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# Quasiparticle losses at the surface of superconducting tunnel junction detectors

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Superconducting tunnel junctions (STJs) are promising as high energy resolution x-ray detectors. However, the theoretical limit of the energy resolution of STJs has not yet been reached for several reasons. In many cases quasiparticle losses limit the energy resolution. We have investigated STJs with different multilayer structures by means of low temperature scanning electron microscopy. By measuring the quasiparticle lifetime of Nb junctions with and without Ta passivation at the surface, we have identified quasiparticle losses at the surface of nonpassivated junctions as the dominant loss process. The temperature dependence of the quasiparticle lifetime gives information about the loss mechanism. The influence of quasiparticle traps on the effective quasiparticle lifetime is discussed.
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## I. INTRODUCTION

Superconducting tunnel junctions (STJs), consisting of two superconducting electrodes separated by a thin barrier, have been extensively studied for their application as high energy resolution x-ray detectors in the range of 1–10 keV.[1] An x-ray photon absorbed in one of the superconducting electrodes breaks up Cooper pairs thereby creating excess quasiparticles. These quasiparticles can tunnel through the barrier. By measuring the tunneling current the energy of the x-ray photon can be determined since the number of quasiparticles created is proportional to the absorbed energy.[2] In Nb the mean energy needed to create excess quasiparticles is about $\epsilon \approx 1.7\Delta$.[3,4] As the superconducting gap $\Delta$ is in the range of meV, the excitation energy $\epsilon$ is about 1000 times smaller than the excitation energy for electron-hole pairs in semiconductor detectors. Therefore, the intrinsic energy resolution, which is limited by Poisson statistics, is about 4 eV for 6 keV x rays in Nb junctions. This resolution is more than 10 times better than the energy resolution of semiconductor detectors. However, the best resolution obtained so far with Nb junctions is only 50 eV for 6 keV x rays.[5]

There are several reasons that are responsible for this resolution degradation. When quasiparticles get lost before tunneling, they do not contribute to the signal and the ratio of signal to noise gets smaller. Moreover, the statistics of the tunneling process have to be taken into account.[6] If there are local quasiparticle losses, the detector response becomes spatially inhomogeneous which also decreases the energy resolution. In this article we focus on spatially independent quasiparticle losses only.

For future production of STJ detectors it is of interest to find out which quasiparticle loss processes are dominant. Since we can measure only the effective quasiparticle lifetime which contains all loss processes, we measured the quasiparticle lifetimes at various temperatures to distinguish different quasiparticle loss mechanisms by their temperature dependence.[3]

The most important temperature dependent loss process is the recombination of an excess quasiparticle with a thermal quasiparticle to a Cooper pair. For temperatures well below $T_c$ the intrinsic thermal recombination time $\tau_r$ is given by[7]

$$\tau_r = \tau_0' \sqrt{\frac{T_c}{T}} \exp\left(\frac{\Delta(0)}{k_B T}\right), \tag{1}$$

with a material dependent time constant $\tau_0'$ ($\tau'_{0,\mathrm{Nb}} = 2.9 \times 10^{-12}$ s). We will see that in our experiments the recombination of two excess quasiparticles (self-recombination) can be neglected.

Quasiparticle recombination creates a phonon with an energy $E \geqslant 2\Delta$ which can again break up a Cooper pair. On the other hand, the recombination phonons may get lost due to transmission to the substrate (phonon escape). Thus, the effective recombination time $\tau_r^{\mathrm{eff}}$ is given by the following equation ($\tau_b < \tau_\gamma \leqslant \tau_r$):[8]

$$\tau_r^{\mathrm{eff}} = \left(1 + \frac{\tau_\gamma}{\tau_b}\right) \tau_r , \tag{2}$$

with $\tau_\gamma$ the phonon escape time and $\tau_b$ the pair breaking time.

The phonon trapping factor $p = [1 + (\tau_\gamma/\tau_b)]$ is a constant for $T \leqslant 0.3T_c$ (Ref. 7) and in the limit $\tau_\gamma/\tau_b \gg 1$ the phonon trapping factor is proportional to the electrode thickness $d$ (because $\tau_\gamma \propto d$). Theoretically, for the junctions presented in this article we expect $[1 + (\tau_\gamma/\tau_b)] \geqslant 90$.[9]

Another important quasiparticle loss process is quasiparticle trapping. A quasiparticle trap is a region with a lower

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gap, which can confine quasiparticles. This loss mechanism is illustrated in Fig. 1. In the trap quasiparticle states are available below the bulk superconducting gap $\Delta_{\mathrm{bulk}}$. Quasiparticles which reach the neighborhood of the trap can relax to these states in trapping time $\tau_{\mathrm{trap}}$ by inelastic phonon emission. The trapped quasiparticles do not have enough energy to enter the bulk superconductor again except after absorbing a phonon. They leave the trap with escape time $\tau_{\mathrm{esc}}$. The energy of the trapped quasiparticles is converted by recombination into subgap phonons ($\hbar\Omega < 2\Delta_{\mathrm{bulk}}$), which cannot break up Cooper pairs in the bulk material.

If the response of one electrode is eliminated by a trapping layer away from the barrier, there is an additional quasiparticle loss because all excess quasiparticles can only tunnel once and then get lost in the electrode with the eliminated response. The corresponding loss time is the temperature independent tunnel time $\tau_{\mathrm{tun}}$ which is also proportional to the electrode thickness $d$.[10]

## II. EXPERIMENT

The junctions were fabricated by Space Research Organization Netherlands (SRON).[11] Junctions of two different wafers fabricated in a similar way and having comparable multilayer structures (Al6/1 and Al6/3) were investigated. Figure 2 shows the multilayer structure of two different types of junctions of wafer Al6/1.

The response of the base electrode of all the junctions investigated is eliminated (killed) by adding an effective trapping layer (50 nm Ta) away from the barrier. Excess quasiparticles in the base electrode are trapped effectively in that Ta layer and are removed from the tunneling process. In this way tunneling from the base to the top electrode is suppressed. A killed base electrode is advantageous for investigating surface losses because the signal of the top electrode can be measured without being obscured by back tunneling from the base electrode. Thus, in the following we concentrate on the response of the top electrode only.

We measured two junctions of wafer Al6/1 having different multilayer structures (see Fig. 2). The trilayer of A16/1 consists of Ta (10 nm)/$\mathrm{Nb_{top}}$ (180 nm)/Al (5 nm)/$\mathrm{AlO}_x$ ($j_c = 400(\mathrm{A/cm^2})$/Al (10 nm)/$\mathrm{Nb_{base}}$ (180 nm)/Ta (50 nm). The type A junction [Fig. 2(a)] is covered by the

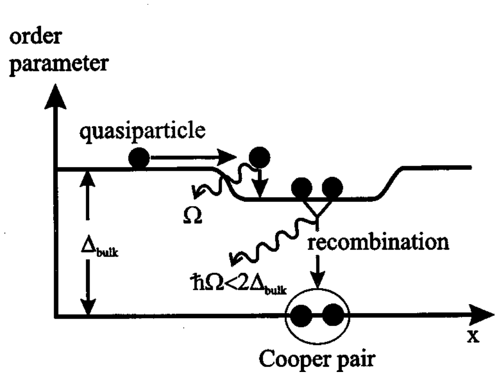


FIG. 1. Quasiparticles may get trapped in regions with a reduced superconducting gap. If trapped quasiparticles recombine, the energy of the recombination phonon is smaller than $2\Delta_{\mathrm{bulk}}$.

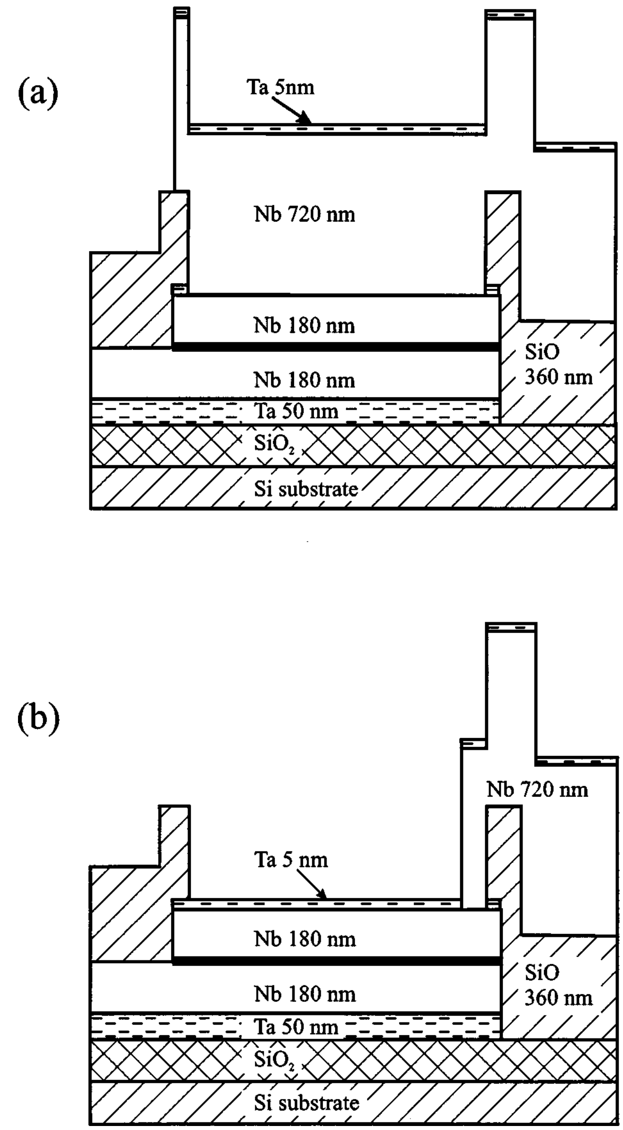


FIG. 2. Schematic layout of two different junctions of wafer Al6/1: (a) type A junction with the top wiring covering the complete top electrode and (b) type B junction with the wiring covering only a small area of the top electrode. The 50 nm Ta layer in the base electrode is an effective quasiparticle trap so the response of the base electrode is eliminated.

top wiring (10 nm Ta/720 nm Nb). Here the upper electrode consists of two different Nb layers which results in a 900 nm total thickness of the upper electrode. The uncovered top electrode of the type B junction is 180 nm thick. The junctions measured are square shaped (type A: $104\times104\ \mu\mathrm{m}^2$; type B: $144\times144\ \mu\mathrm{m}^2$).

One junction of wafer Al6/3 with a special design was measured (junction area: $254\times254\ \mu\mathrm{m}^2$). The trilayer of this junction consisting of $\mathrm{Nb_{top}}$ (200 nm)/Al (5 nm)/$\mathrm{AlO}_x$ ($j_c = 260\ \mathrm{A/cm^2}$)/Al (10 nm)/$\mathrm{Nb_{base}}$ (150 nm)/Ta (50 nm) is half covered by the top wiring (10 nm Ta/800 nm Nb) making the upper electrode of the covered part (type A) effectively 1000 nm thick whereas the top electrode of the uncovered part (type B) is only 200 nm thick. The Ta layer on the top of the uncovered part is only 5 nm thick.

The top electrode and the top wiring are covered with a thin Ta-passivation layer which prevents oxidation of the Nb surface. Oxidized Nb (Ref. 12) is assumed to reduce the quasiparticle lifetime since a layer of normal conducting NbO could be an effective quasiparticle trap. Since Ta has a smaller superconducting gap than Nb, the passivation layer should be thin in order to avoid quasiparticle trapping in the passivation layer.

The quasiparticle lifetimes of all the junctions have also been measured after removing the Ta-passivation layer by reactive ion etching in $SF_6$. By doing this we studied the effect of passivation on the quasiparticle lifetimes. By measuring the quasiparticle lifetimes of junctions with different electrode thicknesses we can also extract some information about surface losses. Since in a thinner electrode more quasiparticles exist close to the surface, surface losses should be more important in a thinner electrode.

The quasiparticle lifetimes were measured by means of low temperature scanning electron microscopy (LTSEM)[13] for various temperatures ranging from 1.75 to 4.3 K. The sample was mounted in a low temperature stage in a standard SEM and irradiated by an electron beam of 5 keV. The penetration depth of 5 keV electrons into the Nb was 120 nm, so that quasiparticles are created only in the top electrode of the junctions. The energy deposition due to the electron beam can be used to simulate x-ray photons, but in contrast to x-ray measurements SEM provides high spatial resolution (about 1 $\mu$m).

During the measurement the junction is current biased in the subgap region of the *I–V* characteristics and the bias voltage shift due to the irradiation is detected. The Josephson current and the Fiske resonances are suppressed by applying a magnetic field parallel to the tunneling barrier.

The exponential temperature dependence of the subgap current of the *I–V* characteristics is used to calibrate the junction temperature. The error of the temperature value is $\Delta T \leqslant 0.1$ K.

For the time resolved measurements the junction is irradiated with short electron beam pulses (typically 100 ns long). The time evolution of the bias voltage shift is amplified and monitored with a digital oscilloscope. These pulses normally show an exponential decay with a time constant equal to the quasiparticle lifetime. As only a small area in the center of the junctions (diameter of about 5 $\mu$m) is irradiated, the influence of quasiparticle losses at the edges of the junctions should be negligible.

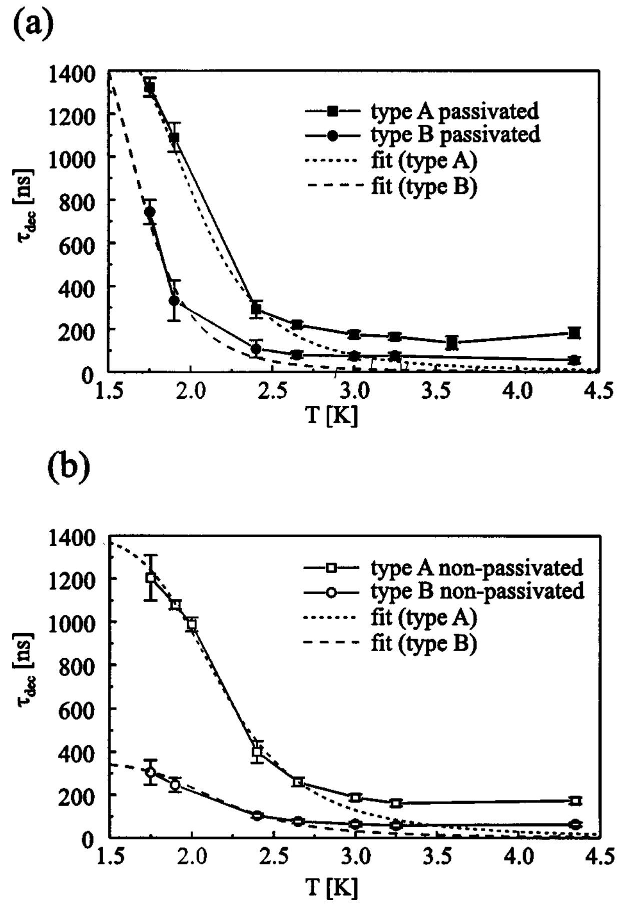


FIG. 3. Measured decay times $\tau_{dec}$ and theoretical fitting curve for sample Al6/1 (a) with Ta passivation and (b) after removing the passivation layer.

## III. EXPERIMENTAL RESULTS

Figure 3 shows the measured decay times $\tau_{dec}$ of sample Al6/1 for various temperatures. For $T > 2.5$ K the decay times are nearly constant. At these temperatures we expect much shorter quasiparticle lifetimes because the effective recombination time $\tau_r^{eff}$ should be in the range of a few nanoseconds. In the limit $\tau_{dec} \leqslant \tau_{RC} = R_D C$ ($C$ is the capacity and $R_D$ the dynamic resistance of the junction) the measured decay time should be given by $\tau_{RC}$. However, $\tau_{RC}$ is shorter than the measured time constants, and we observed that the decay times are independent of the bias conditions. It is interesting that for $T > 2.5$ K the decay times are shorter for the thinner type B junction (also for sample A16/3, see Fig. 4). We cannot explain this phenomenon. In the following we concentrate on the decay times for $T < 2.5$ K which we assume to be equal to the quasiparticle lifetimes.

The quasiparticle lifetimes of the passivated and the nonpassivated type B junction are shorter than the lifetimes of the type A junction. This is expected because of the proportionality of $\tau_r^{eff}$ and $\tau_{tun}$ to the top electrode thickness $d$.

After removing the passivation layer the quasiparticle lifetimes of both types of junctions at $T \leqslant 1.9$ K are shorter than the lifetime of the corresponding junctions with passivation. This effect is more pronounced for the thinner type B junction which indicates that an additional quasiparticle loss occurs at the surface of the nonpassivated junctions.

Figure 4 shows the quasiparticle lifetimes of sample Al6/3. Again we concentrate only on the decay times for $T < 2.5$ K. In this temperature regime the lifetimes at the passivated type A position are even shorter than the lifetimes at the passivated type B position. On the other hand, the lifetimes at the passivated type A position of sample Al6/3 are much shorter than the corresponding lifetimes of sample Al6/1 (passivated type A). Obviously, there is an additional quasiparticle loss at the type A position of sample Al6/3.

After removing the passivation layer the lifetimes at the type B position are reduced, whereas the lifetimes at the type A position remain unchanged.

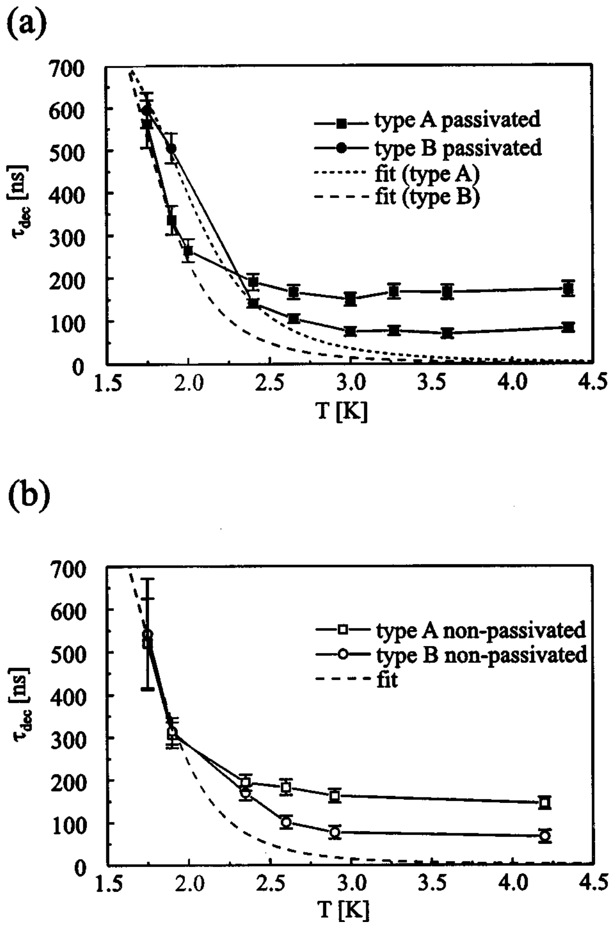


FIG. 4. Measured decay times $\tau_{dec}$ and theoretical fitting curve for sample Al6/3 (a) with Ta passivation and (b) after removing the passivation layer.

## IV. DISCUSSION

In Sec. III we have seen that additional quasiparticle loss occurs at the surface of the nonpassivated junctions. To investigate this effect more quantitatively we adjust a fitting function to the measured curves (Figs. 3 and 4). The following theoretical model is only valid for data points at $T<2.5$ K because of the unknown origin of the measured decay times at $T>2.5$ K.

In our time resolved experiments (see Sec. II) the detector response always shows exponential decay. This exponential decay would not be observable if self-recombination plays an important role because self-recombination is time dependent. For this reason we can neglect self-recombination. This is not surprising because at our experimental temperatures ($k_B T/\Delta \geq 0.1$) the thermal quasiparticle density is supposed to be higher than the excess quasiparticle density. By neglecting self-recombination, all temperature dependent quasiparticle losses can be characterized by an effective recombination time $\tau_{eff}=p\tau_r$, with the intrinsic recombination time $\tau_r$ [see Eq. (1)] and the phonon trapping factor $p$ [see Eq. (2)].

The effective recombination depends in particular on the superconducting gap $\Delta$. Calculating the function $\tau_{eff}=p\tau_r$ for different values of $\Delta$ and $p$, we can show that in first order a small change in $\Delta$ is equivalent to a change in $p$. In the following approximation $c$ describes the variation of the phonon trapping factor ($p$ is fixed) and $\delta$ the reduction of the superconducting gap:

$$cp\tau_r(\Delta) \approx p\tau_r[(1-\delta)\Delta]; \quad \delta \leq 0.3. \tag{3}$$

The evaluation for different values of $c$ shows that $\ln c \approx -6.9\delta$, e.g., a reduction of the superconducting gap of 10% ($\delta=0.1$) corresponds to a reduction of the phonon trapping factor of 50% ($c=0.5$). So we can define an effective phonon trapping factor $p^*=cp$ which includes the reduction of $\tau_{eff}=p^*\tau_r$ by a reduced superconducting gap. To adjust the fitting function unambiguously we do not vary $\Delta$, only $p^*$. This is justified because the error of the approximation of Eq. (3) is smaller than the error bars of the measured decay times.

All temperature independent quasiparticle losses are summarized in the time constant $\tau_c$. Then the effective quasiparticle lifetime $\tau_{life}(T)$ is given by

$$\frac{1}{\tau_{life}(T)} = \frac{1}{p^*\tau_r(T)} + \frac{1}{\tau_c}, \tag{4}$$

with $\Delta=1.47$ meV (from the $I$–$V$ characteristic). We adjust the calculated function $\tau_{life}(T)$ to the experimental data ($\tau_{dec}$) by varying the two fitting parameters $\tau_c$ and $p^*$.

Since we expect that a quasiparticle trap is created at the surface of the nonpassivated Nb junctions through oxidation, we will now discuss the influence of quasiparticle traps on the fitting parameters $\tau_c$ and $p^*$. We assume that the quasiparticle trap is away from the barrier so that trapped quasiparticles cannot tunnel. We distinguish between the limit of an effective quasiparticle trap and the limit of a weak quasiparticle trap. For the classification of quasiparticle traps, which depends on the reduction of $\Delta_{trap}$ with respect to $\Delta_{bulk}$ and on the proximity effect, we assume a trapping layer thickness of a few nanometers. Following the proximity model of Golubov *et al.*[14] the proximity effect can be characterized by the proximity parameter $\gamma_m$. A large $\gamma_m$ corresponds to a large gap reduction.

The scattering processes of an effective quasiparticle trap are illustrated in Fig. 5(a). In this limit $\Delta_{trap}$ is strongly reduced ($\Delta_{trap}<0.5\Delta_{bulk}$) with a proximity parameter $\gamma_m>1$ which results in a small probability that a trapped quasiparticle can leave the trap ($\tau_{trap}/\tau_{esc} \ll 1$). As the quasiparticle density in the trap is enhanced with respect to the bulk material, the recombination time in the trap $\tau_r^{trap}$ is much faster than $\tau_{esc}$ and $\tau_{trap}$. For this reason quasiparticle trapping with subsequent recombination in the trap is an additional loss process. The corresponding loss time $\tau_{trap}$ can be shown to be approximately independent of the temperature in the limit of an effective trap.[14] So the fitting parameter $\tau_c$ can be reduced by an effective quasiparticle trap.

The scattering processes of a weak trap are illustrated in Fig. 5(b). In the limit of a weak trap the reduction of the superconducting gap of the trap is small ($\Delta_{bulk}>\Delta_{trap}$

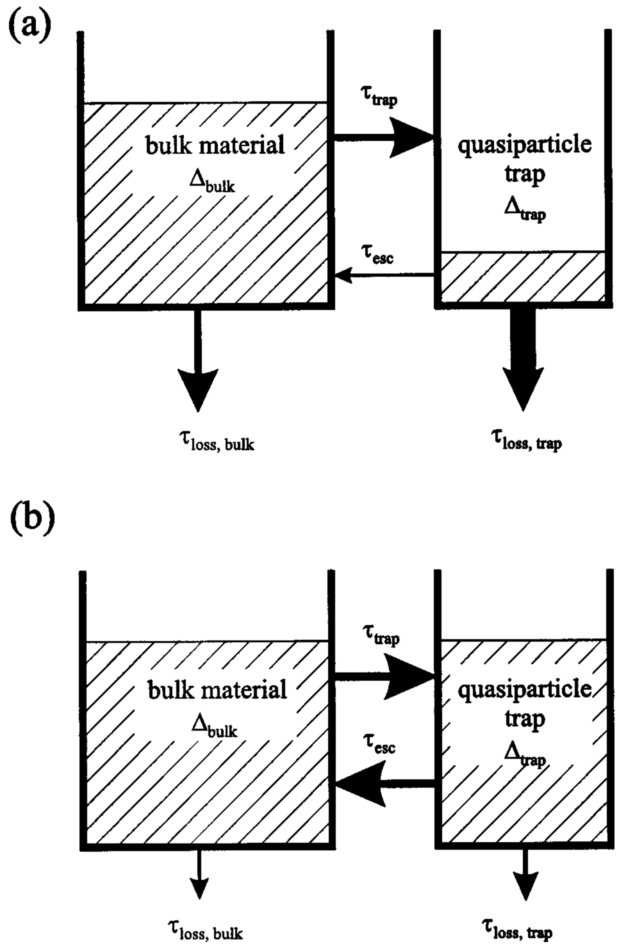


FIG. 5. Simple model to illustrate the scattering rates of (a) an effective quasiparticle trap and (b) a weak quasiparticle trap. The quasiparticles are in two reservoirs (the bulk material and the quasiparticle trap) which they can leave in directions shown by the arrows. A thick arrow indicates that the corresponding scattering rate is high.

$>0.9\Delta_{bulk}$). The escape time $\tau_{esc}$ and the trapping time $\tau_{trap}$ are in the same range and shorter than the effective loss time in the trap and in the bulk material ($\tau_{esc}, \tau_{trap} < \tau_{loss,trap}, \tau_{loss,bulk}$). In this case the quasiparticles of the bulk material and of the trap are in equilibrium and quasiparticle trapping is not an additional loss process. In calculating the effective quasiparticle lifetime of the system, it must be taken into account that the recombination time in the trap is shorter than in the bulk material because of the reduced superconducting gap $\Delta_{trap}$. Using Eq. (3) it is possible to describe the recombination of the whole system with the intrinsic recombination time of the bulk material $\tau_r(\Delta_{bulk})$ and an effective phonon trapping factor $p^*$:

$$\frac{1}{\tau_{eff}} = \frac{a}{p_1 \tau_r(\Delta_{bulk})} + \frac{b}{p_2 \tau_r(\Delta_{trap})} \approx \frac{1}{p^* \tau_r(\Delta_{bulk})}. \quad (5)$$

The two constants, $a$ and $b$, which describe the probability that a quasiparticle stays in the bulk material and in the trap, respectively, can be shown to be approximately independent of the temperature in the limit of a weak trap.[14] Because of Eq. (3) $p^*$ is smaller than $p_1$. Hence $p^*$ can be reduced by a weak quasiparticle trap.

The results of the fitting procedure are shown in Tables I and II. As we have measured the quasiparticle lifetimes only for $T \geqslant 1.75$ K, the saturation of the quasiparticle lifetimes at the value of $\tau_c$ at low temperatures cannot be observed directly. For this reason the errors of $\tau_c$ can be rather large.

The passivated type B junction of wafer Al6/1 has very good properties (see Table I). As the fitting parameter $\tau_c$ of this junction is in the range of the tunneling time ($\tau_{tun} \approx 2$ $\mu$s), $\tau_{tun}$ is the dominant loss time at low temperatures. So almost all quasiparticles contribute to the signal and the energy resolution of this junction is therefore not limited by quasiparticle losses in the top electrode. After removing the passivation layer, $\tau_c$ of this junction is strongly reduced. The fitting parameter $\tau_c$ of the nonpassivated type A junction is also a bit shorter than $\tau_c$ of the passivated type A junction. As this effect is more pronounced for the thinner type B junction, we can assume that this additional temperature independent loss is caused by an effective quasiparticle trap at the surface of the junctions consisting of normal conducting NbO.

The fitting parameters $p^*$ (effective phonon trapping factors) are approximately proportional to the thickness of the upper electrode. This is expected because the phonon trapping factor is proportional to the film thickness $d$. So we can regard the upper electrode of the type A junction as a homogenous Nb film.

The fitting parameters $p^*$ of the nonpassivated junctions are about two times larger than the fitting parameters $p^*$ of the corresponding passivated junctions. This indicates that the Ta-passivation layer is a weak quasiparticle trap.

We will now compare these results with the measurements of sample Al6/3 (see Table II). An important difference to sample Al6/1 is that $p^*$ at the type A position is very small and does not change after removing the passivation. Hence the reduction of $p^*$ is not caused by a surface effect. Probably the interface between the two Nb layers (wiring and top electrode) of the type A electrode forms a weak quasiparticle trap which reduces $p^*$. The fitting parameter

TABLE I. Fitting parameters $\tau_c$ and $p^*$.

| Al6/1 | $\tau_c$(ns) | $p^*$ |
|---|---|---|
| Type A passivation | 1650±150 | 55 ($50 \leqslant p^* \leqslant 80$) |
| Type A nonpassivation | 1400±100 | 95±15 |
| Type B passivation | 1800 ($\tau_c \geqslant 1300$) | 10±2 |
| Type B nonpassivation | 350±70 | 23±7 |

TABLE II. Fitting parameters $\tau_c$ and $p^*$.

| Al6/3 | $\tau_c$ (ns) | $p^*$ |
|---|---|---|
| Type A passivation | $700 \leqslant \tau_c \leqslant 1500$ | 10±2 |
| Type A nonpassivation | $700 \leqslant \tau_c \leqslant 1500$ | 10±2 |
| Type B passivation | 800±50 | 25±3 |
| Type B nonpassivation | $700 \leqslant \tau_c \leqslant 1500$ | 10±2 |

$p^*$ of the type B junction is smaller after removing the passivation. It seems that in this case the oxidation of the Nb surface creates only a weak quasiparticle trap. This would also explain why $\tau_c$ of the nonpassivated junctions is not reduced.

## V. CONCLUSIONS

It is possible to separate different quasiparticle loss processes by measuring the quasiparticle lifetime of junctions with different film thicknesses at various temperatures. In this way we have shown that quasiparticle losses at the surface of Nb STJs reduce the quasiparticle lifetime considerably. For this reason in nonpassivated junctions many quasiparticles get lost before tunneling. A thin Ta-passivation layer at the surface prevents such strong surface losses. However, the measurements also show that the Ta passivation should not be too thick, because we have seen that a 10 nm Ta layer already acts as weak quasiparticle trap.

Quasiparticle losses can also occur at the interface between two superconducting layers. Obviously these losses are very sensitive to the fabrication process.

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