NIOBIUM NITRIDE COATED SUPERCONDUCTING CAVITIES

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1.- INTRODUCTION

We have previously presented results concerning the characterization in the X band of superconducting niobium nitride (NbN) films prepared by reactive DC magnetron sputtering (1). The main advantages of this material for RF cavities are: low secondary electron emission coefficient (2), good immunity against radiation and chemical agents and less serration in oxidation than niobium, indicating less RF residual losses and higher RF breakdown fields (3). Moreover, thermal breakdown due to the quenching of certain microscopic regions from the superconducting state to the normal state is reduced thanks to the high transition temperature. Finally, NbN exhibits lower RF losses than Nb, at the same temperature (1).

Two techniques are usually employed to prepare the so-called δ-NbN phase having good superconducting properties: reactive sputtering in a nitrogen-argon atmosphere, and thermal diffusion of nitrogen into niobium followed by a rapid quench cooling.

Sputtered NbN films have granular type microstructure, yielding high B_{C2} - and J_c - fluxoid properties (4), and are well-adapted for the protection of surfaces with simple forms. However for complicated geometries it is rather difficult to obtain homogeneous NbN films by this technique. Thermal diffusion technique has been used for the first time about forty years ago (5), but has been rarely employed since then, due to the difficulty for obtaining the δ - NbN phase at temperature below 1300°C. For our application to RF cavities, the NbN obtained by thermal diffusion is intended not to be of the granular type structure, having thus lower RF residual losses R_{res} and higher RF breakdown fields B_{crit}. The results reported here are related to NbN obtained by the thermal diffusion technique on bulk niobium at temperatures well below 1300°C.

2.- PREPARATION OF NIOBIUM NITRIDE

The phase diagram of NbN (6) shows different phases, among which the δ phase is the needed one for good superconductivity; it has a cubic B1 structure , exhibits high transition temperatures ($\leqslant$ 17 K), but is stable only at temperatures higher than about 1300 °C. Its preparation under other conditions entails either quenching, or impurities such as, for instance, oxygen (7,8).

The NbN samples have been prepared at KfK (Karlsruhe). The bulk niobium used is first cleaned, then heated at high temperature (1850 °C) during two hours at 10^{-7} Pa, then cooled down to about 800°C, ($\simeq 10^{-8}$Pa), where gaseous nitrogen is admitted at a pressure of 1 Pa during 5 minutes for the nitridation process. The oven is then turned off and nitrogen is rapidly admitted until 1 bar, for the cold quenching process (9). Using this simple technique, we have prepared a TE011 mode cavity resonating at 9 GHz,and cylindrical rods used as test samples for the determination of the transition temperature using the inductive method at low frequency ($F \leqslant 1$ MHz).

3.- CHARACTERIZATION

The measurement of T_c is performed with the inductance variation method on a magnetic coil, the core of which is the NbN rod. The measurement frequency is in the 100 kHz to 1 MHz band. The magnetic coil is part of a resonant circuit which is excited by a tunnel diode and the resonant frequency is recorded as a function of temperature. The transition

temperature is obtained with good precision ($< 10^{-2}$ K) through fitting, and the penetration depth is given by the eigenfrequency variation. The values obtained are then compared with the results given by measurements at high frequencies.

The apparatus used for characterization at high frequencies (9 GHz) has been described in a previous paper (1). The surface resistance is derived from measurements of the quality factor Q as a function of temperature. The penetration depth is determined from the variation of the eigenfrequency F_o of the cavity as a function of temperature T, when T increases towards T_c. If ΔF is the variation of the resonant frequency F_o, the following relation is given by the perturbation theory:

$$\Delta F \;/\; F_o \;=\; \Delta\lambda . \pi \mu_o F_o \;/ G$$

where $\Delta\lambda$ is the variation of the penetration depth and G the geometrical factor.

4.- EXPERIMENTAL RESULTS. DISCUSSION

RF measurements have been performed with a cavity oscillating at 9 GHz in a TE011 mode. Following the BCS theory, the penetration depth λ_o at T = 0 K and the transition temperature T_c are obtained by the fit of (fig.1):

$$\Delta\lambda(T) \;=\; \lambda_o / \; \left\{1-(T/T_c)^4\right\}^{1/2} = \; \lambda_o Y$$

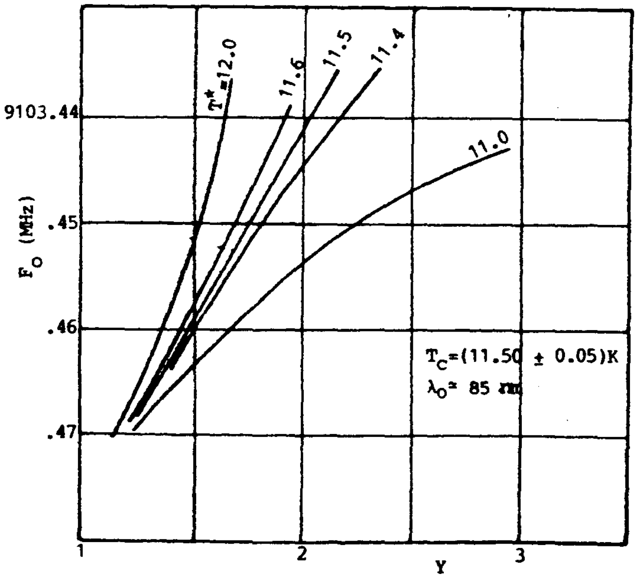

Fig. 1 - Cavity resonance frequency as a function of temperature

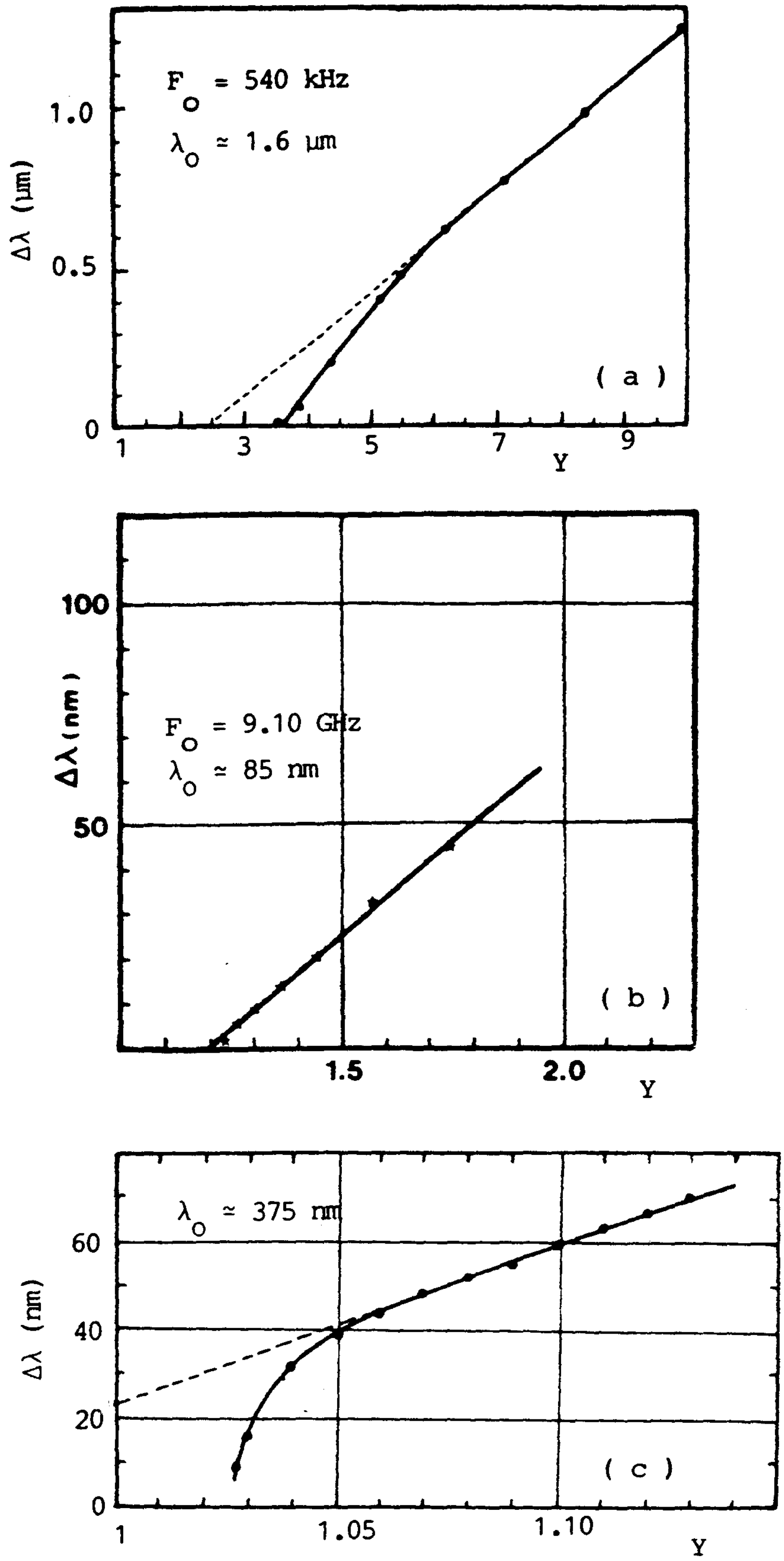

Fig. 2 - Variation of penetration depth versus Y

(a,b) : thermally diffused NbN

(c) : sputtered NbN

$$Y = 1/\left(1 - (T/T_c)^4\right)^{1/2}$$

Figures 2a and 2b show the variation of penetration depth versus Y, respectively at low frequency (inductive method) and high frequency. In fig.3 we report some results obtained for R_S in the temperature range $5.5 < T < 9$ K. On the same figure are plotted the BCS theoretical curve for niobium at 9 GHz, along with the best results we obtained earlier with sputtered NbN (1): it should be emphasized that thermal diffused NbN exhibits a surface resistance which is 2 to 3 times lower than the theoretical value for niobium.

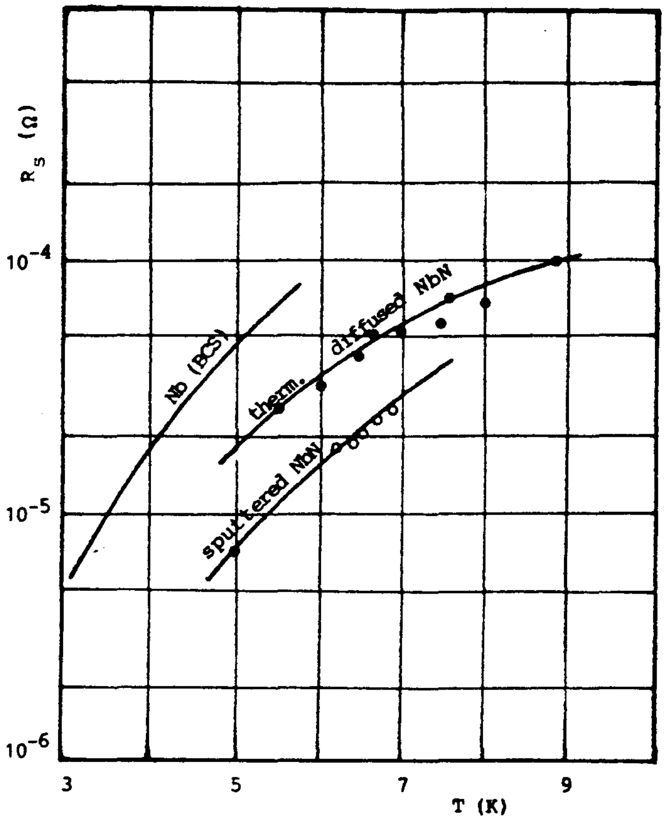

Fig. 3 - Surface resistance as a function of temperature

Following the BCS theory, the energy gap $\Delta(0)$ at $T < T_c/2$ is obtained by plotting R versus (1/T). The value found, $\Delta(0) = 1.4\ kT_c$ for $5.5 < T < 7.5$ K, is relatively low and could be due to weak superconducting regions, likely $NbN_{0.8}O_{0.2}$ (3), or due to N rich NbN existing at surfaces and interfaces. With sputtered NbN, we have measured values for $\Delta(0)/kT_C$ ranging from 1.7 to 2 in the same temperature range and increasing with film thickness (1).

Comparing the results obtained at both low and high frequencies, it is observed that the T_C value is nearly the same. The relatively low value obtained for T_C shows that the δ phase is not stoechiometric. The results have been appreciably improved by operating a very rapid cold quenching from nitridation temperature down to room temperature, or slower nitridation at 600°C. Using a furnace equipped with a fast cooling turbine and under the same elaboration conditions as above, we obtained better results with $T_c \geq 14$ K. Finally, recently improved diffusion conditions allow T_c as high as 16 K (9).

As discussed in (11), the results on NbN, i.e.:

- $\Delta(0)/kT_c$ values decrease with decreasing temperature (10)
- fitted λ_o-values depend on frequency and temperature range (1),

are indicative for weak superconducting regions. These inhomogeneities are the result of the complicated Nb-N-O phase diagram where the strain in nitridation and oxidation plays an crucial role - besides the nucleation (8,11).

The $T_c \simeq 11.5$ K of the NbN can be explained by excess N given by this high pressure ($\simeq 1$ bar) nitridation. The slope changes in fig.2a at $Y < 6$ show then, that even more N (or O) is built into the surface, lowering locally the superconducting interaction. The proximity effect of these regions enhances the slope λ_O for $Y \gtrsim 6$, explaining so the high value of $\lambda_o \simeq 1.6$ µm. For even lower Y and higher frequencies (fig.2b) the fit parameter is reduced further, making the value $\lambda_o = 85$ nm obtained by GHz measurements plausible. The difference between the GHz results, $\lambda_o = 85$ nm for diffused NbN (fig.2b) and $\lambda_o = 375$ nm for sputtered NbN (fig.2c) (1) may be caused by $NbN_{1-x}O_x$ differing due to preparation differences. The slope changes λ_o with Y occurring for this sputtered NbN below $Y \simeq 1.05$ - compared to changes for $Y \lesssim 6$ or $Y \lesssim 2$ (fig.2a,b) - indicate that a continuous spectrum of "T_c values" exist at $NbN\text{-}Nb_2O_5$ surfaces. This is in contrast to Nb where the larger coherence length ($\simeq 10$ nm) yields averaging of NbO_x ($x < 1$) and Nb regions (11). These continuous gap changes at NbN surfaces show up also in Δ- values decreasing with temperature, depending on preparation. Thus high Δ/kT_c - values above 1 are observed for temperatures above $T_c/2$ (1), whereas value below 1 are typical well below $T_c/2$ (10,12). In line with the results summarized in (11), also for NbN there exist no correlation between Δ/kT_c and residual losses. Thus $\Delta/kT_c \simeq 0.7$ has been observed for $R_{res} < 10^{-7}\,\Omega$.

5.- CONCLUSION

The NbN obtained by thermal diffusion of nitrogen has good RF superconducting properties, and in addition it has the merit of easy operation and can be applied to complicated geometries (9). Besides, the Nb_2O_5 oxide layer is thinner than in the case of Nb, shows less serration and greater stability. With optimized diffusion conditions, greater T_c can be expected, giving thus better RF properties. Recent experiments on the Cornell type cavity (13) and on a helix cavity (14) both coated with a NbN film by the technique described above, show that the maximum attainable electric field is essentially the same than that obtained with the uncoated niobium cavity. Yet more experiments are needed both at high and low frequencies, i.e. for high and low β structures, to improve reasonably the maximum field and to check the endurance of the layers against repeated thermal cycles.

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