Analysis of the medium field Q-slope in superconducting cavities made of bulk niobium

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Abstract. The quality factor of superconducting radio-frequency cavities made of high purity, bulk niobium increases with rf field in the medium field range (peak surface magnetic field between 20 and about 100 mT). The causes for this effect are not clear yet. The dependence of the surface resistance on the peak surface magnetic field is typically linear and quadratic. This contribution will present an analysis of the medium field Q -slope data measured on cavities treated with buffered chemical polishing (BCP) at Jefferson Lab, as function of different treatments such as post-purification and low-temperature baking. The data have been compared with a model involving a combination of heating and of hysteresis losses due to “strong-links” formed or weakened at niobium surfaces during oxidation, which correlate to $\delta\Delta/kT_c$ changes by baking.

Keywords: surface impedance, niobium, radio-frequency cavities
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1. Introduction

Superconducting radio-frequency (rf) cavities made of bulk niobium are commonly used in particle accelerators for a wide variety of applications. The quality factor of a cavity, $Q_0 = \omega U/P = G/R_s$, is defined as the ratio between the cavity stored energy (U) and the power dissipated in the cavity walls in one rf radian (P/ω). An equivalent definition is the ratio of the geometry factor G , given by cavity shape and rf mode, and the surface resistance R_s of the wall material. A typical plot of the cavity quality factor as a function of the peak surface magnetic field, B_p , in the TM_{010} mode in the range 20–100 mT often shows a more or less marked degradation, referred to as “medium field Q -slope” [1, 2]. Understanding the origin of the medium field Q -slope is important in order to develop a cavity treatment which minimizes it, allowing to produce cavities with reduced cryogenic losses and enhanced rf breakdown fields B_{crit} . In this paper we will present an analysis of the medium field Q -slope data of Nb cavities with a resistance ratio of $RRR > 200$, which did undergo different treatments such as low-temperature “in-situ” baking and post-purification and with different crystallographic properties, such as polycrystalline niobium with different average grain sizes and single crystal. The surface treatment before high-power rf test at temperatures below 4.3 K is a standard one used for niobium cavities and consists of buffered chemical polishing (BCP) with a mixture of HNO_3 , HF and H_3PO_4 in ratio 1:1:1, removing approximately 20 μm of niobium from the inner cavity surface¹, followed by an high pressure rinse (HPR) with ultra-pure water to eliminate surface contaminations, especially dust particles.

¹ After initial removal of $\approx 100 \mu m$.

2. Models for the medium field Q-slope

One of the causes for the increase of the surface resistance, R_s , at increasing rf field is the thermal impedance between the inner cavity surface, where the heat is generated, and the helium bath, cooling the outer cavity surface. Halbritter [3] introduced the following ansatz for R_s as a function of B_p :

$$R_s(T, B_p) = R_{s0}(T) \left[1 + \gamma^*(T) \left(\frac{B_p}{B_c} \right)^2 + O(B_p^4) \right], \quad (1)$$

where R_{s0} is the surface resistance at about 20 mT being the sum of the BCS surface resistance, $R_{BCS0}(T)$, and the residual resistance, R_{res}^0 . $B_c = 200$ mT is the critical field at $T = 0$ of niobium [4] and T is the He bath temperature. The medium field Q-slope is represented by the parameter $\gamma^*(T)$ which depends on $R_{BCS0}(T)$, Kapitza resistance, R_K , thermal conductivity, κ , and wall thickness, d . Recently, Gurevich [5] estimated the increase of the BCS surface resistance due to the pair-breaking effect caused by a high rf field and added an additional term to $\gamma^*(T)$, inversely proportional to T^2 . R_{res}^0 is due to a homogeneously distributed part, e.g., by inelastic surface scattering at localized states of Nb_2O_5 with density n_L , $R_{res}^{\text{IN}} \propto [\omega \min(l, \lambda) n_L]^2 \simeq 10^{-9} \Omega (f/\text{GHz})^2$ and by a localized part at weak/strong links (WL) in distances a_J ,

$$R_{res}^{\text{WL}} = \frac{2(\omega \mu \lambda)^2}{1 + (\omega \hbar / 2eJ_{cJ} R_{bl})^2} \frac{\lambda_J^3}{a_J R_{bl}}, \quad (2)$$

due to the WL normal leakage current resistance $R_{bl} > R_{bn} \simeq \pi \Delta / 2eJ_{cJ}$ of those junctions with the Josephson penetration depth $\lambda_J(T) \propto 1/J_{cJ}(T)^{0.5}$ given by the critical Josephson

current density $J_{cJ}(T)$ [6]. Our experimental results often show a linear increase R_s vs. B_p , in addition. A possible cause for such dependence is the presence of hysteresis losses due to Josephson fluxons (JF) at WL, as described by Halbritter in Ref. [6]. Such WL are oxide-filled boundaries caused by oxidation of fresh Nb surfaces (OWL) or by enforced oxidation of already existing grain boundaries (GWL). J_{cJ} of these WL is of the order of $10^9 - 10^{11}$ A/m², much higher than that of “weak-links” in high- T_c superconductors but still smaller than the critical depairing current density $J_{cp} \simeq 4 \times 10^{12}$ A/m². Halbritter gives the following expression for the hysteresis losses, R_{hys} :

$$R_{hys} \left(B_p \right) \approx \frac{4}{3\pi} \frac{\omega}{2J_{cJ} \left[1 + \left(\omega/\omega_0 \right)^2 \right]^{3/2}} \frac{2\lambda}{a_J} B_c \left(\frac{B_p}{B_c} \right) = R_{res}^1 \left(\frac{B_p}{B_c} \right), \quad (3)$$

where ω_0 is a characteristic JF nucleation frequency and where the finite flux penetration depth, $\lambda_{JF}(\omega)$, and the finite WL depth, w , are omitted. ω_0 decreases from 5 GHz for $J_{cJ} \simeq 10^{10}$ A/m² to about 0.1 GHz for $J_{cJ} \simeq 10^{11}$ A/m² and to about 10 MHz for $J_{cJ} \simeq 10^{12}$ A/m² (Abrikosov-Josephson fluxons) and $\omega_0 \approx$ MHz for slow Abrikosov fluxons (AF) [6]. The dependence given by Eq. (3) has been observed for 1.5 GHz thin films cavities with GWL's with R_{res}^1 between 100-1000 n Ω [7]. R_{hys} , like R_{res}^{IN} and R_{res}^{WL} , are predicted to be temperature independent for $T < T_c/2$.

3. Analysis of experimental results

Since both, quadratic and linear dependencies, of R_s vs. B_p were measured the experimental data have been fitted by the following expression for the field and temperature dependence of the surface resistance:

$$R_s(T, B_p) = R_{s0}(T) + R_{res}^1(T) \left(\frac{B_p}{B_c} \right) + R_{s0}(T) \gamma^*(T) \left(\frac{B_p}{B_c} \right)^2. \quad (4)$$

The fitting parameters are R_{s0} , γ^* and R_{res}^1 and are subjected to the constraint of being greater than zero.

Table 1 shows the values of the fitting parameters and of the fit correlation factor r^2 obtained from a comparison with R_s vs. B_p data measured on a polycrystalline Nb single cell cavity, resonating at 1.467 GHz in the TM₀₁₀ mode, published in Ref. [8] in detail. The cavity was post-purified by heat treatment at 1400 °C for 4 h in a vacuum furnace. The values shown in Table 1 are averaged over seven consecutive rf tests after this post-purification, separated by 10µm BCP, before and after a low-temperature “in-situ” baking at temperatures between 70 and 180 °C for 48 h. Baking yields a slight increase of Δ/kT_c , of γ^* and of R_{res}^0 , whereas R_{res}^1 increases by a factor of two in Table 1. Data taken at 2.2 K, above the superfluid transition temperature of liquid He, show a dominant quadratic dependence with γ^* values above about fifteen [8]. Figure 1 shows the increase of the surface resistance, $R_s - R_{s0}$, as a function of B_p/B_c before and after baking at 105 °C for 48 h. Table 2 shows the average values of R_{res}^1 , γ^* , R_{s0} and r^2 obtained from fits using Eq. (4) with data on the TM₀₁₀ (1.466 GHz) and on the TE₀₁₁ (2.819 GHz) mode in a polycrystalline Nb single cell cavity published in Ref. [9] in detail. Before post-purification the quadratic term is the main component as medium field Q -slope for both modes, while directly after post-purification the linear term dominates. Baking significantly enhances R_{res}^0 and R_{res}^1 where a 5% increase of Δ/kT_c is obtained, also. The values of the fit parameters for the TM₀₁₀ mode after post-purification are consistent with the ones showed in Table 1 on a different cavity. The increase of R_{res}^0 by post-

purification was mainly due to an increase of the trapped residual Earth's magnetic field due to a drift of the current in the compensation coils. Figure 2 shows $R_s - R_{s0}$ as a function of B_p/B_c before and after post-purification for the TM_{010} and TE_{011} modes. Table 3 shows the values of the fit parameters and the fit correlation factor obtained from a comparison with data measured on a single cell cavity made of single crystal niobium, resonating at 2.256 GHz in the TM_{010} mode [10]. The cavity had been post-purified at 1250 °C for about 12 h. The values in Table 3 indicate that, before baking at 120 °C for 48 h, the dependence of R_s vs. B_p is linear and becoming enhanced by baking as R_{res}^0 and Δ/kT_c do drastically. Plots of $R_s - R_{s0}$ as a function of B_p/B_c for the single crystal cavity measured at different temperatures, before and after baking, are shown in Fig.3, indicating at low B_p a similar linear slope between 1.55 and 2K before baking where at higher fields the $R_s(B_p)$ -growth slows down. Remarkable in Tables 2 – 3 are the larger increases of Δ/kT_c and of R_{res}^1 by baking after post-purification than without, e.g., after consecutive 10µm BCP of Table 1. This is in line with observations of B.Visentin [2] and hints to oxide growth depending on the initial state of the surface and to O redistribution, enhancing Δ/kT_c , dominating over O pick-up.

4. Discussion

Common to Table 2 and 3 are fresh, relaxed Nb, e.g., by post purification, yielding by baking larger increases of Δ/kT_c and larger values of R_{res}^1 . Increases of Δ/kT_c from, e.g., 1.70 to 1.80 in Table 2 or 1.83 to 1.90 in Table 3, are due to precipitation of dissolved O towards Nb_2O_5 WL or Nb_6O , as inferred by reduced $R_{BCS}(4.2K)$ values [3, 6]. The

expansion of the Nb₂O₅ layer of WL reduces J_{cJ} , which enhances R_{res}^{WL} and R_{res}^1 , according to Eqs. (2, 3), confirmed by Tables 1-3. For single crystals without extended grain boundaries GWL, $R_{res}^0 \simeq R_{res}^{WL} < 1 \text{ n}\Omega$ is minimal and the initial linear growth slows down with further increasing B field, likely due to the small $\lambda_{JF}(\omega)$ and w values of the OWL's and due to J_{cJ} increasing with distance from the surface. Such details will be discussed in forthcoming publications. Because of our simplified approach, our fits shift weight from R_{res}^1 to $\gamma^* R_{s0}$ or vice versa, as obvious in Table 1–3, and so a temperature dependence of $\gamma^* R_{s0}$ may yield the temperature dependence of R_{res}^1 .

For a more quantitative analysis we model the single crystal data of Table 3 first and we add then oxidized grain boundaries to model the data of Tables 1-2. $R_{res}^0 < 1 \text{ n}\Omega$ at 2.2GHz, qualitatively hints to OWL with large $J_{cJ} (> 10^{11} \text{ A/m}^2)$ and very small leakage current values ($J_{cJ} R_{bl} \gg 100\pi\Delta/2e$) caused by oxidation of fresh, $RRR > 200$ Nb single crystal surfaces. One can speculate that such OWL before baking may show $a_J \simeq 100 \text{ nm}$ and $J_{cJ} \simeq 8 \times 10^{11} \text{ A/m}^2 \simeq J_{cp}/5$ with $R_{bk} J_{cJ} \simeq 1000 R_{bn} J_{cJ}$ where AF [5] may be created above H_{c1J} with $\omega_0 \simeq 0.1 \text{ GHz}$ yielding R_{res}^{WL} and R_{res}^1 -values summarized in Table 4a. Subsequent baking yields further oxidation and O precipitation, decreasing J_{cJ} towards $J_{cJ} \simeq 6 \times 10^{11} \text{ A/m}^2$ with $R_{bk} J_{cJ} \simeq 700 R_{bn} J_{cJ}$ and $\omega_0 \simeq 0.09 \text{ GHz}$ describing Table 3 well. To model the data of Tables 1-2, we consider oxidized grain boundaries (GWL) with an average grain size of $a_J \simeq 100 \mu\text{m}$ before and $\simeq 2 \text{ mm}$ after crystal growth during post purification at temperatures greater than 1200 °C for several hours. In Table 4b we assume R_{res}^1 to be solely due to GWL with $J_{cJ} \simeq 8 \times 10^{10} \text{ A/m}^2$ with $R_{bk} J_{cJ} \simeq 50 R_{bn} J_{cJ}$ (ω_0

$\simeq 0.5$ GHz) and 4×10^{10} A/m² $R_{bt}J_{cJ} \simeq 10R_{bn}J_{cJ}$ ($\omega_0 \simeq 5$ GHz) before and after post-purification, respectively. The values of R_{res}^1 are a factor 2 to 17 higher than the fit results of Table 2, while the values of R_{res}^{WL} indicate a negligible contribution of GWL to the residual resistance R_{res}^0 , as is good epitaxial films [6]. The observed increase of R_{res}^{WL} and R_{res}^1 by baking and post-purification can be described by a reduction of J_{cJ} and growth of w by O segregation mainly towards OWL and GWL, modeled in Table 4a. Crystallites growth without O (NbO) evaporation by post-purification at 1200°C reduces the density of extended GB where all the O segregates too, yielding a lower J_{cJ} , as proposed by Halbritter [6] to explain the increase of the linear slope coefficient for niobium films with increasing grain size. The changed ω -dependence of R_{res}^1 from $\propto 1/\omega^2$ to $\propto \omega$ is due to the J_{cJ} decrease by baking yielding a change of ω_0 modeled well in Table 4b.

To model the quadratic slope coefficients γ^* before and after post-purification, at frequencies 1.467 GHz and 2.819 GHz, in Table 4b we use Equation (8a) of Ref. [3]. γ is the sum of γ^* and an additional pair-breaking term, given in Ref. [5]. Thermal conductivity and Kapitza resistance values for polycrystalline Nb were obtained from Refs. [11-13] for wall thickness of 2.8 mm. The thermal conductivity of the single crystal Nb was obtained from Ref. [14]. The values of Table 2 at 2 K are roughly consistent with theoretical estimate of γ^* in Table 4b, while the pair-breaking contribution (γ) seems to be too large. In addition, γ^* is predicted to be reduced below 2 K due to the exponentially decreasing BCS losses, but such dependence was not observed. Even the addition of the pair-breaking effect as proposed by Gurevich does not explain the higher value of γ^* below 2 K. It is also interesting to notice the slow temperature dependence of the product

$R_{s0}\gamma^*$ in Table 3, increasing from 25 n Ω to 28 n Ω between 2 and 1.55 K. The estimated value of the quadratic slope coefficient is reduced by post-purification, due to the increased thermal conductivity of the niobium.

The observed frequency dependencies $R_{res}^1 \propto \omega^\alpha$ with $\alpha \simeq -2$ to $+1$ fit Eq. (3) with parameters summarized in Sect. 4 and Table 4, and make physical sense: $J_{cJ} < J_{cp}$ is decreasing with heating in air or UHV baking (< 300 °C), $J_{cJ}R_{bl}$ and ω_0 do increase with widening of the Nb₂O₅ OWL by enforced leakage current and JF character. Small R_{res}^1 values in Table 1 are an indication of short OWL, whereas larger γ^* values are related to more localized losses in weaker links indicated by GWL. Small $\delta\Delta/kT_c$ changes ($< 2\%$) around 1.84 by baking seem to be related to defective Nb saturated with O and Nb₆O precipitates, as observed by repeated oxide growth causing strain and dislocations [3,15]. Fresh RRR > 200 Nb, e.g., after post-purification, shows $\delta\Delta/kT_c$ changes up to 5% by baking with $\Delta^0/kT_c \simeq 1.7 - 1.8$ and a marked R_{res}^1 component indicative to many OWL, typical for crack corrosion [15].

5. Conclusion

The analysis of the increase of the surface resistance as a function of the peak surface magnetic field ($B_p \simeq 20 - 100$ mT, so called “medium field Q -slope”) on rf cavities made of bulk niobium shows the presence of both linear and quadratic terms. The linear term has been interpreted as due to hysteresis losses by “strong-links” created at Nb surfaces by oxidation. In the case of single crystal Nb, OWL are nucleated or in the case of fine grain Nb, the oxidation is widening already existing grain boundaries (GWL) with model

parameters summarized in Tables 4. The quadratic term is due to overheating of the cavity inner surface due to the thermal impedance of the niobium and of the niobium/helium interface.

A cavity made of single crystal niobium exhibits a marked linear dependence of R_s vs. B_p and a strong temperature dependence of the slope coefficient which is altered by the low-temperature baking. In addition, baking yields Δ/kT_c increase between 1 – 5 % indicating that O redistribution is more important than O pick-up. These effects cannot be completely explained by our simplified model and require further investigation. A decrease of the linear slope coefficient along with an increase of the quadratic one was obtained from the analysis of the data on polycrystalline cavities below 2 K, in line with more inhomogeneous oxidation and locally enhanced losses [6, 15]. In addition, recent results on temperature maps of the outer cavity surface as a function of B_p [16] show that localized defects contribute to the medium field Q -slope, confirming localized heating as suggested in Ref. [3, 6].

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Figure legends

1. Increase of the surface resistance as a function of B_p/B_c before and after baking at 105 °C for 48 h measured at 2 K (a) and 1.37 K (b) for a single cell cavity which was post-purified. Solid lines represent fits with Eq. (4).

2. Increase of the surface resistance as a function of B_p/B_c before and after post-purification measured at 2 K in the TM₀₁₀ (a) and TE₀₁₁ (b) modes of a single cell cavity. Solid lines represent fits with Eq. (4).

3. Increase of the surface resistance as a function of B_p/B_c before (a) and after baking (b) at 120 °C for 48 h measured at several temperatures on a single cell made of single crystal niobium after post-purification.

Tables

1. Average values of fit parameters to Eq. (4) and correlation factor r^2 obtained from a comparison with R_s vs. B_p data at 2 K and 1.37 K, before and after baking at different temperatures in the range 70–180 °C for 48 h. Data were taken on a niobium single cell cavity resonating at 1.467 GHz in the TM₀₁₀ mode which was previously post-purified. The average value of Δ/kT_c increased from 1.823 ± 0.006 to 1.853 ± 0.008 and the temperature-independent residual resistance, R_{res}^0 increased from 5.3 ± 0.1 nΩ before baking to 6.3 ± 0.1 nΩ after baking.

T = 2 K		
	Before baking	After baking
R_{s0} (nΩ)	21.6 ± 0.1	15.2 ± 0.2
R_{res}^1 (nΩ)	11.6 ± 1.1	20.2 ± 1.9
γ^*	0.04 ± 0.10	0.10 ± 0.24
r^2	0.952	0.950
T = 1.37 K		
	Before baking	After baking
R_{s0} (nΩ)	4.81 ± 0.03	4.82 ± 0.06
R_{res}^1 (nΩ)	2.27 ± 0.31	4.39 ± 0.47
γ^*	1.36 ± 0.14	1.46 ± 0.17
r^2	0.982	0.988

2. Average values of the fit parameters R_{res}^1 , γ^* and R_{s0} and correlation factor r^2 for both, TM₀₁₀ (1.466 GHz) and TE₀₁₁ (2.819 GHz) modes, at 2 K before and after baking, before and after post-purification treatment on a niobium single cell cavity. The average values of the temperature-independent residual resistance and Δ/kT_c are also indicated.

Before post-purification				
	Before baking		After 100-120 °C baking	
	TM ₀₁₀	TE ₀₁₁	TM ₀₁₀	TE ₀₁₁
R_{res}^0 (nΩ)	5.6 ± 0.6	10.1 ± 0.9	9.0 ± 0.5	12.3 ± 0.9
Δ/kT_c	1.705 ± 0.015	1.818 ± 0.013	1.807 ± 0.016	1.828 ± 0.015
R_{s0} (nΩ)	26.9 ± 0.2	51.5 ± 0.4	16.7 ± 0.2	36.3 ± 0.2
R_{res}^1 (nΩ)	7.46 ± 1.46	0.003 ± 3.7	27.1 ± 1.3	5.32 ± 2.40
γ^*	1.55 ± 0.11	2.76 ± 0.14	0.97 ± 0.13	3.34 ± 0.14
r^2	0.995	0.994	0.998	0.997
After post-purification				
	Before baking		After 100-120 °C baking	
	TM ₀₁₀	TE ₀₁₁	TM ₀₁₀	TE ₀₁₁
R_{res}^0 (nΩ)	8.5 ± 0.6	12.0 ± 1.2	17.0 ± 0.2	14.8 ± 0.7
Δ/kT_c	1.690 ± 0.006	1.692 ± 0.014	1.824 ± 0.003	1.780 ± 0.009
R_{s0} (nΩ)	28.7 ± 0.1	79.3 ± 0.3	23.3 ± 0.3	52.7 ± 0.3
R_{res}^1 (nΩ)	16.4 ± 1.1	24.1 ± 2.8	34.1 ± 2.0	71.9 ± 1.5
γ^*	0.01 ± 0.07	0.00 ± 0.06	0.00 ± 0.13	0.25 ± 0.03
r^2	0.987	0.988	0.993	0.989

3. Fit coefficients and correlation factor of Eq. (4) for data on a post-purified 2.256 GHz single crystal niobium cavity taken at different temperatures before and after baking at 120 °C for 48 h. The value of the temperature-independent residual resistance, R_{res}^0 , is 0.8 ± 0.4 n Ω before baking and 10.0 ± 0.3 n Ω after baking and for Δ/kT_c an increase from 1.827 ± 0.032 to 1.900 ± 0.005 is obtained.

Single crystal cavity, before bake				
T (K)	R_{s0} (n Ω)	R_{res}^1 (n Ω)	γ^*	r^2
2	29.1 ± 0.2	12.3 ± 1.3	0.00 ± 0.06	0.983
1.84	18.3 ± 0.2	14.3 ± 1.1	0.00 ± 0.08	0.994
1.69	8.7 ± 0.2	11.8 ± 1.0	0.95 ± 0.14	0.997
1.55	4.2 ± 0.1	20.0 ± 0.6	0.00 ± 0.2	0.999
Single crystal cavity, after bake				
T (K)	R_{s0} (n Ω)	R_{res}^1 (n Ω)	γ^*	r^2
2	24.3 ± 1.1	55.4 ± 2.1	1.03 ± 0.05	0.980
1.84	15.0 ± 1.0	43.5 ± 1.5	1.77 ± 0.13	0.978
1.69	11.8 ± 0.2	36.7 ± 0.6	2.52 ± 0.08	0.995
1.55	10.1 ± 0.2	24.5 ± 0.8	2.81 ± 0.07	0.975

4a. Theoretical estimate of R_{res}^{WL} , of the linear coefficient R_{res}^1 and of the quadratic term γ^* of Table 3 using Eqs. (2, 3) with parameters J_{cJ} , R_{bl} , ω_0 and a_J described in Section 4 for strong-links due to oxidation of a single crystal Nb surface before and after baking. The niobium thermal conductivity and Kapitza resistance are supposed to be unchanged by the low-temperature baking.

	Before baking	After baking
$R_{res}^{WL} \text{ (n}\Omega\text{)}$	1.74	7.3
$R_{res}^1 \text{ (n}\Omega\text{)}$	13.1	86
$\gamma^* \text{ (2 K)}$	0.26	0.26
$\gamma \text{ (2 K)}$	1.92	1.92

4b. Theoretical estimate of R_{res}^{WL} , of the linear (R_{res}^1) and quadratic (γ^* and γ) coefficients at 1.467 and 2.819 GHz before baking, before and after post-purification of polycrystalline niobium using oxidized grain boundaries (GWL), only.

	Before post-purification		After post-purification	
	1.467 GHz	2.819 GHz	1.467 GHz	2.819 GHz
$R_{res}^{WL} \text{ (n}\Omega\text{)}$	0.02	0.09	0.01	0.03
$R_{res}^1 \text{ (n}\Omega\text{)}$	131	40	35	50
$\gamma^* \text{ (2 K)}$	0.67	1.47	0.19	0.78
$\gamma^* \text{ (1.37 K)}$	0.12	0.42	0.06	0.26
$\gamma \text{ (2 K)}$	2.33	3.40	1.72	2.59
$\gamma \text{ (1.37 K)}$	0.80	2.17	0.74	2.01