

# INVESTIGATION OF FREQUENCY BEHAVIOR NEAR $T_c$ OF NIOBIUM SUPERCONDUCTING RADIO-FREQUENCY CAVITIES\*

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## Abstract

This paper will present a systematic investigation of the resonant frequency behaviour of niobium SRF cavities subject to different surface processing (nitrogen doping, nitrogen infusion, 120°C bake, EP, etc.) near the critical transition temperature. We find features occurring in frequency versus temperature (FvsT) data near  $T_c$  that seem to vary with surface processing. Emphasis is placed on one of the observed features: a dip in the superconducting resonant frequency below the normal conducting value which is prominent in nitrogen doped cavities and appears to be a signature of nitrogen doping. This gives further insights on the mechanisms responsible for the large increase in performance of cavities subject to this surface treatment. The magnitude of this dip in frequency is studied and related to possible physical parameters such as the concentration of impurities near the surface and the cavity design resonant frequency. A possible explanation for the meaning of this dip is discussed, namely, that it is a result of stronger coupling between electrons and phonons within the resonator.

## INTRODUCTION

Superconducting radio-frequency (SRF) cavities are one of the leading technologies in cost effective particle acceleration. They allow for high accelerating gradients while maintaining high quality factors. It has been shown that the performance of these cavities is strongly dependent on the near surface impurity structure due to the supercurrents that flow in this region. As such, preparation of the first couple hundreds of nanometers of the RF surface is of the utmost importance. The performance of cavities subject to various surface treatments treated and tested at FNAL are shown in Fig. 1. The surface treatments used to prepare the cavities shown in Fig. 1 are outlined in Table 1. Certain surface treatments allow for very high accelerating gradients such as the 120°C bake, nitrogen infusion, and 75/120°C bake [1-3]. Another surface treatment, such as nitrogen doping, allows for very high quality factors [4,5].

In addition to differences in performance, these surface treatments vary the resonant frequency as a function of the temperature of cavities. In addition, these surface treatments appear to influence interesting features that arise in frequency vs temperature (FvsT) data near the transition temperature ( $T_c$ ) of the cavity. This paper presents a systematic investigation of these features in FvsT data near  $T_c$ . Although five distinct features are presented, one feature in particular, a prominent dip in the superconducting

resonant frequency of the cavity below the normal conducting value that occurs just below the transition temperature, is emphasized.

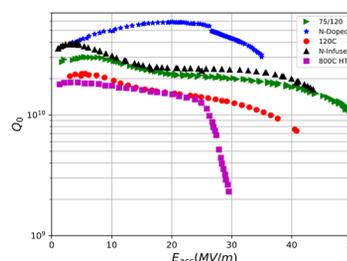


Figure 1: Quality factor vs accelerating gradient ( $Q_0$  vs  $E_{acc}$ ) data taken at 2 K for 1.3 GHz SRF cavities subject to the surface treatments outlined in Table 1.

## ZOOLOGY OF F vs T FEATURES

The resonant frequency of a cavity is measured at low fields with a network analyser as the cavity is warmed up through transition. The temperature is measured using Cernox RTD temperature sensors. As the cavity temperature increases, the resonant frequency of the cavity decreases due to the temperature dependence of the penetration depth, which increases with temperature. This is well described by the Gorter-Casimir two-fluid equation

$$\lambda(T) = \frac{\lambda_0}{\sqrt{1 - \left(\frac{T}{T_c}\right)^4}}, \quad (1)$$

where  $\lambda_0$  is the penetration depth at zero Kelvin. However, deviations from this equation have been observed in experimental data as it does not consider the effect of the superconducting gap [6]. It is understood that any model that considers a gap will show that the penetration depth will more rapidly approach its zero-temperature value than one that does not [7]. As such, the superconducting gap of a resonator will influence its penetration depth into the surface, which in turn varies the profile of the resonant frequency as a function of temperature. The model states that as the temperature approaches  $T_c$ , the penetration depth goes to infinity, causing a discontinuity from the normal conducting value, which is governed by the normal conducting skin depth. Experimental data of Nb SRF cavities shows that the resonant frequency of the cavity remains finite in this region. Not only is the frequency finite, cavities also display five unique and distinct features in this region. Figure 2 outlines the five unique features observed in FNAL F vs T data.

\* This work was supported by the United States Department of Energy, Offices of High Energy Physics and Basic Energy Sciences under Contracts No. DE-AC05-06OR23177

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Table 1: Outline of Several SRF Cavity Surface Treatments along with the Resulting Mean Free Path Estimate

Surface Treatment	N-Doping	N-Infusion	120C Bake	75/120C Bake	EP
	800 C x 3 hrs in UHV	120 C x 48 hr in 25 mTorr N	800 C x 3 hrs in UHV	800 C x 3 hrs in UHV	800 C x 3 hrs in UHV
	800 C x 2 min in 25 mTorr N		120C x 48 hrs in UHV	75 C x 4 hrs in UHV	EP
	800 C x 2min in UHV			120C x 48 hrs in UHV	
	5um EP				
<b>MFP (nm)</b>	60-300	~1-50	~1-20	~1-20	~1000

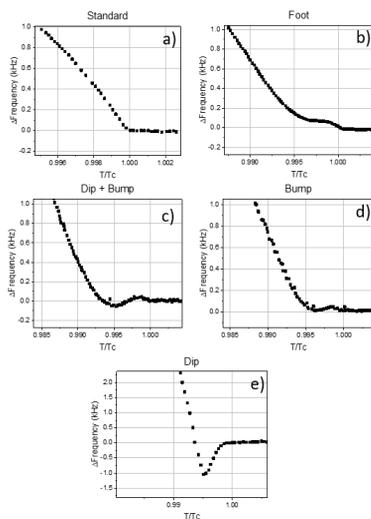


Figure 2: The five features observed in FNAL F vs T data. The normal conducting frequency is set to correspond to 0 Hz for each case.

The first feature is a lack thereof, in which there is a sharp transition from the superconducting (SCing) to the normal conducting (NCing) frequency, labelled “Standard”. The second feature is described as a foot, which has a small hump before going NCing. The third feature is a dip with a bump, indicating that the frequency goes slightly below the NCing value and then back above it, after which it decreases back to the NCing value. The fourth feature is quite similar and only has a small bump. Lastly, there is a prominent dip in the SCing frequency below the NCing value. An explanation for the origins of this dip will be discussed later. Although all five features are fundamentally interesting, this prominent dip in the frequency is the most striking. As such, a systematic investigation of this dip will now be discussed.

## INVESTIGATION OF DIP IN F vs T

### Signature of N Doping

To investigate when this dip in the resonant frequency below the normal conducting value occurs, a single cell 1.3 GHz TESLA shaped Nb SRF cavity was subject to the following surface treatments with a surface reset in between to compare same surface morphology; 75/120°C bake, nitrogen infusion, and nitrogen doping. The cavity resonant frequency was then recorded using a network analyser as the cavity was warmed up through transition. The results are outlined in Fig. 3. Note that each curve is corrected for pressure differences within the dewar. Although the inset on the lower left of Fig. 3 shows the expected differences in temperature dependence of the curves due to different impurity structure, it is striking to see that the three treatments produce three of the five observed features near  $T_c$  discussed in Fig. 2. This signifies that the surface preparation alone is the cause of these differences. The cavity post 75/120°C bake shows a foot near  $T_c$ . Nitrogen infusion, instead, gives a small dip with a bump. The cavity post nitrogen doping gives a prominent dip in the frequency vs temperature data. For all the data studied (48 sets), this prominent dip occurs 100% of the time in nitrogen doped cavities and appears to be a signature of it. Using the 48 sets of data studied, Table 2 outlines the number of instances in which the surface treatments discussed in Table 1 displayed the features shown in Fig. 2. All 27 studied nitrogen doped cavities displayed a prominent dip in the frequency just below the transition. Note that although there exists one instance of a dip in a cavity post nitrogen infusion and one instance of a dip in a cavity post 75/120°C bake, both of these dips were quite small. The effect of the different cavity parameters on this dip will now be explored.

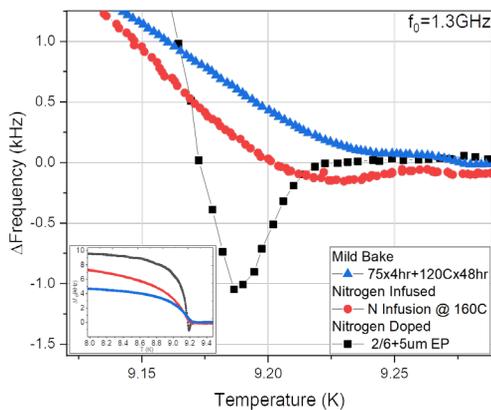


Figure 3: Frequency vs temperature data for a 1.3 GHz single cell cavity subject to three different surface treatments. The inset shows a zoomed-out plot of the three profiles.

Table 2: Occurrences of Features near  $T_c$  Observed for Various Surface Treatments. Blank entries signify that there were no instances of that feature for that surface treatment.

	N Doped	N Infused	75/120 C	120C	EP
Dip	27	1	1		
Foot		1	4		
Bump			1		1
Dip + Bump		2		1	
Standard		1	4	1	3

### Effect of Cavity Design Frequency on the Dip

To study the effect of cavity design frequency on the dip, four cavities of different resonant frequencies were treated to the same 2/6 nitrogen doping surface treatment. The four resonant frequencies used were: 650 MHz, 1.3 GHz, 2.6 GHz, and 3.9 GHz. The resulting frequency vs temperature profiles are shown in Fig. 4. It is observed that the higher the design frequency of the cavity, the larger the dip. In fact, the figure on the right in Fig. 4 shows that the dip depth appears to increase linearly with the design frequency. One simplified possible model to explain this linear relationship could stem from the frequency dependence of the NCing skin depth. Because it goes as the square root of the inverse of frequency, higher resonant frequency cavities will experience a shorter skin depth. As such, the discontinuity between the normal conducting skin depth and the penetration depth that occurs at the superconducting transition may increase with frequency, increasing the depth of the observed dip because the skin depth must effectively “catch up” to the penetration depth.

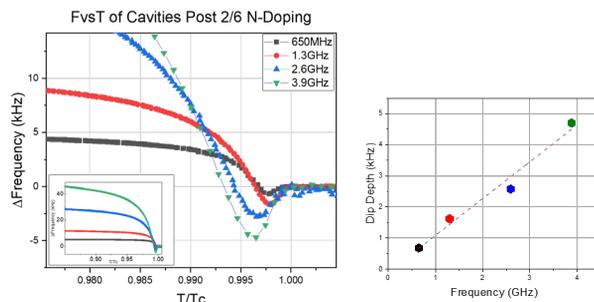


Figure 4: (Left) Frequency vs temperature data for cavities of different resonant frequencies subject to 2/6 N-doping. The lower left inset gives a zoomed-out picture of the FvsT profiles. (Right) Comparison of the dip depth versus the resonant frequency.

### Effect of Nitrogen Mean Free Path on Dip

To study the effect that nitrogen concentration might have on the dip, one single cell 1.3 GHz TESLA shaped Nb SRF cavity, TE1RI003, was treated to a variant of a nitrogen doping surface treatment (3/60 N-Doping). The cavity was then tested after sequential removal of the surface via EP. This sequential removal decreases the concentration of interstitial nitrogen that exists within the RF layer. The cavity resonant frequency through warm up and the  $Q_0$  vs  $E_{acc}$  curves after each step of removal are shown in Fig. 5.

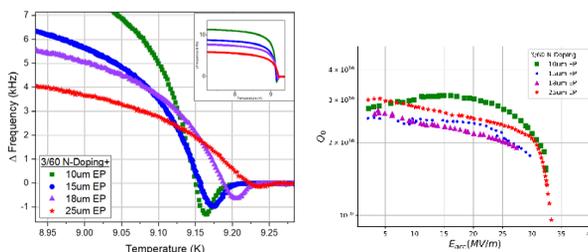


Figure 5 (Left) Effect of nitrogen concentration on the dip in FvsT data for cavity TE1RI003. The inset shows a zoomed-out picture of the profiles. (Right) A plot of  $Q_0$  vs  $E_{acc}$  after various amounts of removal all taken at a temperature of 2 K.

After a 10  $\mu\text{m}$  removal of the RF surface, TE1RI003 had a prominent dip in frequency just below  $T_c$ . The  $Q_0$  vs  $E_{acc}$  curve for this test shows a typical N-doped profile, with the existence of the anti-Q slope phenomenon [4]. After removing an additional 5  $\mu\text{m}$ , giving a total of 15  $\mu\text{m}$  of removal, the dip depth decreased while the transition temperature of the cavity increased. Compared to the previous test, the  $Q_0$  of the cavity was not as high; it also has little to no anti-Q slope. Removing another 3  $\mu\text{m}$  of the surface causes the magnitude of the dip to decrease further and the transition temperature to increase slightly. It should be noted that the  $Q_0$  vs  $E_{acc}$  curve of the cavity after a total of 18  $\mu\text{m}$  of removal of the surface showed the onset of high field Q-slope [8]. However, data was only taken to 27 MV/m to avoid quenching the cavity at 2 K for this test. Further measurements at  $\sim 1.5$  K showed that the cavity reached accelerating gradients up to 36.5 MV/m and was power limited. The final test shown in Fig. 5 is after a total

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removal 25  $\mu\text{m}$  of the RF surface. The magnitude of the dip is now very small, with a depth of about  $\sim 100$  Hz, which is just above the noise level. In addition, the  $T_c$  increases further, up to a value of  $\sim 9.26$  K. In summary, as the concentration of nitrogen gets lower, the doping effect gradually diminishes in: 1) the temperature dependence of the penetration depth, 2) the depth of the dip, 3) the  $Q_0$  vs  $E_{\text{acc}}$  curves.

Some trends with mean free path (MFP) near the surface can be observed from this sequential removal study. The mean free path of the cavity after each step of removal was extracted using SRIMP [9]. The MFP values obtained were then plotted against the depth of the dip in frequency, measured relative to the normal conducting value, and against  $T_c$ . The results are shown in Fig. 6.

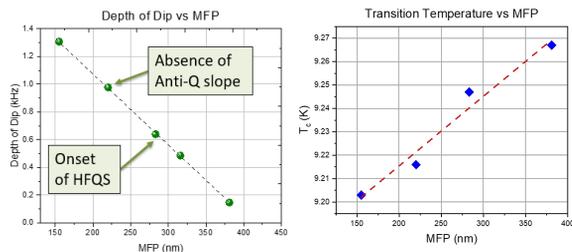


Figure 6: Effect of MFP on (Left) dip depth (Right) transition temperature.

As can be observed above, dip depth appears to vary linearly with the mean free path of the cavity. A mean free path of 220 nm for the cavity corresponds to an absence in the anti-Q slope, as observed in Fig. 5 after 15  $\mu\text{m}$  of removal. A mean free path of 283 nm is obtained when the cavity had the onset of high field Q slope. Continuing the linear trend, the dip is expected to disappear entirely at 400 nm.

The transition temperature also appears to have a linear relationship with the mean free path of the cavity. From the plot, it is seen that the effect of nitrogen interstitial on  $T_c$  is small, varying only 60 mK for MFP values that range from 150 nm to 380 nm.

## DISCUSSION

### Origin of Dip

Although this dip in the resonant frequency below the normal conducting value just below the superconducting transition have been discussed elsewhere [10,11], a short summary of this phenomenon is presented here followed by possible implications. Because the normal conducting skin depth decreases with temperature (due to the longer scattering times of electrons in the Drude model), this skin depth can be pushed into the anomalous regime, making non-local effects important. As a result, Mattis-Bardeen (MB) theory can be used to gain insight on the mechanisms responsible for this dip in the frequency. In MB theory [12], the conductivity of a superconductor is described by

$$\sigma_s = \sigma_1 + i\sigma_2, \quad (2)$$

where  $\sigma_1$  and  $\sigma_2$  are conductivities of quasi-particles and Cooper pairs. The surface impedance of the superconductor normalized to the NCing conductivity  $\sigma_N$  is

$$\frac{Z_S}{Z_N} = \left( \frac{\sigma_s}{\sigma_N} \right)^{-1/2}. \quad (3)$$

For there to exist a dip in the frequency below the NCing value, the penetration depth must be longer than the NCing skin depth  $\delta_N$ . For this to occur, the penetration depth must increase relative to the value of  $\delta_N$  as the temperature decreases through transition. Using (3), for values where of  $(\sigma_s - \sigma_N)/\sigma_N \ll 1$  and if the superconductor is in the dirty limit, Varmazis *et al.* obtain the following expression for the change in the superconducting penetration depth very close to and below  $T_c$ :

$$\Delta\lambda = \frac{\delta_N}{4} \left( \frac{\sigma_2}{\sigma_N} - \Delta \left( \frac{\sigma_1}{\sigma_N} \right) \right), \quad (4)$$

where the second term in the parentheses denote a change in the quasi-particle conductivity. This states that for there to exist a dip in the resonant frequency of a cavity close to and below  $T_c$ , the increase in the conductivity of the super-electrons must be larger than the increase in the conductivity of the quasi-particles. Thus, the superelectrons in a superconductor are responsible for the existence of a dip in the frequency.

There are two cases that will determine the existence of this dip in a superconductor which involve the mean free path  $l$ , the extreme anomalous skin depth  $\delta_{N\infty}$ , and coherence length  $\xi_0$ :

1.  $\delta_{N\infty} < \xi_0$ : dip is likely to exist regardless of the value of  $l$
2.  $\delta_{N\infty} > \xi_0$ : a dip may exist if the mean free path is sufficiently low enough.

In the case of niobium, case 2 is applicable. This means that the mean free path of the cavity will play a role in determining if the phenomenon in question will occur.

This last point is important in helping to understand why only nitrogen doped cavities seem to experience this dip. As outlined in Table 1, electropolished cavities have very long mean free paths ( $\sim 1000$  nm). As such, this fails the case 2 condition laid out above, implying that there should be no dip in the frequency near  $T_c$ . Indeed, Table 2 shows that electropolished cavities display only one distinct feature in frequency near  $T_c$  in addition to the otherwise expected sharp transition to a plateau in the normal conducting regime. In addition, cavities subject to 120 C bake, 75/120 C bake, and nitrogen infusion tend to have a very short mean free path at the surface; however, this is true very close to the surface. Further from the surface the mean free paths become very long, failing again the case 2 condition. This, along with the fact that cavities subject to these surface treatments do not display a prominent dip, hint that perhaps the immediate RF surface is not the only

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region of importance. To contrast this, nitrogen doped cavities have interstitial nitrogen all throughout the RF layer, with mean free paths that are significantly shorter than that of electropolished cavities, as shown in Table 2. As a result, cavities subject to this surface treatment are expected to, and do indeed, show a dip in the frequency below the normal conducting value close to and below  $T_c$ .

### Possible Implications: Strong Coupling

In addition to being fundamentally interesting, the dip in the frequency below the normal conducting value near  $T_c$  might be indicative of a difference in superconducting parameters of resonant frequency cavities such as the superconducting gap. In BCS theory, when plotted against the temperature, there exists a peak in the quasi-particle conductivity,  $\sigma_1$ [13]. This peak is called the coherence peak and occurs at temperatures of  $\sim 0.85 T/T_c$ . This coherence peak comes from the increased conductivity of quasi-particles that arises from the breaking of Cooper pairs by thermally activated phonons [14-16]. To couple with the above studies of the effect of nitrogen concentration and frequency on the dip, the effect of the superconducting gap and resonant frequency on the coherence peak are calculated and shown in Fig. 6. Note that by naïvely using BCS theory and leaving the gap as a free parameter, many of the elements of strong coupling are ignored; this work however, serves as a mere introduction to present possible paths forward.

Figure 6 shows that the resonant frequency of a cavity strongly controls the height of the coherence peak. Higher frequencies give lower coherence peak heights. Note that the resonant frequency also weakly alters the width, which tends to be centred around  $\sim 7$  K. In contrast, the superconducting gap  $\Delta$  appears to strongly control the width of the coherence peak, with higher values of the gap yielding thinner widths of the coherence peak. Note that the gap has little effect on the height of the peak.

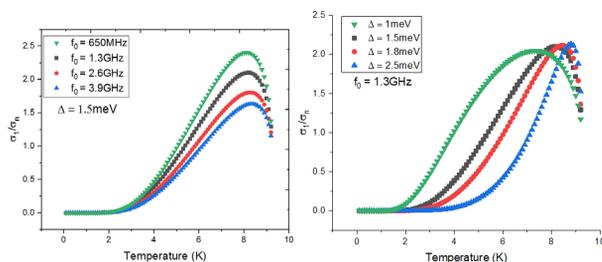


Figure 6: (Left) BCS calculations of the quasi-particle conductivity for various resonant frequencies of cavities with a superconducting gap of 1.5 meV. (Right) BCS calculations of the quasi-particle conductivity for various superconducting gaps for a resonant frequency of 1.3 GHz. Both figures are done for niobium with a  $T_c$  of 9.25 K.

One can't help but notice the similarities between the plots in Fig. 6 and the left figures in Fig. 4 and Fig. 5; perhaps the dip in the resonant frequency might have some connection to the coherence peak. To investigate this possibility, the data set taken on a Nb sample within a 60 GHz

resonator that is presented in Fig. 3 of Klein *et al.* [14] is used. The Nb FvsT data this paper presents has this dip in the frequency just below  $T_c$ , as is shown in black in Fig. 7. Note that Klein *et al.* report preparing the Nb samples with nitric acid. Using the method laid out in the paper, the given  $Q_0$ vsT and FvsT data are used to calculate the quasi-particle conductivity. This calculation of the conductivity is shown in Fig. 8 and gives the coherence peak shown in Fig. 5 of Klein *et al.* This peak is recalculated, but this time with a generated data set for the FvsT curve so that it more closely resembles the FvsT data obtained for the 75/120 C bake shown in Fig. 3 (i.e., the dip no longer exists and the temperature dependence of the penetration depth changes). With this fictitious 75/120 C bake data, the width of the coherence peak is drastically reduced, shown in red in Fig. 7. As learned from Fig. 6, thinner coherence peaks indicate stronger superconducting gaps, meaning that the absence of the dip indicates stronger coupling. However, this is just a fictitious set of data used to explain the current theory; in changing the FvsT profile, the  $Q_0$ vsT profile that is also necessary to calculate the conductivity was left untouched. It is possible that real data will show that the dip indicates stronger coupling between the electrons and phonons within the superconductor. Whatever the result, it is obvious that the dependence of the resonant frequency on the temperature is important in obtaining values of the superconducting gap of the resonator.

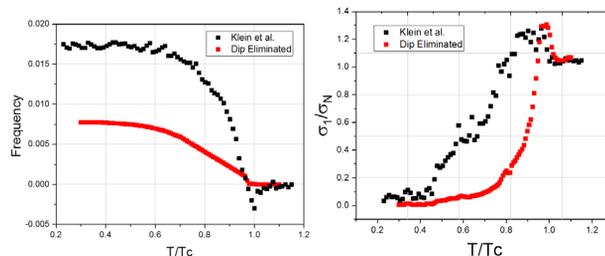


Figure 7: (Left) FvsT data taken from Fig. 3 in [14] is shown in black. In red is a generated set of data made to replicate the FvsT profile of a 75/120 C bake cavity. (Right) the quasi-particle conductivities calculated using the FvsT data shown in the left plot.

## CONCLUSION

Nitrogen doped cavities show a prominent dip in frequency below the normal conducting value just before the transition temperature. Larger concentrations of nitrogen as well as higher cavity design frequencies cause the depth of this dip to increase in magnitude. It is believed that this dip is indicative of a larger superconducting gap, which can be measured by the width of the coherence peak that occurs in the quasi-particle conductivity in BCS theory. Preliminary calculations show that the elimination of this dip and changing the FvsT does indeed vary the width of the coherence peak. Future work includes the calculation of the coherence peak using FNAL data.

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