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ANALYSIS OF TUNNEL BARRIERS IN Nb/Pb JUNCTIONS

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Riassunto

Un certo numero di applicazioni delle giunzioni tunnel superconduttive (STJ) quale ad esempio la rivelazione di particelle ionizzanti mediante STJ nel regime di Giaever richiede che le correnti di perdita sotto il gap siano quanto più basse possibili. La loro minimizzazione puo' essere ottenuta solo con la comprensione dei contributi di corrente. Vari fenomeni, come perdite causate da una cattiva qualità dell'ossido, oppure effetti correlati con l'interfaccia superconduttore-ossido portano ad un comportamento diverso da quello aspettato.

Con lo scopo di caratterizzare e ottimizzare le nostre barriere tunnel, la caratteristica tensione-corrente (I-V) è studiata in dettaglio. A questo scopo sono state realizzate un certo numero di giunzioni Nb/Pb su substrati di vetro Corning e zaffiro. Vari spessori di ossido sono stati ottenuti esponendo all'aria la superficie di niobio per temperature e tempi variabili. Alle diverse ossidazioni corrispondono valori di R_{nn} da 5 a $5 \cdot 10^6$ ohm·mm^2. Le caratteristiche I-V sono state studiate fino a valori di corrente di poco inferiori al danneggiamento irreversibile della giunzione.

Tutte le misure sono state fatte sia a temperatura ambiente che alle temperature dell'azoto e dell'elio liquido e sono state registrate da un sistema di acquisizione dati. Dall'analisi dell'I-V, del Ln(I/V)-V, e della conduttanza sono ricavati alcuni parametri delle barriere.

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ABSTRACT

The application of superconducting tunnel junctions as a particle detector requires the minimization of subgap currents which can be obtained only after the understanding of the different tunnel current contributions. Various phenomena, like leakage caused by low oxide quality, or effects related to the superconductor-oxide interface, make deviations from the expected behaviour. In order to characterize and optimize the quality of our tunnel barriers, the current vs. voltage (I-V) characteristics are investigated in detail.

For this purpose we made a set of Nb/Pb junctions on both Corning glass and sapphire substrates. Various oxide thickness were obtained by air exposure of the niobium layer at different temperatures and times. Different oxidations correspond to R_{nn} values from 5 up to $5 \cdot 10^6$ ohm·mm^2. The I-V characteristics have been investigated up to current values below the irreversible damage of the junctions.

All measurements were carried out at room, liquid nitrogen and liquid helium temperatures and were recorded by a data acquisition system. From the analysis of both the I-V, the Ln(I/V)-V, and the conductance, and from their temperature dependence some tunnel barrier's parameters are determined.

1. - INTRODUCTION

In the past several applications of superconducting tunnel junctions STJ, most of them using the Josephson dc or ac effect, has been developed(1).

In these applications the quasi-particle currents do not play a determinant role. On the contrary in some applications like particle detectors using STJ in the Giaever regime, a very low subgap current is required i.e. leakage currents as low as possible are desired.

In order to understand the different contributions to the currents crossing the tunnel barrier, a detailed study of the barrier structure is required. Since the most common kind of junctions is made of niobium films and the tunnel barrier is built by the niobium native oxides, then it is clear the importance of studying the niobium oxides.

The high chemical reactivity between the oxygen and a niobium clean surface makes the oxidation process fast and difficult to control. Studies on the niobium oxidation process has been done(2,3,4) and different oxides with different behaviours has been found. Several oxidation methods can be used for growing a thin layer of niobium oxide:

- thermal and dry oxidation, where the oxide grown is dominated by the oxygen diffusion,
- plasma oxidation, where a direct injection of oxygen ions makes the tunnel barrier made of oxides more amorphous,
- wet oxidation, which in general, is not used for electronic devices.

After the surface cleaning or at the end of the deposition of a niobium thin film, the oxidation process starts as soon as the oxygen is introduced into the vacuum chamber. The NbO oxide grows very fast and saturates with a mean thickness of 5 Å. The NbO oxide is inhomogeneous, with metallic behaviour, and consists of chunks with the typical NaCl structure.

On the top of NbO grows the dielectric Nb_2O_5 oxide. This oxide is fairly smooth and grows slowly, either in oxygen or in air.

The formation of the Nb_2O_5 oxide starts from NbO_6 octahedral units connected through oxygen atoms at their vertices. This lattice corresponds to the NbO_3 stoichiometry. In order to accomodate the stoichiometry to Nb_2O_5, some octahedral collaps making geometrical planes where they connect through sides instead of vertices. These planes are called crystallographic shear planes CSP and allow differencies in stoichiometry which can be accomodated in regular structures. As a result of the transformation the NbO_6 leads to the growth of microcristalline amorphous Nb_2O_5 with channels which are regions with flat oxygen deficencies. In any case the oxygen diffusion from the Nb_2O_5 to the niobium, leads to the formation of a permanent layer of NbO interface oxide.

Faults, grain boundaries and CSP are extended defects and yelds a set of blocks or columns. Moreover impurity and oxygen vacancies forms localized defects populated by electrons close to Nb_4^+ sites.

As a result the insulation properties of Nb_2O_5 are weakned indeed, the usual simple trapezioidal tunnel barrier can not be used and a more complicated model is needed.

The height of the tunnel barrier of Nb_2O_5 oxide is known to be of about 1.5 eV. Corresponding to extended defects a lower effective barrier is present whereas localized defects can generate resonant tunneling with the lowest affective barrier height. Due to the presence of this three tunnel mechanisms, mainly three ways are in charge of the tunnel current (5,6,7):

- tunnel current through the potential barrier,
- Current flow through the channels in the Nb_2O_5 oxide,
- Resonant current through the oxygen vacancies which generate the resonant tunneling.

Furthermore sometime small metallic links are present and tunnel junctions with barriers made of native oxide, shows true leakage currents.

As far as the superconducting properties of the niobium films are concerned the amorphous growth of the Nb_2O_5 strains the metallic surface enlarging defects in the superficial niobium. This effect promotes an interstitial oxygen diffusion which leads to a worse superconducting surface.

This worse surface superconductivity can be described in terms of a non-BCS density of state and in terms of the presence of states below the gap. It leads in the I-V characteristic to knees at $V \approx \Delta_1 + \Delta_2$ and subgap currents; but a detailed discussion of this topic is out of the purpose of this paper.

In constrast junctions made of artificial barriers, yelds good tunnel properties when made of oxides (Al_2O_3, MgO, ..) grown without difects.

The investigation of the dynamical conductance dI/dV gives informations on the barrier shape but can not identify the different contributions to the total current and the corresponding parameters of the complex model described before.

A way to show up the different contributions to the tunnel current is the use of the logarithmic derivative:

$$g(V) = | d\,Ln(I/V) / dV |$$

according to(5) g(V) is I indipendent; g(V) different from zero shows that the Ohm law cannot be applied. Moreover the logarithmic derivative g(V) shows a different behaviour for different tunnel currents and, the maxima of g(V) indicate at which voltage a new tunnel contribution, opens(5,6). In particular in case of resonant tunneling g(V) shows a sharp maximum for voltages in the range 20-60 meV in this case the positive and negative voltages V^{+}_{max}, V^{-}_{max} at which the maxima appear are symmetric respect to the origin ($| V^{+}_{max} | = | V^{-}_{max} |$). The presence of a broad and higher maximum in the range of 100-500 meV shows the tunnel through the channels. A similar maximum of g(V) at voltages of the order of 1-1.5 eV is due to the presence of the real barrier; the voltage value where this peak appear can be asymmetric ($| V^{+}_{max} | \neq | V^{-}_{max} |$) in case of barriers with an asymmetric shape.

The object of this work is the analysis of the different tunnel contributions in our junctions. These measures, allowing the identification of the contributions to the tunnel currents, are an useful tool for the minimization of subgap currents.

2. FABRICATION AND MEASUREMENTS

Tunnel junctions are made up with our usual technique(8). A Niobium thin film with a thickness of about 5000 Å is deposited by r.f. sputtering. Measurements of resistivity ratio R(300)/R(77) and critical temperature T_c are done as a test of the niobium quality. Sometimes a slighty better quality of the niobium on the sapphire respect to the niobium on the glass substrate is observed.

Niobium patterning is obtained by photolitografic process. Three square junctions with area ranging from 50x50 to 1000x1000 μm^2 are fabricated on each substrate. A soft sputter-cleaning makes a fresh niobium surface, after that, the barrier is obtained by thermal oxidation of the niobium thin film. In this process the oxide is grown by an exposure to the air and the oxide thickness increase with the exposure time and depends on the humidity and temperature.

Later, in order to improve the reliability, the barrier was grown into the vacuum system just after the sputter-cleaning and without breaking the vacuum. A monitored leak of dry oxygen is used in this oxidation process. Unfortunately in our laboratory the counterelectrode is deposited into another vacuum system. An extra layer of oxide grows during the transfer from the sputtering system to the evaporation chamber making these barriers slightly similar to the barriers obtained by air exposure.

Lead counterelectrodes are evaporated by heating lead chunks into a mobibdenum boat. The thickness, monitored by a cristal oscillator is usually on the order of 1 μm.

The fabrication process ends with the photolithography of the second electrode and with the spinning of a thin protective layer of negative photoresist KTFR (Kodak).

All measures were carried out at room, liquid nitrogen and liquid helium temperatures using the usual four contact configuration.

Cryogenic measurements are done wiring our device into a special insert and as soon as the measure finish a special dryer is used in order to avoid the humidity.

Voltages measurements are performed using a low noise True Instrumentation Amplifier (TIA) with an input impedance greather than 10^{13} ohm $\|$10pF and a input current lower than 1pA(9). In our configuration the TIA makes both a preamplification of signals coming from the junction, and a de-coupling between the acquisition system and the low noise electronics. The acquisition is done by a digital plotter HP7090A and data are recorded on floppy disks. Later on a personal computer HP87 computes both the dI/dV, the Ln(I/V) and the g(V).

In order to characterize our devices as a first step the I-V characteristics are measured for bias voltages on the order of few mV. Because of the low bias linearity of junctions in the normal state, this measure gives the low bias resistance R_{nn}.

After the measure of R_{nn}, high bias I-V characteristics are measured up to voltages below the irreversible damage of the junctions. The maximum attained bias depends strongly on the values of R_{nn}; for example junctions with R_{nn} on the order of Mohms can be polarized at bias as high as 1 eV while for junctions with R_{nn} of few ohm the maximum bias is about 50 meV.

4. EXPERIMENTAL RESULTS AND CONCLUSIONS

For several measurements of our Nb/Pb junctions the I-V characteristic, at enough high bias, is asymmetric, for this reason a carefull wiring is needed in order to have always the same configuration.

The asymmetry in the I-V characteristic sometime is found to change while the temperature changes from 300K to 77K, this behaviour can be ascribed to some thermal changes of the barrier.

Measurements of R_{nn} (300K)/R_{nn}(77K) are not able to distinguish the difference in the oxidation process. Measurements at liquid helium are the only true test of the junction quality; only devices showing good superconducting gap structure and low subgap currents are considered.

I-V characteristics of these junctions were numerically acquired, and the calculation of Ln(I/V) was immediately done. The slope of the Ln(I/V) versus V of this junctions, sometime had a slight decrease. In that point, the calcolation of the derivative of Ln(I/V) i.e. the calculation of g(V), as explained in Fig. 1 shows a clear peak.

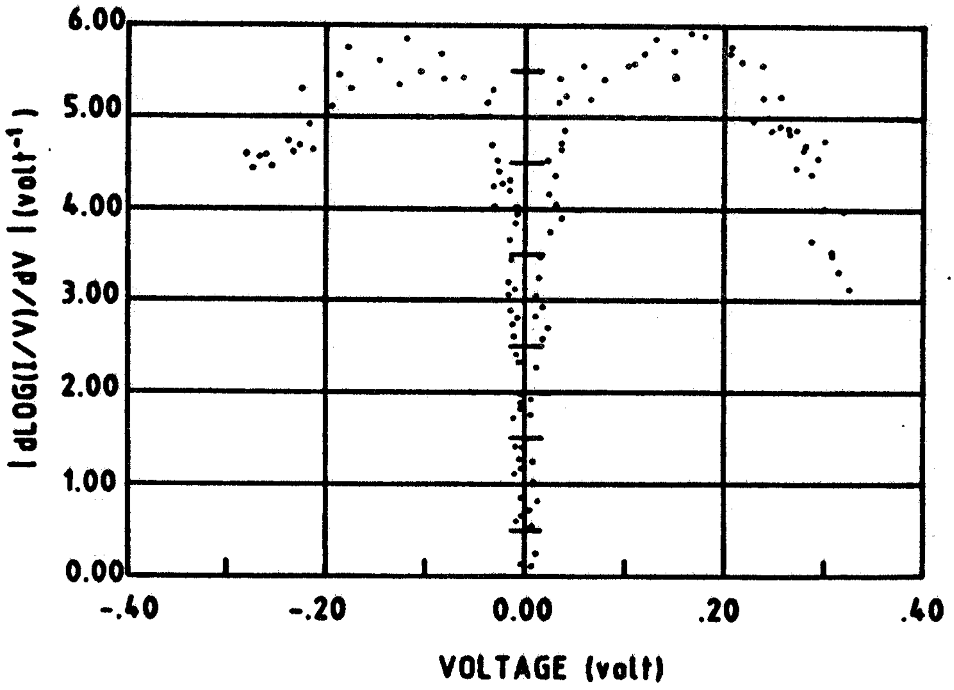

FIG. 1 - g(V) for the junction 198H2M04. The clear symmetric peaks are due to the presence of channels in the barrier with a height of 150 mV.

Many junctions were measured. Several times we found peaks of g(V) for voltages in the range of 100-150 mV, once there was a peak at 30 mV (8). Peaks increase their voltage position changing the temperature from 77K to 4.2K, but, due to the thermal spreading, they disappear for measurements at 300K.

The symmetry of peaks at low bias agree with the symmetry of the low bias I-V characteristic; in contrast as shown in Fig. 2 the observed presence of peaks at -1.1 eV and +1.3 eV again, confirms the asymmetry in the barrier height.

From our data and in agreements with ref. (5,6,7,8,10) it is possible to explain the observed broad symmetric peaks of g(V) for bias in the range of 100-150 mV as the presence of tunnel channels in the oxide. Likewise the peak observed at 30 mV can be explained by the presence of a resonant tunneling in that device.

In conclusion we proved the presence of tunnel channels in the Nb_2O_5 oxide for our junctions. Up to now is not yet possible to relate the observed behaviour of g(V) with the fabrication process. Its beleaved that this goal will be reached soon with further measurements.

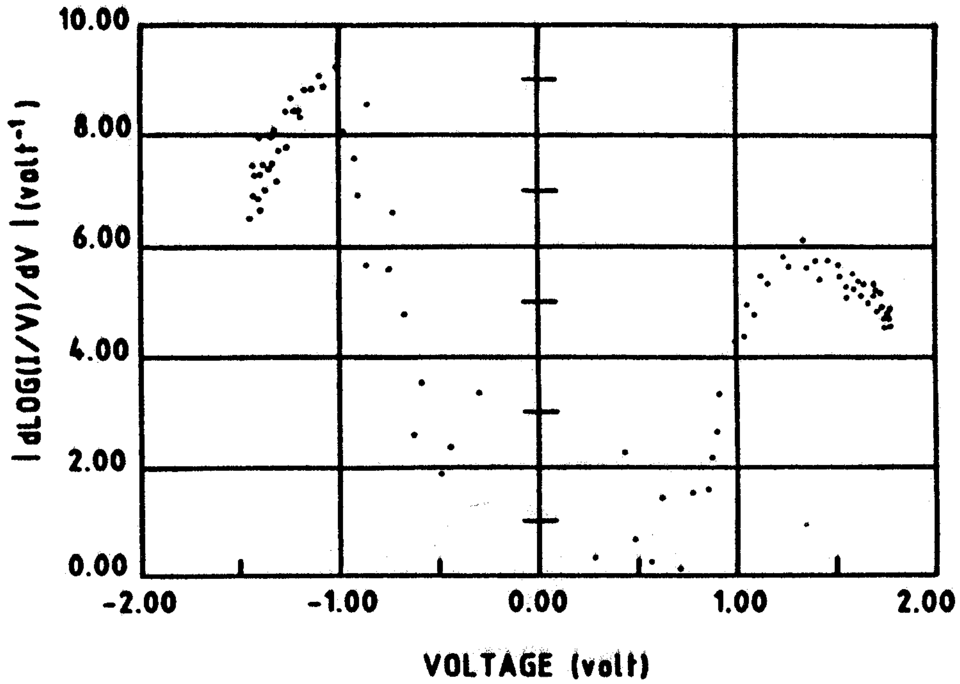

FIG. 2 - g(V) for the junction 212H6L01. The asymmetric peaks are due to the barrier with a height of -1.1 eV and 1.3 eV.

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