Annealing effect on the reproducibility of Josephson Junctions made by ion irradiation.

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Abstract. We have studied the annealing effects on the transport properties of High Tc Josephson Junctions (JJ) made by ion irradiation. Several JJ were measured for different annealing times and the experimental data were compared to numerical simulations. We have successfully used a vacancy-interstitial annihilation mechanism to describe the evolution of the JJ coupling temperature (T_J) and the homogeneity of a JJ array, related to the evolution of the defects density mean value and its distribution width. For sufficient long annealing times (t > 600 min), ΔT_J was significantly reduced. This result appears to be very encouraging for future applications where the spread in JJ characteristics has to be as low as possible.

1. Introduction.

In the last years research on High Tc (HTc) Josephson Junctions (JJ) has been very intense¹. This interest is related not only to the understanding of superconducting properties, but also to the important possibilities for technological applications. It has been suggested that superconducting JJ can be an alternative to semiconductor-based high-speed electronics² for some specific applications. Recently the SCENET roadmap on superconducting electronics³ detailed the efforts needed to be done to turn JJ into a key technology for electronics and detection systems. Most of these applications require close packed arrays of junctions, whose characteristics (critical current Ic and normal state resistance Rn) have to be uniform over large areas and on large scales (from a few to tens of thousands of junctions)⁴. A very promising technique recently developed for the fabrication of HTc JJ is based on ion irradiation⁵. Good quality and highly reproducible⁶,⁷ JJ can be built, thanks to the optimization and control of the fabrication process⁸. Such a JJ is made of a microbridge of HTc material covered by a mask leaving a small aperture across it (of width d ranging from 20 to 100 nm). High-energy ions are used to create a local disorder through the slit: since YBa₂Cu₃O₇⁺ is a d-wave superconductor, disorder decreases the transition temperature of the irradiated area (Tc'). Above Tc' and up to a temperature called T_J, a Josephson coupling takes place between the two superconducting electrodes, leading to a Superconductor/Normal/Superconductor (SNS) JJ.
In general, annealing has been used to increase the long-term stability of JJ$^0$ and it is believed to homogenize the defects profile in the JJ through a diffusive process$^{10}$. Following this idea, JJ annealing can be thought as a mean to achieve a higher reproducibility in an array of JJ made by ion irradiation, but there are no studies related to this question reported in the literature, to the best of our knowledge. Indeed, little work has been done to understand the origin of the spread in the JJ characteristics and how to reduce it to increase the JJ characteristics reproducibility.

2. Simulations and experimental details.

We have measured the transport properties of several HTc JJ made in a 150 nm thick c-axis oriented Y$_{1-x}$Ba$_x$Cu$_2$O$_y$ (YBCO) film grown on SrTiO$_3$ single crystal by Pulsed Laser Deposition. Details of the JJ fabrication are given elsewhere$^6$.$^{11}$. The shadow mask used to define the junctions is made of polymethyl methacrylate photoresist in which 20 nm wide slits are opened by electron beam lithography. The samples were irradiated with 100 keV and 150 keV oxygen ions (O$^+$) with doses $\phi_{100keV} = 5.5 \times 10^{13}$ ions/cm$^2$ and $\phi_{150keV} = 6.75 \times 10^{13}$ ions/cm$^2$. These doses were chosen to induce the same defects density in the center of the barrier, i.e. the same corresponding critical temperature $Tc'$.

We have used SRIM$^{12}$ Monte-Carlo simulations to model the defect density (and therefore the geometry) of a junction made of 150 nm YBCO layers irradiated with 100 keV and 150 keV oxygen ions. This code provides the “Defects Lateral Distribution” (DLD) created by ions impinging a surface, i.e. the local defect density dpa (displacements per atom). To account for the finite width of the slit $d$, we integrate the DLD over the width. We therefore end with a defect profile along the JJ. We then calculate $Tc'$ corresponding to the local dpa, knowing that disorder decreases the critical temperature according to a depairing like Abrikosov-Gorkov law$^{13}$ (since YBCO has a d-wave superconducting order parameter) :

$$\ln \left( \frac{T_{c'}}{T_{c}} \right) = \psi \left( \frac{1}{2} \right) - \psi \left( \frac{1}{2} + \frac{0.14 \ dpa \cdot T_{c}}{0.0375 \ T_{c}} \right)$$

(Eq. 1)

where $T_c$ is the critical temperature of a blank sample, namely 90 K. Experimentally, we have shown that for a given JJ, $Tc'$ corresponds to the defect density in the centre of the barrier ($dpa_0$)$^{14}$. In a JJ array there is an experimental scattering in $Tc'$ ($\Delta Tc'$), corresponding to different $dpa_0$ values for each JJ with a distribution width $\sigma_{dpa0}$. This generates a spread in the Josephson coupling temperature ($\Delta T_J$), which is easier to measure experimentally. Since we have shown that $\Delta Tc'$ is proportional to $\Delta T_J$ over a wide temperature window$^{14}$, we can make a direct comparison between calculations and data.

We then compute the evolution of the DLD during the annealing process through random walk simulations. Two different mechanisms have been considered : defects diffusion and vacancy-interstitial annihilation. For the diffusion mechanism we consider a constant isotropic hopping probability $P_h$ for a defect to move sideway, and (1-2$P_h$) the probability to stay, for each time increment $\Delta t$. For the recombination mechanism the algorithm is the same but the defects have a probability $P_r$ to recombine with a vacancy (and thus disappear) and (1-$P_r$) to remain. Calculations were made typically for 10000 JJ with different hopping and recombination probabilities. Due to the stochastic nature of the process, the defect density in the barrier is not univocally determined. In fact at each time step $t$, the local dpa, and in particular $dpa_0$, is governed by a distribution of probabilities of $dpa_0$ named $P(dpa_0)$, which is characterized by a mean value $\langle dpa_0 \rangle$, which gives the mean $Tc'(t)$ using equation 1 and a width ($\sigma_{dpa0}(t)$) which essentially determinates $\Delta Tc'(t)$. Using the numerical simulations we have studied the evolution of $P(dpa_0)$ as a function of the annealing time, which depends on the physical mechanism acting in the annealing process.

What is interesting for irradiated JJ is that both parameters, $\langle dpa_0 \rangle$ and $\sigma_{dpa0}$ can be directly related to important features of the system. The first one gives the JJ $Tc'$, setting the Josephson regime and the
operating window for devices using this technology. The second determinates the spread in $T_c'$ or the homogeneity of a JJ array, which is a key feature for technological applications.

3. Results

Figure 1 shows the normalized resistance as a function of temperature $T$ of JJ irradiated with 100 keV $O^+$ for different annealing times ($t = 10$, 90', 600' and 3600') at 80°C. The transition corresponds to the Josephson coupling of the junctions $T_j$. Below $T_j$ a typical RSJ like behavior is observed\textsuperscript{15}. $T_c'$ and $\Delta T_c'$, as well as $I_c$ and $\Delta I_c$ were found to follow well the evolution of $T_j$ and $\Delta T_j$, as already observed\textsuperscript{14}. As $t$ is increased, $T_j$ increases and the normal resistance $R_n$ decreases (Figure 3 inset). These results are consistent with a decrease of the effective density of defects in the barrier by diffusion or vacancies annihilation. For short annealing times $\Delta T_j$ increases as $t$ increases but finally decreases for long annealing times ($t > 600'$). For sufficient long annealing times the scattering in $T_j$ is even smaller than the initial spread, i.e. a better array of JJ was achieved.

![Figure 1](image1.png)

**Figure 1:** Normalized resistance as a function of temperature $T$ for Josephson Junctions irradiated with 100 keV $O^+$ ions for different annealing times ($t=10$, $t=90'$, $t=600'$ and 3600').

Figure 2 presents the critical current $I_c$ as a function of $T$ for the same JJ after different annealing times ($t = 10$, $t = 90'$ and $t = 600'$). For $T_c'<T<T_j$, $I_c$ increases as $T^2$ according to the de-Gennes Wertammer model of proximity effect\textsuperscript{16} as already noticed\textsuperscript{6,8}. The high sensitivity of the critical current to the defects distribution and its important temperature dependence makes it especially sensitive to the spread in JJ properties. The scattering in $I_c$ increases for short $t$, following $\Delta T_j$. However, as $t$ increases ($t > 600'$) the spread in $I_c$ starts to reduce. More data are needed to confirm this trend. The $I_cR_n(T_c')$ product as function of $t$ for the JJ irradiated at 150 keV and 100 keV is shown in the insert of figure 2. It is worthwhile noticing that $I_cR_n(T_c')$ product more than doubles its value for $t = 30'$.

Annealing effects as such a low temperature (80°C) is rather surprising given the oxygen diffusion constant in the ab plane reported in the literature\textsuperscript{10,17}, but was already observed in oxygen irradiated films\textsuperscript{10}. We have therefore considered the here above-mentioned mechanisms to understand the experimental results: one based on diffusion, the other one on the interstitial-vacancy annihilation process which may occur efficiently at rather low temperature.
Figure 2: Critical current as a function of the reduced temperature $T/T_J$ of JJ irradiated with 100 keV O$^+$ ions for different annealing times ($t=10'$, $t=90$, and $t=600'$). Arrows indicate the mean value and the spread of $I_c$ at $T_c'$. The insert shows the $I_c.R_n$ product for the JJ irradiated at 100 keV and 150 keV as a function of the annealing time.

The Josephson coupling temperature as function of the annealing time was calculated for the two mechanisms: diffusion and annihilation (Figure 3). The experimental data for low temperature annealing of a JJ array (10 Josephson junctions) was used to adjust the hopping and recombination probabilities. Both mechanisms reproduce very well the experimental data considering a $T_c'$ saturation of around 55K, far from the 90K corresponding to the non-irradiated YBCO. This would indicate that low temperature annealing cannot fully heal the damage done by the ion irradiation and is different from ion irradiated Si where almost all the defects can be healed by the annealing process$^{18}$.

Figure 3: Evolution of the JJ transition temperature as a function of the annealing time for the two mechanisms. Experimental data correspond to the mean value a JJ array of 10 JJ. The insert shows the normal resistance as a function of the annealing time for this samples.
It is worth noticing that from the temporal behavior of $T_c'$ (related to the mean value $\langle d\rho_0 \rangle$) it is not possible to distinguish between the two mechanisms, since both fit rather well the experimental data.

Let us now compute the standard deviation $\sigma_{d\rho_0}$ which controls $\Delta T_c'$. Figure 4 displays the results of the simulations (diffusion dotted line and annihilation solid line) and the experimental data for both 100 and 150 keV irradiated samples.

![Figure 4](attachment:image.png)

**Figure 4:** $\Delta T_J$ spread ($\Delta T_J$) as a function of $T_J$ for the JJ made on the same wafer by 100 keV (closed symbols) and 150 keV (open symbols) oxygen ions irradiation. Shown in solid line are the simulation results for the vacancy-interstitial annihilation model, and in dashed lines for the diffusion mechanism.

The maximum amplitude of $\Delta T_J$ has been adjusted for the simulations to account for the experimental one. The only adjustable parameter is the initial homogeneity of the JJ, which controls the shape of the curves. The dispersion of the initial defects distribution used for the fitting is 3% for the JJ irradiated at 150 keV and 7% for the ones irradiated at 100 keV: these number are consistent with the experimental data ($\Delta T_J^0 = 1.1K$ and 3.1K respectively). This result can be explained by considering the slits size spread as the source of $\Delta T_J$, as previously shown. In no case the diffusion process is able to reproduce the data, whereas the annihilation one does. The reason is rather fundamental. The initial increase of $\Delta T_J$ in both cases is due to the stochastic nature of the processes. But the decrease is related to the efficiency of a mechanism to heal the system. Further details about this will be given shortly.

4. Conclusions.

Ion irradiation is an interesting and promising technique to develop new HTc superconductive electronics using Josephson Junctions. Low temperature (80°C) annealing modifies the characteristics of the JJ: the coupling temperature $T_J$ increases, together with the critical current and the IcRn product. When considering an array junctions, we have shown that long time annealing (10 hours) can reduce the scattering in $T_J$, and therefore in Ic. Numerical simulations clearly show that an interstitial-vacancy mechanism is mainly responsible for this behavior. These results are very encouraging for applications requiring a great number of identical Josephson Junctions.

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