formation of Nb hydrides. Similar trapping of H by substitutional Ti diffused to fill vacancies is attributed to the comparable high- Q cavities realized by that technique[5, 18].

It is interesting to note that a very similar phenomenon was reported by Ballantini *et al.* in 1999[19], with dry oxidation of a niobium cavity after high temperature heat treatment. One may speculate that perhaps that also populated the near surface in a way which effectively inhibited the formation of particular hydride forms within the rf surface. The decrease of $R_s(B)$ to ~ 70 mT was

observed even in the early explorations with UHV-baked X-band cavities given dry oxidation at SLAC by Wilson *et al.*[20]

While the role of hydrides in the occurrence of “high-field Q drop” is becoming increasingly substantiated[17], it is not yet clear how one may characterize the influence of hydrogen on the lower field SRF surface resistance of Nb. Whether its presence is key to the occurrence of “mid-field Q slope” in “un-doped” Nb and its absence allows the occurrence of the “extended Q-rise”, or whether its bound presence is in some way instrumental in creating such favorable conditions is yet unknown.

ANALYZING OBSERVED $R_S(B_{pk}, T)$

In the context of cavity process development to exploit the “nitrogen-doping” high-Q phenomenon for the use by the LCLS-II project, several single cell and 9-cell 1.3 GHz cavities were systematically tested over a range of temperatures. To interpret the performance of variously prepared cavities, it is helpful to separate temperature-dependent contributions to the effective rf surface resistance, $R_{s-eff} = G/Q_0$, where G is a geometry factor, from temperature-independent ones. For each cavity test, the data set was fitted using the function:

$$R_{s-eff} = R_{resid}(B_{pk}) + R_{BCS}(B_{pk}, T) [= \frac{A(B_{pk})}{T} e^{-\frac{U}{T}}]$$

We take the simplifying assumption that the effective gap/ T_c is constant, with $U = 17.02$ K, for $2\Delta = 1.84$ meV. This form is general enough to accommodate a thermally activated quasiparticle distribution that is influenced by the presence of super-current flow required to support B_{pk} such that the gap peaks in the quasiparticle spectrum may broaden with B but with fixed Δ . We also take the assumption that local heat flux remains low enough over the range of measurements that the superfluid He bath temperature adequately reflects the active rf surface temperature.

We illustrate our analysis approach using measurements made on the single cell cavity RDT-5, a standard 1.3 GHz TESLA-style fine-grain niobium cavity. It received a sequence of three preparations and tests. It was first prepared with a typical BCP etch, then given an incremental 4 μm electropolish, then subjected to a nitrogen-doping protocol with 180 minute, 800°C vacuum heat treat, 2-minute exposure to ~30 mTorr N_2 @ 800°C, followed by 6 minute 800°C vacuum “anneal”, and 4 μm electropolish. (For such a protocol, we use the short-hand notation: 2N/6+4.) After each chemical preparation the cavity was high-pressure rinsed with ultrapure water, evacuated and cryogenically tested at multiple temperatures. (The standard Q_0 vs. field plot for the last test is presented in Figure 1.) Each data set was then fitted as a whole to the function above. The resulting fit parameters $R_{resid}(B_{pk})$ and $A(B_{pk})$ are shown in Figure 2 with the resulting fitted function $R_{BCS}(B_{pk}, 2.0K)$. We

plot $A(B_{pk})$ in semi-logarithmic form following the logic of [7]. The equivalent fitted function $Q_0(B_{pk}, T_i)$ for the doped case is also plotted in Figure 1 for illustration.

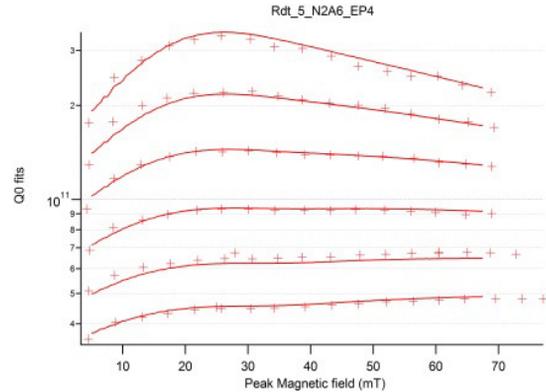


Figure 1: Q_0 vs. B_{pk} for RDT-5 after 2N/6+4 doping protocol at 2.0, 1.9, 1.8, 1.7, 1.6, and 1.5 K, together with the resulting fitted function.

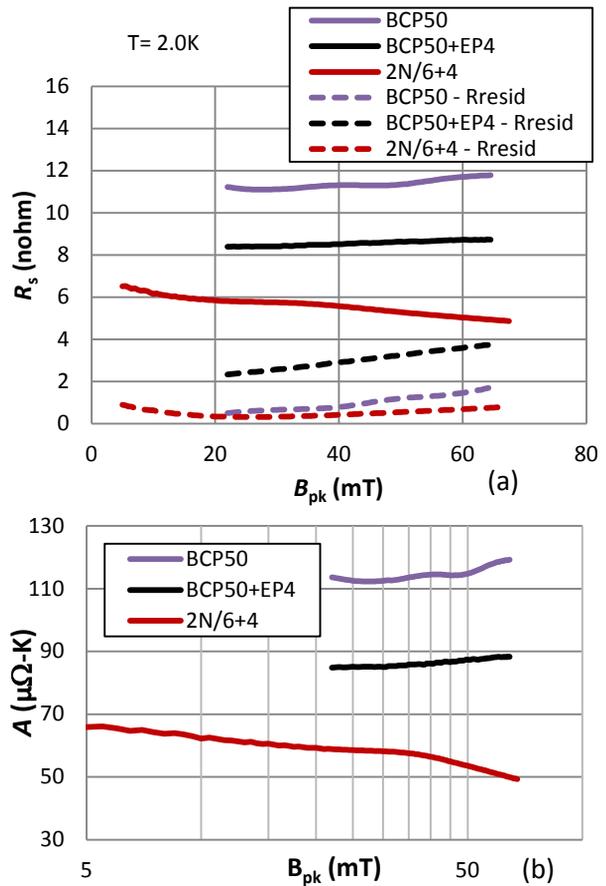


Figure 2: Fitted separation of “BCS” and “residual” components of $R_s(B_{pk})$ for three cavity RDT-5 preps.

The same analysis has thus far been applied to 15 different single-cell cavity tests, each with a different surface preparation and 7 different 9-cell cavity tests, four of which had nominally the same surface preparation, that being the one presently selected for use on the LCLS-II cavities[21-23]. We summarize here the results and some of the preliminary interpretations.

In support of the high Q R&D program of the LCLS-II project there was an initial exploration of protocol parameters used for nitrogen doping of cavities to obtain high Q s. A systematic analysis correlating $R_s(B_{pk})$ and cavity quench fields with the protocol parameters was begun[24]. This work is yet on-going, but we note here some example cases that illustrate in particular how $A(B_{pk})$ changes with different surface conditions. We anticipate such analyses may yield further material insights and guidance for performance optimization.

Of six 9-cell cavities processed at JLab, two were processed with alternative protocols, and the resulting performance test data was given the same analysis as four cavities that received the current standard 2N/6+5 protocol. Figure 3 shows the fitted performance of AES031 which was tested after 20N/30+16 and 20N/30+26 treatments, and AES034, which received a 2N/30+10 protocol. For comparison, we also include the average fitted performance obtained from tests on the cavities using the 2N/6+5 protocol and the fitted performance from a single-cell cavity RDT-15 that received a 20N/50+15 treatment protocol.

For comparison, in Figure 3 we also show the predictions of $R_{BCS}(B_{pk}, 2.0K)$ and the corresponding $A(B_{pk})$ from the Xiao theory, using a parameter set with “textbook” values for niobium $\Delta_0/kT_c(0)=1.85$, $T_c(0) = 9.25$ K, $\xi_0 = 38$ nm, $\lambda_L(0) = 39$ nm [25-27], and $l = 9$ nm at 1.3 GHz.

COMMENTS

- A significant variety of doping protocol parameter sets yield $R_{s-eff} < 10$ n Ω at $B_{pk} \sim 60$ mT.
- Although statistics are yet quite limited, there appear to be systematic variations of $A(B_{pk})$ with variations of the surface doping with nitrogen, particularly in the $B_{pk} = 20$ -70 mT range.
- We offer no explanation for the consistently-encountered low-field decrease of $R_{resid}(B_{pk})$.
- Comparison of the 2N/6+5 and 2N/30+10 results show significantly increased R_{BCS} with $B_{pk} > 20$ mT.
- The 10 μ m incremental EP on AES031 from 20N/30+16 to 20N/30+26 very significantly flattened the R_{BCS} contribution, but left it still substantially lower than what is obtained by classical BCP or EP treatments.
- The lowest results for $R_{BCS}(B_{pk}, T)$ from conditions sampled so far appear to be similar to the predictions of Xiao’s dynamic quasiparticle thermal equilibrium model in the $B_{pk} = 20$ -70 mT range.

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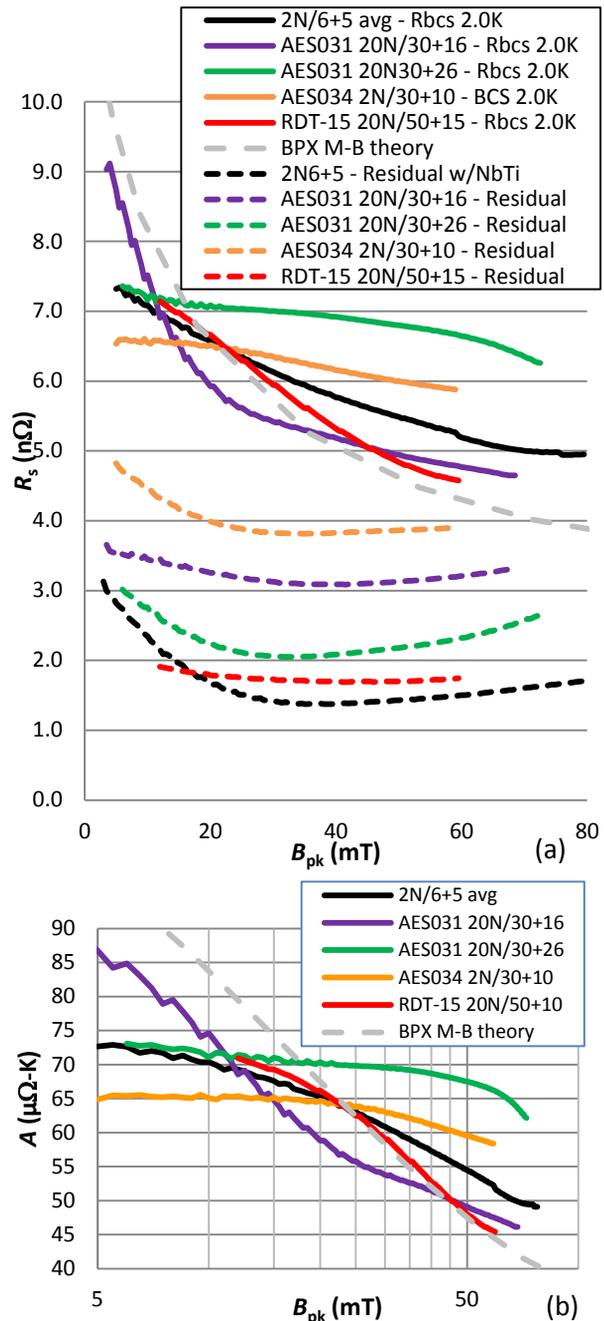


Figure 3: Fitted separation of “BCS” and “residual” components of $R_s(B_{pk})$ for cavities prepared with various doping protocols. (a) $R_{resid}(B_{pk})$ and $R_{BCS}(B_{pk}, 2.0K)$, (b) fitted prefactor $A(B_{pk})$.

REFERENCES

- [1] D. C. Mattis and J. Bardeen, Physical Review 111, 412 (1958).
- [2] J. Halbritter, Zeitschrift für Physik 238, 466 (1970).

- [3] P. Dhakal, et al., in International Particle Accelerator Conference 2012 (New Orleans, Louisiana, 2012), p. 2426.
- [4] A. Grassellino, et al., Superconductor Science and Technology 26, 102001 (2013).
- [5] P. Dhakal, et al., Physical Review Special Topics - Accelerators and Beams 16, 042001 (2013).
- [6] B. P. Xiao, et al., Physica C: Superconductivity 490, 26 (2013).
- [7] G. Ciovati, et al., Applied Physics Letters 104, 092601 (2014).
- [8] S. Sridhar and J. E. Mercereau, Physical Review B 34, 203 (1986).
- [9] A. Gurevich, Phys. Rev. Lett. 113, 087001 (2014).
- [10] Y. Trenikhina, et al., in SRF2013 (Paris, France, 2013), p. 583.
- [11] D. Gonnella, et al., in 27th Linear Accelerator Conference (Geneva, Switzerland, 2014).
- [12] T. H. Metzger, et al., Journal of Physics F: Metal Physics 15, 779 (1985).
- [13] D. Gonnella and M. Liepe, in International Particle Accelerator Conference 2014 (Dresden, Germany, 2014), p. WEPRI064.
- [14] D. C. Ford, et al., Superconductor Science and Technology 26, 105003 (2013).
- [15] D. C. Ford, et al., Superconductor Science and Technology 26, 095002 (2013).
- [16] G. Pfeiffer and H. Wipf, Journal of Physics F: Metal Physics 6, 167 (1976).
- [17] A. Romanenko, et al., Superconductor Science and Technology 26, 035003 (2013).
- [18] D. Richter, et al., Physical Review B 27, 6227 (1983).
- [19] R. Ballantini, et al., in 9th International Workshop on RF Superconductivity, edited by B. Rusnak (Santa Fe, NM, 1999), p. 211.
- [20] P. B. Wilson, et al., in 1973 Particle Accelerator Conference (IEEE, 1973), p. 104
- [21] M. Liepe, et al., in International Particle Accelerator Conference 2014 (Dresden, Germany, 2014), p. WEPRI062.
- [22] A. Grassellino, in International Particle Accelerator Conference 2015 (Richmond, Virginia, 2015), p. MOYGB2.
- [23] A. D. Palczewski, et al., in International Particle Accelerator Conference 2015 (Richmond, Virginia, 2015), p. WEPWI019.
- [24] A. D. Palczewski, et al., in 27th Linear Accelerator Conference (Geneva, Switzerland, 2014).
- [25] V. Novotny and P. P. M. Meincke, J. Low Temp. Phys. 18, 147 (1975).
- [26] C. Kittel, Introduction to Solid State Physics (Wiley, 1996).
- [27] R. D. Parks, Superconductivity (Dekker, New York, 1969).