RESULTS FROM ATOMIC LAYER DEPOSITION AND TUNNELING SPECTROSCOPY FOR SUPERCONDUCTING RF CAVITIES*

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Abstract

Atomic Layer Deposition (ALD) is a process that synthesizes materials in successive monolayers, at rates up to 1 micron/hour. We have been using this technique at Argonne as a possible way to improve superconducting radio frequency (SCRF) cavities performances. Initial experiments using tunneling spectroscopy and ALD have led to a new model for dissipation mechanisms occurring at the surface and news ways of controlling SCRF surfaces, as well as suggesting ways to significantly improve the operating gradients of superconducting cavities. Initial measurements of ALD treated samples show significant improvement over untreated cavity-grade Nb samples. We will report these results.

INTRODUCTION

For over three decades, the SCRF cavities performances have been continuously improved to reach now reproducible quality factor of 10^10 and max accelerating field E of 30 MV/m. However, unresolved mysteries among which High field Q-slope and its mitigation by a mild baking treatment remain and prevent cavities from reaching the intrinsic Niobium limit believed to be around 50-55 MV/m. The interactions between Nb surface superconductivity and the native oxides are complex and not fully understood; a fundamental investigation of the microscopic mechanisms by which oxygen and the complex set of niobium oxides influence cavity performance seems now to be unavoidable. We will present first, a tunneling spectroscopy study of the mild baking effect on cavity-grade electropolished Nb samples that indentify magnetic impurities as a heretofore unrecognized contributor to the dissipation mechanism at the surface of Nb: In a second part we will present the Atomic Layer Deposition technique and the first attempts to improve the Nb surface superconductivity by getting ride of the oxides: A uniformly 2nm thick Alumina film has been deposited by ALD on Niobium sample and followed by a High temperature heat treatment in Ultra High Vacuum (UHV). In conclusion, we will present the first results obtained on coated cavities and the future projects associated with ALD.

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THE MILD BAKING EFFECT

As we reported previously [1] tunneling measurements on cavity-grade Nb directly probe the surface superconductivity. Our results provide new insights into the Q-slope problem and the baking effect. Air exposed, electropolished samples reveal a surface gap parameter characteristic of clean, bulk Nb (Δ=1.55 meV) but the tunneling density of states (DOS) is considerably broadened, samples treated using the same mild baking step that reduces the Q-slope (e.g., 120°C for 24h - 48h) show much sharper DOS and reduced zero-bias conductance (Figure 1 Center and right) and this effect prevails whether the sample is baked in air or in vacuum.

Niobium oxides have been widely studied in their bulk, crystalline form: Nb_2O_5 is an insulator with a band gap > 4 eV, NbO_2 is a weak Peierl semiconductor with a gap of 0.1 eV and NbO is metallic and superconducting below 1.4 K. Beyond these three main categories, each of these oxides has the unique property to stabilize a substantial off-stoichiometry, a pristine air-exposed Nb surface develops then a more complex set of oxides with a continuum of gradually increasing oxygen concentration NbO_X from interstitial oxygen (x<1) near the bulk niobium, up to the top most Nb_2O_5 (x=2.5). Oxygen vacancies in the Nb_2O_5 (NbO_X with 2<x<2.5) create Nb4d1 ions that carried an unpaired electron. These ions defects can then be seen as localized charge and spin carriers. Cava et al. [2] showed that indeed, sub-stoichiometric bulk crystalline Nb_2O_5 develops magnetic moments and metallicity, both increasing with the oxygen vacancies concentration.

Several mechanisms can lead to a broadening of the DOS: proximity effect due to a metallic overlayer, strong structural disorder that induces a mean free path $l \ll \xi$, the superconducting coherence length (35 nm for Nb). However all these effects will decrease the gap amplitude and shift the quasiparticles peaks to lower energy. This is in contradiction with our results that show (Figure 1 Right) a shift of the peaks to higher energies than the bulk Nb gap Δ=1.55 meV. To our knowledge only one mechanism can explain these results: Pair-breaking caused by inelastic scattering on magnetic impurities.

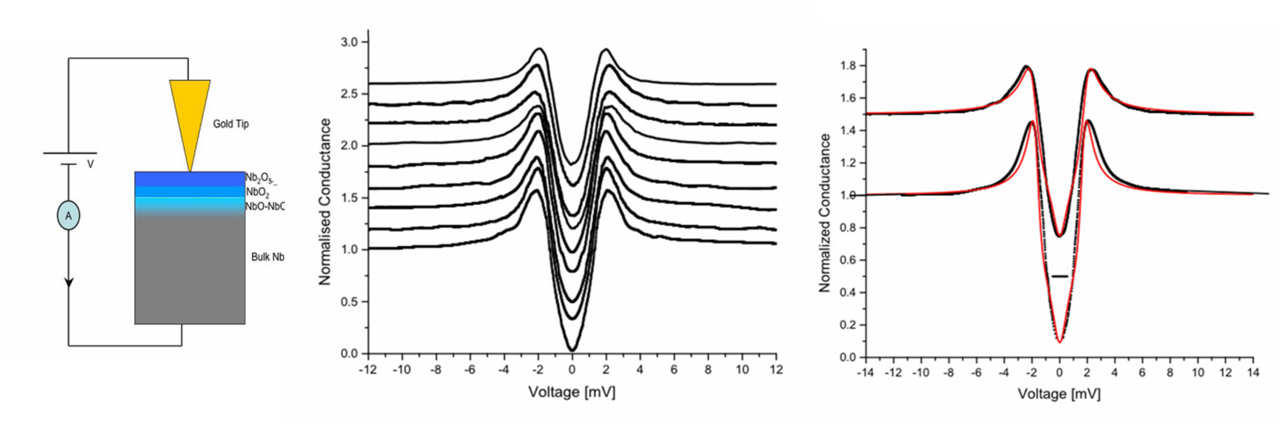

Figure 1: Left: schema of the point contact experiment. Center: 10 conductance curves measured at 1.7K on a baked Nb single crystal. Right: Average conductance curves for a baked (bottom) an unbaked (top) Nb sample. In red the corresponding fits with the Shiba theory.

The Shiba theory [3] describes the effect of paramagnetic magnetic impurities on the superconducting DOS in the strong coupling limit: Additional quasiparticles state develops at an energy ε inside the gap where ε is a parameter that depends on the coupling strength (the weak coupling limit, or Abrikosov-Gorkov theory, is given by ε=1). The scattering rate of quasiparticles, $\alpha=\Gamma/\Delta$, where Γ is the quasiparticles lifetime, broadens the DOS and shift the peaks to energies E> Δ (Figure 1 Right). The best fit gives: Δ=1.55 meV, ε=0.62, $\alpha_{Unbaked}$=0.3 for an unbaked sample and Δ=1.55 meV, ε=0.62, α_{baked}=0.2 for a baked sample. From these parameters, we can extract the corresponding concentration of magnetic impurities and find 0.2% for an unbaked sample and 0.13% for a baked sample. The total amount of quasiparticles inside the gap can be calculated by integrating conductance curves between +Δ and -Δ. We find the ratio of the areas for baked and unbaked curves to be equal to ~1.5, the dissipation mechanism of a baked sample is then reduced by the same amount with respect to an unbaked sample. This result is in good agreement with the ratio of the Quality factor Q at low field for unbaked and baked cavities.

The microscopic origin of the mild baking effect and the related decrease of magnetic scattering can be explained by a simple mechanism: Irreversible local rearrangements of oxygen in the niobium oxides occur even at such low temperature; the growth of a thicker [4], more stable NbO_2 Peierls insulator, non magnetic by nature, acting as a protective layer. NbO_2 prevents then partially the superconducting pairs from being scattered by magnetic impurities present in the Nb_2O_5 located on top of NbO_2. As the probability of a pair to tunnel through an insulator decreases exponentially with its thickness, the drop off of magnetic impurities concentration, as felt by the superconductor, requires only a few Angstrom thick stoichiometric NbO_2.

ATOMIC LAYER DEPOSITION

Atomic Layer Deposition (ALD) [5] is a method of synthesizing materials in single atomic layers that can achieve growth rate of a few microns/hr. We are studying an alternative solution to remove the oxides from the Niobium surface by ALD coating cavity-grade electropolished Nb samples with a uniform 2-3nm Al_2O_3 protective overlayer and heat them in UHV at high temperature [6]. The coated samples were baked in vacuum at temperature ranging from 250-500 C and the XPS spectrum of the Nb3d core level was measured simultaneously. After each thermal treatment the samples transferred in air to the point contact apparatus and the surface superconducting DOS was probed by tunneling spectroscopy at a temperature of 1.6K.

We found reproducibly that a low temperature baking from 220-380C leads to reduced superconducting features: a gap Δ< 1.55 meV and an increased inelastic scattering parameter that fills the gap with normal electrons and increases the dissipation. At temperature above 450 C, however, Δ recovers the bulk niobium gap value of 1.55 meV and the inelastic scattering parameter decrease drastically down to 0.08 meV. At 250 C for 2h (green), the XPS spectrum shows a reduction of Nb_2O_5 into sub-oxides besides oxygen diffuses on a distance of the order of λ, thus increasing the oxygen concentration and the dissipation in the induced current layer. The strong increase of the inelastic parameter Γ=0.9 meV and the decrease of the gap Δ=1.45 meV, extracted from the fit of the conductance spectrum, indicates an important surface pollution of the superconducting niobium.

At higher baking temperature: 380 C during 20h (in orange), the surface oxides are now composed mostly of metallic oxides NbO and NbOx (x<1) with a small amount of NbO2. The tunneling conductance curve reveals a pronounce decrease of the superconducting gap

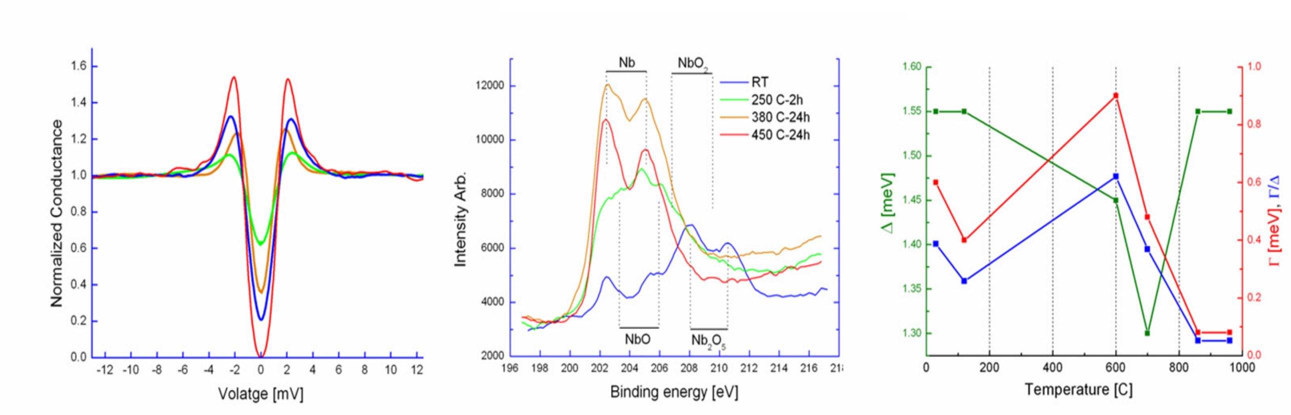

Figure 2: Left: Tunnel conductance curves at 1.7K. Center: corresponding XPS spectrum. Left: evolution of the gap and inelastic scattering parameter as a function of the baking temperature.

down to Δ=1.3 meV, consistent with the presence of a thicker metallic overlayer on top of niobium. As interstitial oxygen diffuses on several mm >> λ for the baking parameters, a cleaning of the interface oxides/Nb leads to the decrease of the inelastic scattering parameter Γ=0.35 meV. Finally for temperature above 450 C during 20h (in red), the XPS spectrum shows that niobium oxides are now composed of pure NbO together with a sharpening of the metallic niobium peaks Nb^o. The related tunneling spectrum shows a recovering of the bulk Niobium gap Δ=1.55 meV and a decrease of the zero bias conductance to less than 5%, indicative of a strong reduction of the normal electrons inside the gap: Γ=0.08 meV. In agreement with Arnold theory and previous experiments, the superconductivity is fully induced in a thin, clean NbO metallic overlayer without any reduction of the underneath niobium gap Δ=1.55 meV. The coated cavity baked at this temperature should show better performances, close to the ideal limits of a perfect Nb superconductor.

CAVITY TEST AND FUTURE PROJECTS

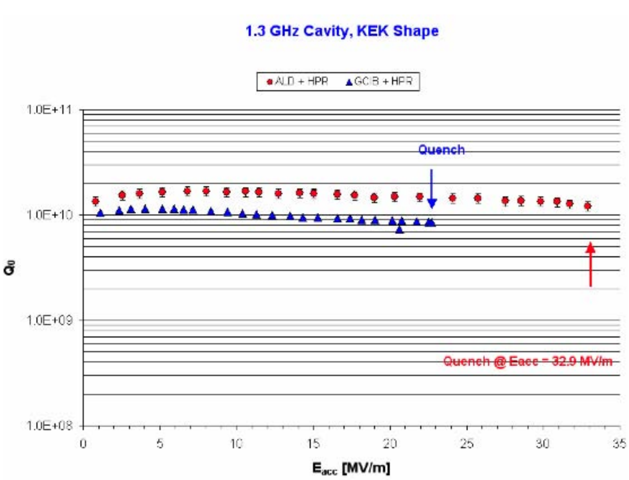

Figure 3: RF test of a 1.3 GHz single cell cavity before (blue) and after (red) ALD coating.

We tested the Atomic layer deposition process on a 1.3 GHz niobium cavity from Jlab. The cavity where inserted into the ALD line and coated with 10nm an Alumina layer covered after by 3nm of niobium pentoxide Nb2O5 to avoid multipacking effect. Fig.3, summarize the RF test before and after the ALD coating, without any baking. This first result proves clearly that not only the ALD layered structure didn't do any harm to the cavity but on top of that increased the Quality factor by a factor of 3, the maximum accelerating field Emax by a factor of 1,5 and at last suppressed totally the High field Q-slope. We are still investigating the possible reasons for such improvements.

In principle a wide range of metals among which elemental niobium, metal oxides, nitrides or more complex compounds can easily be achieved by ALD even superconducting such as NbN, V3Si, Nb3Ge.

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