Strong Meissner screening change in superconducting radio frequency cavities due to mild baking

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(Received 11 December 2013; accepted 4 February 2014; published online 18 February 2014)

We investigate “hot” regions with anomalous high field dissipation in bulk niobium superconducting radio frequency cavities for particle accelerators by using low energy muon spin rotation (LE-μSR) on corresponding cavity cutouts. We demonstrate that superconducting properties at the hot region are well described by the non-local Pippard/BCS model for niobium in the clean limit with a London penetration depth λL = 23±2 nm. In contrast, a cutout sample from the 120°C baked cavity shows a much larger λ > 100 nm and a depth dependent mean free path, likely due to gradient in vacancy concentration. We suggest that these vacancies can efficiently trap hydrogen and hence prevent the formation of hydrides responsible for rf losses in hot regions.

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Superconducting radio frequency (SRF) cavities are the key technology for future particle accelerators for high-energy physics, nuclear physics, light sources, and accelerator-driven subcritical reactors. The invention of the “cold” technology, where the beam accelerating structures are made of superconducting niobium instead of normal conducting copper, revolutionized accelerators, cutting the required operational power by orders of magnitude and allowing to routinely achieve very high accelerating gradients >40 MV/m at the 100% duty factor.1

Several decades of SRF R&D at laboratories and universities worldwide have lead to the successful realization of niobium cavities that reliably achieve very high gradients and quality factors.1,2 However, these structures suffer from a systematic effect of decreasing efficiency for increasing accelerating voltages. This long-standing critical problem is due to the emergence of highly dissipative “hot” regions on cavity surface—a phenomenon known as the high field Q slope. Extensive studies2 demonstrated that all parts of the cavity surface become “hot” regions as soon as the local amplitude of the magnetic field reaches Brf ≈100 mT. For electropolished cavities an empirically found treatment, 120 °C baking for 48 h, causes “hot” regions to disappear, and the whole cavity surface becomes “cold” in a sense that there is no anomalous extra losses emerging at Bfr ≥100 mT.

Despite intense investigations,2 the nature of the hot regions and the mechanism of the 120 °C baking are still subjects of debate. Understanding the cause of these losses and finding the best ways to overcome them are the keys to push SRF cavities performance and to significantly reduce costs for current and future accelerators worldwide. A way to gain this understanding is by studying the differences in microscopic superconducting properties of hot regions and non-dissipative cold regions. Precise microscopic measurements of the magnetic field profile B(z) inside superconductors recently became possible with the development of the low energy muon spin rotation technique (LE-μSR).3,4 The unmatched sensitivity of LE-μSR was demonstrated on thick films of Nb in the clean limit, where a clear evidence for a nonlocal electromagnetic response5 was found, a finding beyond the reach of other existing techniques. Coupled with the ideally suitable depth range of 0–130 nm and spot size of about 1 cm, LE-μSR is an ideal probe to address the problem of hot region emergence.

In this Letter, we directly reveal the superconducting properties within 130 nm from the surface at the hot region of an electropolished (EP) niobium SRF cavity and in the cold non-dissipative region from the 120 °C baked EP cavity. This is achieved by measuring $B(z)$ beneath the surface with LE-μSR: due to the Meissner effect the superconductor expels the applied external field from its interior on a length scale given by the London penetration depth $\lambda_L$, a fundamental microscopic parameter of a superconductor which is directly related to the density of Cooper pairs and the electron mean free path. We demonstrate that the hot region is well described by the non-local clean limit BCS/Pippard electrodynamics with $\lambda_L = 23±2$ nm. In contrast, a cutout sample from the 120 °C baked cavity is found to have a much larger $\lambda > 100$ nm consistent with a strongly suppressed electron mean free path. Interestingly and counter-intuitively, a much larger penetration depth—normally indicative of a weaker, dirtier superconductor—actually leads to a much lower high rf field dissipation. Identification of the physical mechanism for hot region mitigation suggests an alternative route by, for example, impurity doping.

In order to obtain samples with well-characterized microwave dissipation, the best and straightforward way, although destructive, is to perform temperature mapping measurements on state-of-the-art SRF cavities followed by dissection of areas of interest from the walls. Such an approach was proven to be extremely useful in past investigations of field emission and thermal breakdown6 and lately for the high field losses.7–9 For our studies we used two niobium SRF cavities of TESLA elliptical shape10 with a residual resistivity ratio, RRR ~ 300. After manufacturing both cavities were electropolished for removal of about 120 μm material using a standard solution of

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All three samples have been extracted from these locations using a low rotary speed milling machine with no lubricant to minimize contamination. The interior walls of the EP sample (100–6) for the unbaked cavity, and subjected to buffered chemical polishing (BCP) for 20 h at 120 °C as a last step. No hydrogen degassing at 600–800 °C was applied to these cavities. Detailed measurements of the microwave dissipation in the fundamental TM_{010} mode with f_0 = 1.3 GHz were performed at T = 2 K using both standard phase-lock techniques and, independently, by a temperature mapping system similar to Ref. 13, attached to the outside cavity walls. Such local thermometry consists of 576 Allen-Bradley carbon resistors arranged in 36 boards (equally spaced every 10° around the cavity rotational axis) with 16 thermometers in each board. It measures the heating of the outside cavity wall caused by the microwave dissipation on the inside surface. The local temperature increase ΔT at each thermometer location is proportional to the dissipated power on the inside wall, ΔT ≈ P_{diss} ∝ R_s(B)B^2, thereby providing a direct measurement of the local surface resistance R_s since the distribution of surface magnetic field is known from numerical calculations.

Quality factors Q_0 of both cavities measured at T = 2 K as a function of B_{peak} are shown in Fig. 1(a). The drastic difference at high fields is a typical result of the 120 °C baking. Typical “unfolded” temperature maps at B_{peak} = 119 mT are shown in Figs. 1(c) and 1(d). A clear difference is apparent; the unbaked cavity shows a much stronger heating within the belt between sensors number 4–12, which corresponds spatially to the high surface magnetic field on the inside cavity surface [Fig. 1(b)]. The surface magnetic field at different sensor locations is shown in Fig. 1(b). Notice that it is very close to the peak surface magnetic field B_{peak} for sensors 4–12. Based on the rf measurements we have selected representative samples—(100–6) for the unbaked cavity, and (340–6) for the 120 °C baked cavity—which (due to 120 °C bake) have drastically different ΔT(B) as shown in the inset of Fig. 1(a). Circular cutouts of ~11 mm diameter were extracted from these locations using a low rotary speed milling machine with no lubricant to minimize contamination. Another sample extracted from the location (30–6) in the EP unbaked cavity and subjected to buffered chemical polishing (BCP) for 20 μm material removal was used to represent unbaked BCP-treated cavities which exhibit similar Q_0 (B_{peak}) behavior to the EP ones. All three samples have been subsequently investigated with LE-μSR.

The μSR technique uses beams of 100% spin-polarized positive muons (μ^+), which serve as sensitive local magnetic probes when implanted inside a sample. At the μE4 beamline at PSI, a high intensity surface muon beam with an energy of ~4 MeV is moderated to ultra-low epithermal energies (~15 eV) in a cryogenically condensed solid Ar film deposited on a 10-K-cold Ag foil. These epithermal muons are subsequently accelerated by electrostatic fields to energies E ≤ 30 keV, corresponding to implantation depths up to ~140 nm in Nb. The schematic of the experimental arrangement and measured muon flux distribution on the sample are shown in Figs. 2(a) and 2(b). Upon implantation, the muon precesses in the local magnetic field at its stopping site. The precession frequency is proportional to the magnetic field and is measured by detecting the anisotropic muon decay (lifetime τ_μ = 2.2 μs): the decay positrons are preferentially emitted in the direction of the μ^+ spin, which allows to monitor the time evolution of the muon spin by registering the positrons in detectors surrounding the sample.

FIG. 1. (a) The intrinsic cavity quality factor Q_0 at T = 2 K as a function of peak surface magnetic field B_{peak} on the cavity surface for the electropolished unbaked (■) and 120 °C baked (▲) superconducting cavities from which samples were dissected; the inset shows the rf heating ΔT(B) for cutout samples—notice the strong correlation between the ΔT increase and Q_0 degradation. (b) Calculated magnetic field distribution on the surface of the cavity at the locations of the temperature sensors. (c)–(d) “Unfolded” temperature maps of the outside walls at B_{peak} = 119 mT for: (c) unbaked electropolished cavity; (d) electropolished +120 °C baked cavity. White areas correspond to ΔT < 10 mK. Locations for sample cutout are marked on the maps by circles.

FIG. 2. (a) Schematic of the experiment; (b) measured muon flux distribution; (c) asymmetry signals A(τ) in normal (top) and superconducting (bottom) states of the EP sample (100–6) at muon implantation energy E = 12.5 keV corresponding to the mean stopping depth of about 42 nm.
The number of positron events at each of the detectors is described by the following form:

\[ N(t) = N_0 \exp(-t/\tau_p)[1 + A(t)] + N_{\text{bkg}}, \quad (1) \]

where \( A(t) = A_0 P(t) \) describes the time evolution of the muon ensemble polarization \( P(t) \), and \( A_0 \) is the experimental decay asymmetry. \( N_{\text{bkg}} \) is a time-independent uncorrelated background. The asymmetry \( A(t) \) is given by averaging over the muon stopping distribution \( n(z, E) \)

\[ A(t) = A_0 \exp\left[-\frac{(\sigma t)^2}{2}\right] \int n(z, E) \cos[\gamma B(z)t + \phi] \, dz; \quad (2) \]

where \( \gamma = 2\pi \times 135.54 \) MHz/T is the muon gyromagnetic ratio, \( \phi \) is the detector phase, and \( \sigma \) is a Gaussian depolarization rate, reflecting the dipolar broadening due to nuclear spins.

The experimental procedure was to perform zero-field cooling to \( T = 3 \) K and then apply a magnetic field, \( B_a \), parallel to the sample surface and transverse to the muon spin [see Fig. 2(a)]. The magnitude of \( B_a \) was confirmed in each case by performing a run in the normal state at \( T = 10 \) K above the transition temperature of niobium (\( T_c = 9.25 \) K) where the Meissner effect is absent. Muon implantation energies of \( 3.3 \leq E \leq 25.3 \) keV were used. See supplementary material for corresponding implantation profiles simulated using the computer code TRIM.SP. For the simulations, niobium oxide (\( \text{Nb}_2\text{O}_5 \)) of 5 nm thickness was assumed as the topmost layer.20 Systematic uncertainty in these simulations is estimated to be of order 2%, which translates into \( \leq 2 \) nm of the mean depth uncertainty.

Several million decay positrons were collected for each muon energy. Examples of asymmetry signals \( A(t) \) obtained on the same sample (100-6) at \( B_a = 15 \) mT, \( E = 12.5 \) keV in the normal and superconducting states are shown in Fig. 2(c). The Meissner effect becomes manifested in the reduction of the precession frequency and the heavily damped \( A(t) \) caused by the broad field distribution in the stopping range of the muons.

All data were analyzed using the program musrfit.21 We used two fit models: a simple Gaussian model, and a numerical time-domain model based on the non-local Pippard/BCS model. For comparing \( B(z) \) between the samples, we first use the well-established Gaussian approximation

\[ A(t) = A_0 \exp\left[-\frac{(\sigma \Delta t)^2}{2}\right] \cos(\gamma \mu B_G t + \phi), \quad (3) \]

where \( B_G \) is in very good approximation equal to \( \langle B \rangle \) which is given by

\[ \langle B \rangle = \int_0^\infty B(z) n(z, E) \, dz. \quad (4) \]

The screened magnetic field \( B_G \) as a function of the mean muon stopping depth

\[ \langle z \rangle = \int_0^\infty z n(z, E) \, dz \quad (5) \]

is presented in Fig. 3 for all three samples.
Thus are also con-

results. However, we found no evi-

dence for surface pinning reported in Ref. 9.

Small nanoscale hydrides have been lately proposed to

be the cause of the anomalous high field dissipation,31 and

the 120 °C baking effect attributed to the injection of vacan-
cies.32 Preliminary structural investigations33,34 are also con-

sistent with this picture. Within this model hydrides remain

superconducting by proximity effect up to the high field dis-
sipation onset (B_{rf} \approx 100 \text{ mT}), which is consistent with the

clean limit Pippard/BCS description at B \leq 25 \text{ mT} of

LE-μSR data for the hot region (100-6). If hydride precipita-
tion is suppressed by the 120 °C baking, then \ell remains low

upon cooldown to T \leq 3 \text{ K}, which is in agreement with our

findings on the baked cavity sample (340-10). Thus our

results can be explained by the assumption that efficiently

trapping hydrogen is the mechanism to eliminate the forma-
tion of nanoscale hydrides (hot regions). These vacancies

introduced by the 120 °C baking may also explain the depth
dependent mean free path after baking.

It is also worth mentioning that we have performed

zero-field measurements on all samples to search for any

near-surface magnetic impurities motivated by recent super-

conducting quantum interference device (SQUID)35 and

point contact tunneling36 results. However, we found no evi-
dence for surface magnetism and no difference between the

two samples.

In conclusion, we have directly measured and com-
pared the magnetic penetration depth in hot (highly dissip-
avive at high rf fields B_{rf} \approx 100 \text{ mT}) regions of bulk niobium

SRF cavities for particle acceleration with that in non-
dissipative regions obtained by 120 °C baking. For the hot
region Meissner screening at B \leq 25 \text{ mT} is well described

quantitatively by Pippard/BCS non-local electrodynamics
with \lambda_L = 23 \pm 2 \text{ nm}. For the 120 °C baked cavity cutout
the magnetic field penetrates much deeper (\lambda > 100 \text{ nm}), and

the decay is well described by a depth-dependent electron

mean free path in the range of 2 \leq \ell \leq 16 \text{ nm}. We propose

that vacancies introduced by the 120 °C baking may pre-
vent hydride precipitation responsible for strong rf losses
in hot regions while also leading to the depth gradient in

the electron mean free path. Our findings suggest impurity

doping of the surface layer as a possible alternative route
for hot region mitigation and further SRF cavity

improvement.

We acknowledge Hans-Peter Weber for his excellent

technical support. Fermilab is operated by Fermi Research

Alliance, LLC under Contract No. De-AC02-07CH11359

with the United States Department of Energy. A.R. and F.B.

were partially supported by the U.S. DOE Office of Nuclear

Physics.

BPC (30-6) sample are \lambda_L = 25 \pm 2 \text{ nm} and d = 15 \pm 1 \text{ nm}.

Both EP (100-6) and BPC (30-6) samples have \lambda_L, which are

reasonably close to the thick Nb film value \lambda_L = 27 \pm 3 \text{ nm}

(Ref. 5) and are somewhat lower than that from bulk magne-
tometry measurements\textsuperscript{27} \lambda_L = 46 \pm 2 \text{ nm}. However, if the

latter is "renormalized" to take into account non-weak

coupling in niobium (\lambda_L \rightarrow \lambda_L/\sqrt{Z}, Z \approx 2.1), we get

\lambda_L = 32 \pm 2 \text{ nm} closer to our values.

Unlike data for the (100-6) hot region sample and (30-6)

BPC sample, data for the (340-10) sample from the baked
cavity could not be described by the Pippard/BCS model

with a single mean free path \ell. In this case we had to follow

a different procedure: values of \lambda_L and d were fixed to those

of the unbaked sample (100-6) and \ell was used as a free pa-

rameter. The assumption of the same \lambda_L is reasonable since

the same bulk niobium is used for both samples.

The assumption of the same d is also reasonable since samples

have the same nanoroughness of the EP-treated surface, and

further support is provided by the Gaussian model fits [see

Fig. 3]. The results of the Pippard fitting procedure (individ-

ual points) are shown in Fig. 4 along with the calculated

depth profiles for different values of \ell (solid lines). Data

indicates a depth-dependent mean free path with the lower

value (\ell = 2 - 4 \text{ nm}) in the first \sim 60 \text{ nm} followed by an

increased \ell \geq 16 \text{ nm} at larger depths. Using niobium concen-
tration n_{\text{Nb}} = 3.6 \times 10^{22} \text{ cm}^{-3}, estimated atomic concen-
tration of scattering centers for \ell = 2 \text{ nm} is \sim 1/(n_{\text{Nb}} \ell^3)

\approx 0.2\%, which is not expected to have a significant impact

on T_c via electron-phonon scattering modification.\textsuperscript{28} Thus

the assumption about the same \lambda_L is reasonable.

Previous microwave cavity studies\textsuperscript{14,29,30} suggested that

one of the effects of the 120 °C baking on niobium may be a

significant decrease of \ell in the first \sim 20–30 \text{ nm} from the

surface causing a crossover from clean (\ell \gg \ell_0) to dirty

(\ell < \ell_0) limit. We observe a strong increase of the penetra-
tion depth in the (340–10) baked cavity sample, which can

be consistently described by such a mean free path suppres-
sion. Our findings are also qualitatively consistent with

measurements of B_{c2} reported in Ref. 27 where it was also

found that 120 °C baking of EP samples leads to an increase

of surface B_{c2} indicating "dirtier" material. A strongly sup-

pressed surface \ell is also in line with the increased surface

pinning reported in Ref. 9.

| Cavity/sample | Applied magnetic field (mT) | \lambda_L (nm) |
|--------------|-----------------------------|--------------|
| EP (100-6)   | 5                           | 21.8(4)      |
|              | 15                          | 23.5(4)      |
|              | 25                          | 24.4(5)      |
| BCP (30-6)   | 28                          | 24.8(4)      |

FIG. 4. Average normalized field vs. mean muon stopping depth. Individual

points correspond to the Pippard fits based on single energy measurements

only. Solid lines for unbaked EP (100-6) and BCP (30-6) samples show
global fits based on the Pippard/BCS model. Solid lines for the EP+120 °C

baking may also explain the depth dependent mean free path after baking.
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