Comment on ''Direct observation of the superconducting energy gap developing in the conductivity spectra of niobium''

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For Nb-surface impedance measurements, various deviations from BCS and Eliashberg theory have been reported. Those claimed deviations from theory are instead related to chemistry, i.e., by taking the Nb-O reaction properly into account. [S0163-1829(99)00642-6]

In a recent paper[1] it was claimed that conductivity data of superconducting Nb films near the gap frequency deviate significantly from the BCS theory, even if strong-coupling corrections are included. This paper does not reference various papers on the Nb where the BCS theory was able to fit even fine details relying on material science.[2–13] Clearly, those experimental results are not compatible with the BCS theory of homogeneous Nb with ideal surface. Instead, the crack corrosion by Nb oxidation sketched in Fig. 1 (Ref. 6) yields weak links, Nb-NbO_x-Nb_2O_{5-y} ($x,y<1$) interfaces and NbO_x clusters which weaken locally the Nb superconductivity[6,13] and globally $2\Delta/kT_c=4.05$ down to 3.85–3.5 and T_c from 9.2 K to 8–7 K for heavy oxidation as observed by surface impedance measurements. In addition, the NbO_x-Nb_2O_{5-y} interfaces reduces the mean free path l by surface scattering and, as weak links, yield by a reduced critical Josephson current density $j_{cJ}(T)$ and a normal leakage current resistance $R_{\rm bl}$, rf residual losses $R_{\rm res}$, penetration depths $\lambda(T)$ jumps and enhancements,[6] and easy flux generation and flux penetration.[11]

In contrast to bulk Nb, sputtered Nb grows in island and the Nb-O interaction is not pinned to ''outer'' surfaces but acts throughout the sputter process because the fresh Nb surface cleans the sputter gas as the best pump of the system. Thus the initial low-T_c values[14] are mainly due to O gettering, especially at boundaries. With further increasing Nb-film thickness the now cleaner sputter gas allows a T_c growth and grain size $a_{\rm GB}$ increase.[14] The NbO_x-Nb_2O_{5-y} at grain boundaries enhance the boundary resistance $R_{\rm bn}$ well above the intrinsic Sharvin resistance $R_{\rm sh}\cong 4\times 10^{-12}$ $\Omega\,{\rm cm}^{-2}$ and reduce j_{cJ}, which by Nb_2O_{5-y} oxygen vacancies is accompanied by the leakage current $R_{\rm bl}>R_{\rm bn}$.

Hence a two component system is appropriate for the conductivity of sputtered Nb films given by the intrinsic conductivity $\sigma_{\rm BCS}$ and the grain-boundary conductivity $\sigma_{\rm bn}$ in the thin-film limit by

$$\frac{1}{\sigma}=\frac{1}{\sigma_{\rm BCS}}+\frac{1}{\sigma_{\rm bn}}=\frac{1}{\sigma_{\rm BCS}}+\frac{\hbar\omega}{a_{\rm GB}2ej_{CJ}(T)[i+\omega\tau_J(T)]} \tag{1}$$

with $1/\tau_J(T)=2ej_{cJ}R_{\rm bl}/\hbar$ as Josephson frequency. The parameters of both components deduced from Ref. 1 are as follows: $RRR=6$ yields $a_{\rm GB}\cong 20\cong l_{\rm res}$ and $R_{\rm bn}\cong 10^{-11}$ $\Omega\,{\rm cm}^2$.[12] The Nb_2O_{5-y}-NbO_x interfaces in the 15-nm-thick Nb film of Ref. 1 show $T_c\cong 8.3$ K (Ref. 6) and degraded gap parameters $\Delta^*<\Delta_0$ (Ref. 13) which yield $j_{cJ}(0)\approx 2\times 10^8$ A/cm^2 at island boundaries and, together, with the above parameters, with Ref. 6, and with Eq. (1) $\lambda(0)\cong 90$ nm as penetration depth. Similarly, the T and ω dependencies shown in Fig. 5 of Ref. 1 are fitted by Eq. (1) with $\sigma_{\rm BCS}(T,\omega)$ of Refs. 1 and 4 and $\sigma_{\rm bn}(T,\omega)$ obtained by the above parameters with $R_{\rm bl}(T<0.8\,T_c)\cong$const and with $j_{CJ}(T)\propto\tanh(\Delta/2kT)$. According to Eq. (1) $\sigma_1(T<0.8\,T_c$, $\omega\ll 1/\tau_J)$ saturates at $a_{\rm GB}/R_{\rm bl}$ which fits to a large number of experiments[11] on Nb films, on bulk Nb,[6] and on YBCO films, e.g., in infrared measurements.[15] Such rf residual losses are the clearest deviation from BCS/Eliashberg theories. A more detailed analysis of the data of Ref. 1 with Eq. (1) may yield insights into the Δ^* degradations at interfaces[13] and, possibly, into deviations from the BCS/Eliashberg theory.

In summary, material science and chemistry are crucial for the surface impedance of superconducting reactive materials, like Nb with its strong chemical bonding to O, H, or N. Hence the claimed deviations from the Eliashberg theory in Ref. 1 seem to be overshadowed by Nb-NbO_x-Nb_2O_{5-y}-interface effects at intrinsic and extrinsic boundaries, as sketched in Fig. 1.

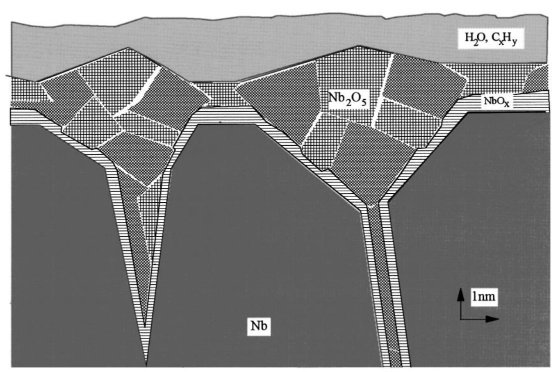

FIG. 1. Sketch on oxidized Nb surface showing crack corrosion by the enhanced Nb_2O_5 volume (factor 3) as compared to Nb. Those Nb-NbO_x-Nb_2O_{5-y}-NbO_x-Nb weak links/segregates ($y,x\leq 1$) may reach depths between 0.01-1/1-10 μm depending on Nb quality and oxidation strength (Refs. 5–8). As planar defects they act as Josephson junctions with $j_{cJ}(T)$ as Josephson critical current and with $j_{\rm bl}(T)$ as normal conducting, leakage current.

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