# Nonequilibrium model of the superconducting tunneling junction x-ray detector

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Giaever-type superconducting tunneling junctions are shown to be sensitive to the ionization energy of low-energy x-ray photons. The Mn  $K\alpha$  and Mn  $K\beta$  x-ray photons from a Fe<sup>55</sup> source were detected with an energy resolution of 67 eV full width at half maximum (FWHM) at an energy of 5.89 keV with Sn/Sn oxide/Sn junctions evaporated on fused silica and operated at a temperature of 0.38 K. In this paper, a model of this detector is presented. The basic features are well described by dynamical Cooper-pair breaking and the relaxation mechanisms of nonequilibrium superconductivity. Quasiparticle diffusion had to be included in order to explain the observed data. The solutions of the four coupled Rothwarf-Taylor equations, one quasiparticle equation and one  $2\Delta$ -phonon equation for each of the two films, agree reasonably well with measured pulse shapes.

#### I. INTRODUCTION

Peturbing a superconductor by an external source of energy will in general lead to a nonequilibrium distribution of quasiparticles and phonons. Nonthermal quasiparticle populations have been produced in the previous years, either by injecting quasiparticles directly into a superconducting film using the double-junction method, or by exposing the films to photons or phonons. The quasiparticle-injection method was introduced in 1962 by Ginsberg<sup>1</sup> in the first experimental determination of the quasiparticle recombination time. The first to illuminate superconducting films with a laser was Testardi.<sup>2</sup> In the latter experiment, the effects could not be explained simply in terms of heating alone. It was suggested that some excess quasiparticles were produced by dynamically breaking Cooper pairs. This work stimulated a series of investigations, both experimentally and theoretically (for a review on nonequilibrium phenomena in superconductivity see Refs.  $3-5$ ).

The perturbed distributions relax to their equilibrium values owing to various relaxation processes. Both quasiparticles and phonons are in general coupled, as two quasiparticles recombining to a Cooper pair produce an excess  $2\Delta$  phonon, which in turn can break a Cooper pair. This leads to the phonon-trapping effect, first discussed by Rothwarf and Taylor.<sup>6</sup> They proposed two coupled energy-independent rate equations, one for the quasiparticles, and the other for the  $2\Delta$  phonons, and they could account for the longer recombination times observed in the experiments than expected from theory.

In this paper, the relaxation of excess quasiparticles and phonons produced by the absorption of a 6-keV x ray in the films of a superconducting tunneling junction is discussed. The results presented in this paper are part of an investigation to use superconducting tunneling junctions as ionizing radiation detectors with high-energy resolution. The experimental results of this investigation have been published elsewhere.<sup>7-10</sup> Basically, the idea is to make use of the fact that the lowest-lying quaisparticle states are separated from the superconducting ground state by an energy gap of the order of meV. This is a thousand times less than in conventional semiconductor detectors. By illuminating Sn/Sn Oxide/Sn junctions with a  $^{55}$ Fe source, a best energy resolution of 65 eV at 5.89 keV was obtained. The charge collected was  $2.5 \times 10^6$  electrons, yielding a minimal ionizing energy of  $\sim$  3 meV. Extrapolating to vanishing electronic noise, the intrinsic resolution of our samples approaches  $0.7\%$  at 5.89 keV. This is an order of magnitude worse than expected from the large number of primary charge produce and is believed to be due to the granuarity of the films. $8,1$ 

The absorption of an x ray in a superconducting film is also interesting from the point of view of the nonequilibrium effects produced. The perturbation induced by the x ray is short and localizable, and has the advantage of a well-defined energy being deposited in the film. Laser experiments have been performed with sufficiently shor laser pulses,  $5<sup>11</sup>$  but they lacked the precise determination ene<br>ave<br>,11 of the deposited energy. The energy of a 5.89-keV x-ray photon expressed in SI units is  $9.84 \times 10^{-16}$  J and is in general much smaller than the energies deposited by the individual pulses of the laser experiments. The random emission of the x-ray photons from the radioactive source prohibits the use of lock-in techniques. Owing to this, great care must be taken to minimize the relaxation losses and to optimize the tunneling process in the junction.

In Sec. II, some basic experimental results are presented, in order to point out the nonequilibrium character of the detector. The tunneling of quasiparticles between two superconducting films, separated by a thin oxide barrier, is treated in some detail in Sec. III. The various relaxation processes are discussed in Sec. IV, where the four coupled equations of the model, one quasiparticle and one  $2\Delta$ -phonon equation for each film, are presented. The solutions are compared with experimental pulse shapes in Sec. V.

#### II. EXPERIMENTAL RESULTS

The first to consider using superconductors are detectors for ionizing radiation were Wood and White<sup>12, 13</sup> in

1969. In the last few years additional results<sup>7-10,14-23</sup> have been published. As has been pointed out above, the potentially high resolving power of superconducting detectors arises from the fact that in a superconductor the lowest-lying excited electronic states (quasiparticles) are separated from the ground state (Cooper pairs) by a gap  $\Delta$ of the order of <sup>1</sup> meV. Hence, a thousand times more "free" charge is expected to be produced compared to a conventional semiconductor detector, which has a gap of the order of <sup>1</sup> eV. As the resolution of a detector is limited by the statistical fiuctuations in the number of charge carriers produced, superconducting detectors could, in principle, offer energy resolutions more than an order of magnitude better than the best semiconductor detectors. In addition, excited phonons are not a priori lost in a superconductor, as their energies (Debye energy  $\sim 10$  meV) can be considerably larger than the gap  $\Delta$ , breaking pairs and contributing to the production of free charge in the detector. In a Sn-juntion detector, one expects the minimal ionizing energy  $\varepsilon$  to be of the order of the superconducting gap of tin, which is  $\Delta_{Sn} = 0.58$  meV. Hence, assuming no losses, the ideal resolution at an energy of 5.89 keV would be

$$
\frac{\Delta E_{\text{FWHM}}}{E}(\text{Sn}) = 2.35 \left[ \frac{0.58 \text{ meV}}{5.89 \text{ keV}} \right]^{1/2} = 0.07\% .
$$

When using a superconducting material as a detector, the question to be considered is how to measure the number of excess quasiparticles induced by the ionizing radiation. Since the work of Giaever,  $24-26$  superconducting tunneling junctions have often been used in studying the physics of quasiparticles<sup>3,4</sup> When applying a voltage difference  $U_B$  between the two films, a thermal current  $I_{\text{th}} \sim \exp(-\Delta/kT)$  will flow across the insulating barrier. In order to operate in the Giaever-type mode, the Josephson current has to be suppressed by applying an external magnetic field parallel to the oxide barrier ( $\sim$  100 G). If the number of quasiparticles is raised owing to the perturbation produced by the x ray, the tunneling current will increase, ideally in proportion to the energy deposited in the films. This implies that the disturbance caused by the transient event should not be so large that the microscopic features of the superconductor are altered. Especially excluded is the superconductor to normal-conductor transition, as this will only act as an energy discriminator.

The geometry of the junctions is shown in Fig. 1. The films were deposited onto the substrate (fused silica) by thermal evaporation and by use of the shadow technique. The pulse-height spectrum of a junction is shown in Fig. 2. The most striking property is the doubling of the  $K_{\alpha}$ - $K_{\beta}$  structure of the <sup>55</sup>Fe source. This is due to the absorption of x rays in either of the films of the junction. As the thickness of the films is much smaller than the attenuation length of the x rays ( $\sim$  6  $\mu$ m), the number of events per film depends on the thickness of the respective films. The relative positions of the two  $K\alpha$  peaks in the spectrum is determined by the different relaxation losses in each film and by the corresponding rise times of the respective pulses. In Fig. 3, both, the pulse height and the rise time are combined in a two-dimensional plot, showing



FIG. 1. Geometry of the junctions

an extended view of the total absorbing peaks of the two films. The rise time of the thinner film is faster because of the shorter tunneling time. The events with lower pulse heights and larger rise times are due to diffusion of quasiparticles produced by x-ray absorption in the current leads. As these quasiparticles first have to diffuse into the tunneling region (the overlapping area of the films), the rise times of the corresponding pulses are larger. The pulse heights are smaller because of the smaller fraction of excess quasiparticles capable of tunneling and also because of the increased self-recombination losses in the narrower current leads. It was initially surprising to observe pulses from either film with the same sign at a fixed biasing voltage. Electrons always tunnel from the film at the higher potential energy to the film at the lower potential energy, but, as we will see in Sec. III, in a superconducting tunneling junction it is possible to exchange quasiparticles in both directions.

Some temperature-dependence measurements were performed to demonstrate the nonequilibrium character of the detector. This dependence was measured both in vacuum and with a helium film on the junction. A sequence of both pulse-height and rise-time spectra with increasing bath temperature is shown in Fig. 4. We observe the following features.

(i) The pulse height reduces with increasing temperature. This reflects the increasing thermal quasiparticle recombination losses at higher temperatures.

(ii) The rise-time separation of the two films decreases at higher temperatures, reflecting the increasing domi-



FIG. 2. Pulse-height spectrum of a Sn-junction detector. The doubling of the  $K\alpha$ - $K\beta$  structure is due to the absorption of x rays in either films. The low-energy tail is from x rays absorbed in the current leads.



FIG. 3. Two-dimensional display of pulse height versus rise time, presenting a close-up view of the total absorbing peaks of the two films.



FIG. 4. Temperature dependence of the pulse-height and rise-time spectra.

nance of the faster recombination times at higher temperatures.

In Fig. 5, the accumulated charge is plotted versus temperature for x rays absorbed in either films ( $K\alpha$  peaks). In one measurement (open symbols) the junctions were in vacuum, and in the other case (solid symbols) a helium film covered the junctions. The pulse heights of the signals are smaller in the presence of a helium film, owing to the enhanced probability for phonons to escape. This shows that phonons play an important role in superconducting junctions. As has been pointed out above, the recombination of two quasiparticles yields a  $2\Delta$  phonon, which in turn can break a Cooper pair. The effective lifetime of the quasiparticles is hence strongly influenced by the rate of the  $2\Delta$  phonons escaping into surroundings. From this one can expect the top layer of the junction (film 2) to be more sensitive to the presence of a helium film [the lower film (film 1) is coupled to the substrate and not covered by the helium film]. In Fig. 6, the ratio of the pulse heights of film 2 (top film) to the corresponding pulse heigts of film <sup>1</sup> (lower film) is plotted against increasing temperature. The larger decrease of this ratio in the presence of a helium film indicates the importance of phonon trapping in our detector. In the model presented in Sec. IV, phonon trapping will be taken into account by introducing an effective quasiparticle recombination time  $\tau_{\text{eff}}$  (which is reasonable in the case of a stationary perturbation), but will follow directly owing to the coupling of the quasiparticle and  $2\Delta$ -phonon equations.

# III. TUNNELING PROCESSES IN SUPERCONDUCTING TUNNELING JUNCTIONS

The tunneling of electrons between two metallic films separated by an insulating barrier can be described by using first-order perturbation theory. An electron state  $|k\rangle$ in metal 1 overlaps with an electron state  $|1\rangle$  in metal 2, and hence a transition from state  $|k\rangle$  in metal 1 to state



FIG. 5. Temperature dependence of the  $K\alpha$  pulse height for x rays absorbed in both films. Solid symbols are with a helium film on the junction, open symbols without.



FEG. 6. Temperature dependence of the pulse-height ratio of film 2 to film 1, demonstrating the importance of phonon escape. This ratio decreases faster with temperature, when the junction is covered with a helium film, due to enhanced phonon escape in the upper film.

 $|1\rangle$  in metal 2 can occur. The amplitude for this transition is given by the matrix element  $T_{kl}$ , and  $T_{lk}^*$  for the corresponding reverse transition. In thermal equilibrium, the coherent nature of the electronic excitations does not affect the transitions, as the Bardeen-Cooper-Schrieffer (BCS) coherence terms cancel in the corresponding expression for the tunneling current. As has been pointed out already by Giaever and Megerle,<sup>26</sup> one only has to substitute the supereonducting quasiparticle density of states into the expression for the tunneling current in a normalconducting junction. We can therefore express the tunneling Hamiltonian in terms of the normal electron opera $tors: <sup>3,27,28</sup>$ 

$$
H_T = \sum_{k,l} \left( T_{kl} c_{l1}^{\dagger} c_{k1} + T_{lk}^* c_{k1}^{\dagger} c_{l1} \right) \,. \tag{1}
$$

The first term destroys an electron state  $|k \rangle$  in metal 1 and creates an electron state  $|l\uparrow\rangle$  in metal 2, and the second term creates  $|k \rangle$  in metal 1 and destroys  $|l \rangle$  in metal 2, where  $k$  and  $l$  denote the momenta and  $\uparrow$  denotes the spin direction of the electron. The Hamiltonian (1) yields the thermal current-voltage characteristic for both normal-conducting and and superconducting tunneling junctions (for details on thermal current-voltage characteristics see Ref. 29).

In order to discuss the tunneling of electrons in superconducting tunneling junctions more generally, we transform (1) into an Hamiltonian describing more accurately the exchange of quasiparticles between the two superconductors. $3$  We substitute the normal electron operators  $c_{k}$  by the quasiparticle operators  $\gamma_{k}$ , defined by Bogliubov-Valatin transformation:

$$
c_{k_1} = u_k^* \gamma_{k_1} + v_k \gamma_{-k_1}^{\dagger}, \nc_{-k_1}^{\dagger} = -v_k^* \gamma_{k_1} + u_k \gamma_{-k_1}^{\dagger},
$$
\n(2)

where  $u_k$  and  $v_k$  are the BCS distribution functions for the pair states:  $|v_k|^2$  is the probability that  $(k \tau, -k \tau)$  is

occupied, and  $|u_k|^2$  the probability that  $(k \uparrow, -k \downarrow)$  is empty.

The Bogliubov-Valatin transformation diagonalizes a special representation of the BCS-Hamiltonian (the model Hamiltonian<sup>28</sup>) which includes in a natural way the single-particle excitations of a BCS superconductor. The operator  $\gamma_{k_1}^+$  corresponds to putting with certainty a single electron into state  $|k\rangle$  and leaving with certainty the other state of the pair,  $|-k \rangle$ , empty. The operator  $\gamma_{k_1}^+$ can therefore be identified as a quasiparticle-creation operator. This excitation blocks that specific pair from participation in the coherence and increases the system energy accordingly. Substituting the normal electron operators  $c_k$  with the quasiparticle operators  $\gamma_k$ , one obtains the superconducting tunneling operator.

$$
H_T^S = \sum_{k,l} T_{kl} (u_1^* \gamma_{11} + v_1 \gamma_{-11}^{\dagger})^{\dagger} (u_k^* \gamma_{k1} + v_k \gamma_{-k1}^{\dagger}) + \text{H.c.}
$$
\n(3)

Carrying out the multiplication, one obtains four terms, each of which can be treated independently. This is permitted because the initial and final states are different for all terms, and no cross products appear in squaring the matrix elements.<sup>3</sup>

The Hamiltonian  $H_T^S$  is then

$$
H_T^S = \sum_{k,l} T_{kl} u_l u_k^* \gamma_{l1}^{\dagger} \gamma_{k1} + \text{H.c.}
$$
 (4a)

$$
+\sum_{k,l}T_{kl}u_{l}v_{k}\gamma_{l1}^{\dagger}\gamma_{-k1}^{\dagger}+\text{H.c.}
$$
 (4b)

$$
+\sum_{k,l}T_{kl}v_l^*u_k^*\gamma_{-l_1}\gamma_{k\uparrow}+\text{H.c.}
$$
 (4c)

$$
+\sum_{k,l}^{k,l}T_{kl}v_{l}^{*}v_{k}\gamma_{-l1}\gamma_{-k1}^{\dagger}+H.c.
$$
 (4d)

We will now discuss each of these four terms.

Term (4a). This term destroys a quasiparticle with the energy  $E_k$  in superconductor 1 and creates a quasiparticle with energy  $E_l$  in superconductor 2 [Fig. 7(a)]. As the coherence factors cancel<sup>3,8</sup> one obtains the familiar expression for the thermal tunneling current:

$$
I = G_{NN} \int_0^{\infty} \rho(\Delta, E) \rho(\Delta, E + eU)
$$
  
 
$$
\times [f(E) - f(E + eU)]dE,
$$

where  $G_{NN}$  the normalconducting conductance of the junction, and  $\rho(\Delta, E)$  is the reduced quasiparticle density of states.

Term (4e). The term (4c) destroys quasiparticles on both sides. Energy is not conserved in this process, and this term hence does not contribute to the tunneling current.

Term (4b). Due to (4b), quasiparticles on both sides of the barrier are created by the breakup of a Cooper pair. This process requires a minimum voltage  $V=2\Delta/e$  to overcome the binding energy of the pair. Again, in thermal equilibrium the BCS coherence factors drop out. This term is responsible for the sharp rise in the  $I-V$ characteristic at voltages  $> 2\Delta/e$ .

Term (4d). The interesting term of  $H_T^S$  is the term



FIG. 7. Tunneling processes in a junction with both films superconducting. (a) is due to term (4a) and (b) due to (4d). In both cases the flow of electrons is in the same direction. This leads to the observation that x-rays absorbed in either of the films yield a signal with the same sign.

$$
v_l^* v_k \gamma_{-l+1} \gamma_{-k+}^{\dagger}.
$$

This term creates a quasiparticle in film <sup>1</sup> and destroys a quasiparticle in film 2. As is apparent from Fig. 7(b), the electron tunnels from <sup>1</sup> to 2 (i.e., from the film with the higher potential to the one with the lower potential). This flow of quasiparticles opposite to the tunneling direction of the electrons is possible through the intermediary of the Cooper pairs.

Owing to the overlap of the wave functions of quasiparticles across the thin barrier, electrons can be exchanged between Cooper pairs across the oxide barrier (this is not to be mistaken as the Josephson effect, which is the tunneling of Cooper pairs across the barrier, and is suppressed in our application by the external magnetic field applied). In thermal equilibrium, and for equal gaps, the transition rates of the two terms (4a) and (4d) are identical and lead to an additional factor 2 in the tunneling current.

With the processes shown in Figs. 7(a) and (b), one can explain why x rays absorbed in either of the films of a superconducting junction yield pulses with the same sign. If an x ray is absorbed in the film at the higher potential, the process in Fig. 7(a) is responsible for the signal. In the other case, the electrons will tunnel according to Fig. 7(b). A consequence of these two tunneling processes is the back tunneling of quasiparticles, shown schematically in Fig. 8. Excess quasiparticles are exchanged back and forth, with the electrons always tunneling in the same direction, until the  $2\Delta$  phonons, resulting from quasiparticle recombinations, escape the films. The first to demonstrate this amplification effect was  $Gray^{3,32}$ . He measured a gain of 4 in the tunneling current by using the quasiparticle injection method in double Al junctions (three aluminium films separated by two insulating barriers). By increasing the temperature, the gain of this superconducting transistor decreases, owing to the increase of thermal recombination.

In our detector, more tunneling electrons are measured for primary quasiparticles produced because of this back tunneling effect. The intrinsical resolution of the super-



FIG. 8. Back tunneling of quasiparticles. If the tunneling rate is faster than the effective quasiparticle lifetime, excess quasipsrticles can be recycled, leading to intrinsic amplification of the tunneling current.

conducting detector, however, will not be improved, as the resolution is determined by the number of primary excitations. The criterion necessary for this intrinsic amplification to occur is that the tunneling rate has to be larger than the effective quasiparticle relaxation rate. At higher temperatures the quasiparticle loss is dominated by thermal recombination, whereas at lower temperatures, quasiparticle diffusion out of the tunneling region and self-recombination are responsible for the quasiparticle losses.

# IV. NONEQUILIBRIUM MODEL OF THE SUPERCONDUCTING DETECTOR

The relevant relaxation processes in a perturbed superconductor are as follows.

(i) Recombination of quasiparticles to Cooper pairs under emission of a phonon with energy greater than  $2\Delta$ .

(ii) Pair breaking by  $2\Delta$  phonons.

(iii) Inelastic scattering of quasiparticles and phonons.

(iv) Breakup of a  $2\Delta$  phonon.

(v) Transmission of phonons from the film into the surroundings.

(vi) Diffusion of quasiparticles in the film.

The rates of these relaxation processes depend strongly on the various parameters of the detector: temperature, geometry of the films, coupling of the films to the substrate, and structure of the films. An additional important quantity in our application is the tunneling rate  $\tau_{\text{tun}}^{-1}$ of quasiparticles into the opposite film. In earlier nonequlibrium junction experiments,  $\tau_{\text{tun}}^{-1}$  was usually neglected, as the perturbing energy could be adjusted accordingly to achieve a good signal-to-noise ratio. In our case, this of course not possible. The tunneling time  $\tau_{\text{tun}}$  can be expressed in terms of the normal-conducting properties of the junction and the thickness of the film:<sup>1</sup>

$$
\tau_{\rm tun} = R_N e^2 N_0 A d \ ,
$$

where  $R_N$  is the normal-conducting resistance of junction (slope of *I*-*V* characteristic at  $U > 2\Delta/e$ ), *e* is the electron charge,  $N_0$  is the single-spin density at Fermi energy,  $A$  is the area of junction, and  $d$  is the thickness of junction film. The resistance  $R_N$  is a property of the insulating barrier and is temperature independent. The dependence of  $\tau_{\text{tun}}$  on the area A (overlap of the two films) is only formal, as it cancels by the inverse dependence of  $R_N$  on A. The tunneling rate depends inversely on the thickness  $d$  of the film, owing to the fact that the tunneling rate is proportional to the product of the transmissivity and the frequency of hitting the barrier. The latter depends inversely on the thickness  $d$  (assuming the mean free path of a quasiparticle to be at least of the order of the film thickness). The parameterized value for tin is

$$
\tau_{\rm tun}(S_{\rm n}) = (2.2 \,\, {\rm nsec})[R_N(\Omega)][A(\mu{\rm m}^2)][d(\mu{\rm m})].
$$

The tunneling times of our detectors varied between <sup>1</sup> and  $10 \mu$ sec.

In an important theoretical paper by Kaplan et  $al.$ <sup>33</sup> the relevant relaxation rates have been calculated for a variety of superconductors. They have not considered phonon trapping in their work, which has to be taken into account when comparing their values with experiment. We have used their calculated thermal relaxation rates. The Rothwarf-Taylor approximation has been adopted; hence the energy dependence of all distributions is omitted. The films are assumed to be homogeneous and isotropic

Before the various relaxation processes are discussed, we comment briefly on the cascade of the "hot" electrons (energy on the order of eV) down to the "cold" quasiparticle (energy on the order of meV). Initially, atoms are ionized along the trajectory of the potoelectron, yielding electrons with an energy of several eV. Owing to the direct electron-electron interaction and inelastic electron-phonon scattering, the mean energy per electron reduces during the cascade. This cooling process is fast ( $\sim$ psec). When the mean energy per electron approaches a few meV, the superconducting coherence becomes effective. At low temperatures, inelastic quasiparticle scattering (under emission of phonons) and phonon pair breaking will be dominant. According to Kaplan et  $a l$ , <sup>33</sup> inelastic quasiparticle scattering is faster than quasiparticle recombination for temperatures  $T < 0.2T_c$ , yielding a population of excess quasiparticles with a mean energy  $({\sim}\Delta)$  before recombination sets in. At the lowest energies, we can neglect inelastic scattering processes, and assume all quasiparticles to have the same mean energy  $\Delta$ . As has been pointed out by Kaplan et al., direct electron-electron scattering is significant only for low quasiparticles energies. This term does not scatter the quasiparticles out of our energy range of interest  $({\sim}\Delta)$  and is therefore omitted. We also neglect the branch mixing time  $\tau<sub>O</sub>$  describing the relaxation of the momentum branch imbalance<sup>3,34</sup> [the hot electrons (energy on the order of eV) all have a momentum larger than  $k_F$ , which would lead to an apparent branch imbalance, but during the cascade, the branches are equally populated due to the inelastic quasiparticle-phonon scattering].

Phonons are virtually absent at low temperatures  $(T < 0.5$  K for Sn junctions) because of the fast phonon pair breaking and the negligible thermal quasiparticle recombination rate. The phonons are nevertheless important, as their escape into the surroundings presents the ultimate decay channel for the deposited ionizing energy.

The scenario of our model is as follows. An x ray is either absorbed in film 1, producing an initial excess population of quasiparticles,  $N_1^0$ , and  $2\Delta$  phonons,  $N_{\omega_1}^0$ , in film 1, or is absorbed in film 2 with the corresponding initial values  $N_2^0$  and  $N_{\omega_2}^0$ , respectively. The time evolution of the excess numbers in either film depends on the various relaxation parameters and on the specific nature of the coupling of these excess quantities.

## A. Thermal quasipaxticle recombination

The loss of excess quasiparticles owing to recombination can be separated into two contributions.

(i) Thermal recombination, where the recombination partner is a thermally excited quasiparticle. This rate is very sensitive on temperature because of the exponential temperature dependence of the thermal quasiparticles density.

(ii) Self-recombination, where the recombination partner is another excess quasiparticle.

If we restrict ourselves, for the moment, to film 1, and take into account recombination, tunneling, and phonon escape only, one derives the following rate equations for the excess number densities:

$$
\frac{d}{dt}n^* = 2\tau_B^{-1}n^*_{\omega} - (2Rn_T)n^* - R(n^*)^2 - \tau_{\text{tun}}^{-1}n^*,
$$
\n(5)\n
$$
\frac{d}{dt}n^*_{\omega} = -\tau_B^{-1}n^*_{\omega} + \frac{1}{2}(2Rn_T)n^* + \frac{1}{2}R(n^*)^2 - \tau_{\gamma}^{-1}n^*_{\omega}.
$$

where R is the recombination coefficient,  $\tau_B^{-1}$  is the phonon pair breaking rate,  $\tau_{\text{tun}}^{-1}$  is the quasiparticle tunneling rate,  $\tau_{\gamma}^{-1}$  is the phonon escape rate,  $n_{T}$  is the density of thermal quasiparticles,  $n^*(t)$  is the density of excess quasiparticles, and  $n_{\omega}^*(t)$  is the density of excess 2 $\Delta$  phonons. The relative factor of 2 in Eq. (5) accounts for the fact that in a recombination process, two quasiparticles are destroyed and only one  $2\Delta$  phonon is created. The two recombination terms are separated in (5), and we define the thermal recombination rate:

$$
\tau_R^{-1} = 2Rn_T \tag{6}
$$

Kaplan et al. have calculated the leading low-temperature expression:

$$
\tau_R^{-1} \sim [\pi (2\Delta/kT_c)]^{1/2} (T/T_c)^{1/2} \exp(-\Delta/kT) \tau_0^{-1} ,
$$

where  $\tau_0$  is a characteristic time for different superconductors. Their calculated value for Sn is  $\tau_0(\text{Sn})=2.30$ nsec. Inserting the expression for the thermal quasiparticle density lensity<br> $n_T \sim 4N(0)\sqrt{\pi/2\Delta kT} \exp(-\Delta/kT)$ 

$$
n_T \sim 4N(0)\sqrt{\pi/2\Delta kT} \exp(-\Delta/kT)
$$

into Eq. (6), one obtains the recombination coefficient:

$$
R = [8N(0)\Delta]^{-1} (2\Delta/kT_c)^3 \tau_0^{-1}.
$$

#### B. Self-recombination

The term proportional to  $(n^*)^2$  describes selfrecombination of the quasiparticles. This term is independent of temperature and important when the quasiparticles are confined to a small volume. Owing to quasiparticle diffusion, this volume will spread out in time, leading to a coupling of self-recombination and quasiparticle diffusion

## C. Quasiparticle diffusion

Quasiparticle diffusion enters in two ways: (i) The volume spanned by the excess quasiparticles evolves with time, yielding a time dependent self-recombination term. (ii) Only quasiparticles above the oxide barrier can tunnel. This fraction of quasiparticles is time dependent because of diffusion, and therefore an additional factor in the tunneling term has to be added. We define  $A(x_0, t)$  to be the ratio of the number of excess quasiparticles above the tunneling area versus the total number of excess quasiparticles in the film. This ratio evolves with time and depends on the position  $x_0$  of the absorbed x ray in the film. For an x ray absorbed in the tunneling region,  $A(x_0, t)$  will initially be <sup>1</sup> and will decrease owing to the diffusion of excess quasiparticles out of the tunneling region. On the other hand, if quasiparticles are produced far away from the tunneling region,  $A(x_0, t)$  will increase from zero to a value depending on the distance of x-ray absorption.

The geometry of our junctions (see Fig. 1) allows a one-dimensional treatment of diffusion. The density of excess quasiparticles along the  $x$  axis is therefore

$$
n(x_0, x, t) = \frac{1}{\sqrt{2\pi v \lambda t}} \exp\left[-\frac{(x - x_0)^2}{2v \lambda t}\right],
$$
 (7)

where  $v = 0.3v_F$  is the average group velocity of quasiparticles,  $v_F = 1.9 \times 10^6 \mu \text{m}/\mu \text{sec}$  is the Fermi velocity in Sn,  $\lambda$  is the mean free path of quasiparticles, and  $x_0$  is the position of x-ray absorption. From this we obtain

$$
A(x_0,t) = \frac{\int_{-3d}^d n(x_0,x,t)dx}{\int_{-d}^{1-d} n(x_0,x,t)dx}.
$$

The lower boundary  $3d$  of the integral in the numerator takes into consideration a first refiection of the quasiparticles at the film boundary  $-d$ . The integrals can be expressed in terms of the error function.

### D. Phonon pair breaking

The phonon pair breaking time  $\tau_B$  has also been calculated by Kaplan et  $al$ .<sup>33</sup> This rate is constant at the low temperatures of interest  $(T/T_c \sim 0.1)$  and a characteristic value of the superconductor. In the case of tin they obtained  $\tau_B(\text{Sn}) = 0.110$  nsec.

#### E. Phonon escape

All the phonon escape rates are essentially free parameters, as the problem of phonon coupling of the films to the surroundings is not well understood. We adopt the usual dependence on film thickness

$$
\tau_{\gamma} \sim \frac{4d}{\eta c_s} \ ,
$$

where d is the thickness of film,  $\eta$  is the phonon transmissivity, and  $c<sub>S</sub>$  is the average phonon velocity. In the model we consider two phonon escapae modes:  $\tau_{\gamma_1}$  is the phonon escape from film <sup>1</sup> into the surrounding, and  $\tau_{\gamma_{12}}$  is the phonon transmission from film 1 into film 2, and we consider the respective terms for film 2.

#### F. Phonon breakup

The phonon breakup time  $t_{2\omega}$  describes the rate at which  $2\Delta$  phonons break into two phonons with energy less than  $2\Delta$ . An order of magnitude estimation for this value can be obtained from experiments where the thickness of the films was varied.<sup>35</sup> In those experiments, no deviation of the linear dependence of the recombination time  $\tau_{\text{eff}}$  with thickness has been observed, indicating that  $\tau_{2\omega}$  must be longer than phonon escape.

# G. Coupling of the two films

Back-tunneling couples the excess quasiparticle densities of the two films. The quasiparticle equation of film <sup>1</sup> hence has two tunneling terms, one loss term due to the transfer of quasiparticles from film <sup>1</sup> into film 2, and one tunneling term, increasing the number of quasiparticles because of tunneling from 2 into <sup>1</sup> (and correspondingly two terms for the equation of film 2).

In order to obtain the equations for the total number of excess quasiparticles and phonons, we multiply the densities with the volume  $V$  spanned by the excess quasiparticles. From the diffusion term (8) we can define a length scale

$$
\Delta x = \sqrt{v \lambda t}
$$

[standard deviation of Gaussian (7)] and define the volume to be

$$
V(t) = \begin{cases} d\Delta x^2 & \text{if } \Delta x < w, \\ d\Delta xw & \text{if } \Delta x > w, \end{cases}
$$

where  $d$  is the thickness of the film, and  $w$  the width of film. The volume term then appears only in the nonlinear self-recombination term.

The four coupled equations of the nonequilibrium model are

$$
\frac{d}{dt}N_1 = 2\tau_B^{-1}N_{\omega 1} - \tau_R^{-1}N_1 - \frac{R}{V_1(t)}N_1^2A_1(x_0, t)\tau_{\text{tun12}}^{-1}N_1
$$
  
+  $A_2(x_0, t)\tau_{\text{tun21}}^{-1}N_2$ ,  

$$
\frac{d}{dt}N_{\omega_1} = -\tau_B^{-1}N_{\omega 1} + \frac{1}{2}\tau_R^{-1}N_1 + \frac{1}{2}\frac{R}{V_1(t)}N_1^2 - \tau_{2\omega}^{-1}N_{\omega 1}
$$
  

$$
-\tau_{\gamma 1}^{-1}N_{\omega 1} - \tau_{\gamma 12}^{-1}N_{\omega 1} + \tau_{\gamma 21}^{-1}N_{\omega 2}
$$
,

 $(8)$ 

$$
\frac{d}{dt}N_2 = 2\tau_B^{-1}N_{\omega 2} - \tau_R^{-1}N_2 - \frac{R}{V_2(t)}N_2^2 - A_2(x_0, t)\tau_{\text{tun21}}^{-1}N_2
$$

$$
+ A_1(x_0, t)\tau_{\text{tun12}}^{-1}N_1,
$$

$$
\frac{d}{dt}N_{\omega 2} = -\tau_B^{-1}N_{\omega 2} + \frac{1}{2}\tau_R^{-1}N_2 + \frac{1}{2}\frac{R}{V_2(t)}N_2^2 - \tau_{2\omega}^{-1}N_{\omega 2}
$$

$$
-\tau_{\gamma_2}^{-1}N_{\omega 2} - \tau_{\gamma_21}^{-1}N_{\omega 2} + \tau_{\gamma_12}^{-1}N_{\omega 1} ,
$$

where  $N_1$  is the number of excess quasiparticles in film 1,  $N_{\omega 1}$  is the number of excess 2 $\Delta$  phonons in film 1,  $N_2$  is the number of excess quasiparticles in film 2, and  $N_{\omega^2}$  is the number of excess  $2\Delta$  phonons in film 2. For the quasiparticle relaxation rates,  $\tau_B^{-1}$  is the phonon pair breaking rate,  $\tau_R^{-1}$  is the thermal recombination rate,  $R/V_1(t)$  is the self-recombination rate in film 1,  $R/V_2(t)$ is the self-recombination rate in film 1,  $A_1(x_0,t)\tau_{\text{tun12}}^{-1}$  is the tunneling rate from film <sup>1</sup> into film 2, and  $A_2(x_0, t) \tau_{\text{tun21}}^{-1}$  is the tunneling rate from film 1 into film 2. For the 2 $\Delta$  phonon relaxation rates,  $\tau_{\gamma_1}^{-1}$  is the escape rate from film 1 into surroundings,  $\tau_{\gamma_2}^{-1}$  is the escape rate from film 2 into surroundings,  $\tau_{\gamma_{12}}^{-1}$  is the transmission from film 1 into film 2,  $\tau_{\gamma_{21}}^{-1}$  is the transmission from film 2 into film 1, and  $\tau_{2\omega}^{-1}$  is the break up rate.

In order to be able to compare the solutions of the model with the measured pulse shapes, we add a fifth equation describing the accumulation of the measured charge  $Q(t)$ :

$$
\frac{d}{dt}Q(t)=i_{+}(t)-i_{-}(t),
$$

where  $i_{+}$  is the excess quasiparticle current, and  $i_{-}$  is the discharge current due to RC time constant. The current  $i_{+}$  is the sum of the two tunneling terms

$$
i_{+}(t) = A_{1}(x_{0}, t)\tau_{\text{tun12}}^{-1}N_{1}(t) + A_{2}(x_{0}, t)\tau_{\text{tun21}}^{-1}N_{2}(t) ,
$$

and the current  $i_i$  is

$$
i_{-}(t) = \tau_{RC}^{-1}Q(t) ,
$$

where  $\tau_{RC}$  is the RC time constant of the preamplifierdetector circuit.

Owing to the large differences in the magnitude of the various relaxation terms some care is required when solving Eqs. (8) numerically. Special numerical algorithms are available for these type of equations.

# V. SOLUTIONS OF THE MODEL

In order to test the physical relevance of Eqs. (8), we have chosen parameters in order to simulate our detector as closely as possible. It is not the intention of this section to make a free fit of the model, as this mould be physically meaningless, considering the large number of free parameters. Having selected a fixed set of parameters, defining a "model-junction detector," we calculated the response of this model junction to the variation of

various parameters. The Eqs. (8) solve simultaneously the response to the absorption of x rays in either films, and the two solutions obtained are hence not independent of each other. The following parameters have been selected.



From these parameters one obtains the following values.



In Fig. 9, the solution for an x ray absorbed in film I is shown on the left-hand side, and the corresponding solution for one absorbed in film 2 on the right-hand side. The calculated pulses are superimposed on the experimental pulses, the latter recognizable by the step structure due to the digitization  $(160 \text{ nsec}/\text{bin})$ .<sup>8</sup> In the upper half, all calculated and measured pulses are shown normalized, and in the lower half the pulses are normalized in respect to a given pulse from an x-ray event absorbed in film 2, showing the relative pulse heights. We show the solutions for two positions of x-ray absorption: one in the tunneling region  $(x_0=0)$  and the other at the other end of the film  $(x_0 = 1000 \,\mu\text{m})$ . A larger mean free path in film 1 is reasonable, because there the evaporation rate was ten times faster than in film 2. This can reduce the quasiparticle mean free path  $\lambda$  in film 1 owing to the higher purity, indicating that possibly impurity scattering dominates quasiparticle diffusion. The relative amplitudes of the solutions were adjusted by choosing the respective phonon transmissivities from the films into the surroundings to be 0.25 for film 1, and 0.<sup>1</sup> for film 2, respectively. A ratio of this order is to be expected, as phonon escape is enhanced for the film coupled to the substrate (film 1) in the absence of a helium film. The pulse height of the solution corresponds to  $3\times10^6$  electron charges and fits surprisingly mell the accumulated charges measured. The solu-



FIG. 9. Solutions of the rate equations compared with experimental pulses. The data on the left is due to x rays absorbed in film 1, and the data on the right is due to x rays absorbed in film 2.  $X_0$  is the position of the x-ray absorption in the films.  $\lambda_1$  and  $\lambda_2$  are the quasiparticle mean free paths of the two films.

tions are insensitive to the relative initial quasiparticle and phonon populations, i.e., choosing an initial phonon to quasiparticle ratio of 1:1 or 1000:1 does not change the pulse height and pulse shape significantly; this is to be expected considering the fast phonon pair breaking time  $(\tau_B = 0.11$  nsec). As can be seen in the lower half of Fig. 9, the pulse heights of events absorbed in the current leads are not modeled well. We believe this to be due to the coarse treatment of quasiparticle diffusion in our model. As has been pointed out above, diffusion is coupled to the nonlinear self-recombination term, which is dominant at low temperatures and for small volumes. Therefore, selfrecombination is believed to be more effective in the narrow current leads because of their smaller volume. This was taken into account by modifying the nonlinear term  $R/V(t)$  to be

$$
\frac{R}{V(t)}\{A(x_0,t)+R_{CL}[1-A(x_0,t)]\}.
$$

 $A(x_0, t)$  is the fraction of quasiparticles in the tunneling region, and  $1 - A(x_0, t)$  the fraction in the current leads.  $R_{CL}$  is a factor proportional to the ratio of the width of the junction to the current leads (in the model junction taken to be 10). All the solutions of this section were obtained with this modified self-recombination term.

In Fig. 10, the solutions for a smaller mean free path  $(\lambda=0.3\mu\text{m})$  in film 2 are presented. The five solutions correspond to an x ray absorbed at  $x = 0$   $\mu$ m, 250  $\mu$ m, 500  $\mu$ m, 750  $\mu$ m, and 1000  $\mu$ m. The experimental pulse shapes have been obtained by making a scatter plot with

the digitized pulses and identifying events in the diffusio band.<sup>8,10</sup> Figure 10 shows good qualitative agreement and supports the existence of quasiparticle diffusion in our detector. The equations were also solved for different temperatures. This is shown in Fig. 11. The solutions do not agree well at lower temperatures. As has been mentioned already, this is believed to be due to the coarse approximation of diffusion, and consequently selfrecombination, which is dominant at these low temperatures.

ln order to demonstrate the importance of back tunneling, the rate equations were solved for both cases, with and without back tunneling. In the two diagrams on the upper half of Fig. 12, the solutions with back tunneling are shown, and in the lower half of Fig. 12 the solution without back tunneling is shown. In the latter case, there is an obvious disagreement with the measured pulse shapes. The calculated value for the accumulated charge without back tunneling is  $1.3 \times 10^6$  e, compared to  $3 \times 10^6$ e in the case with back tunneling, inferring an intrinsic amplification gain of  $\sim$  2.

The results presented in this section indicate that the four coupled rate equations (8) can account for most of the basic features of the detector. At lower temperatures, a complete two-dimensional, diffusive treament of the detector would be more appropriate.

# VI. CONCLUSIONS

Perturbing superconducting tunneling junctions with x-ray photons produces an excess distribution of quasipar-



rticle diffusion is evident from the comparison of the solutions with the experimental pulses. The position of xray absorption is varied between 0 and 1000  $\mu$ m, in steps of 250  $\mu$ m



FIG. 11. Solutions for different temperatures:  $0.45$ ,  $0.55$ ,  $0.62$ ,  $0.72$ , and  $0.78$  K.



FIG. 12. The influence of back tunneling is evident when comparing the solutions with and without back tunneling to the experimental pulse.  $Q_c$  is the normalization factor of the respective pulse-height scales, showing an enhancement of  $\sim$  2 owing to the back tunneling.

ticles and phonons owing to dynamical breaking of Cooper pairs. The relaxation of these distributions can be approximated by four Rothwarf-Taylor equations, one quasiparticle equation and one  $2\Delta$ -phonon equation for each of the two films of the juntion. Because of the back-tunneling effect, the excess quasiparticle distributions in both films are coupled. Quasiparticle diffusion has been included in the model in an approximate way. The agreement of the solutions with measured pulse shapes is satisfactory, showing that the basic features of the detecor are well described by the relaxation of nonthermal quasiparicle and phonon populations. A more accurate treatment of diffusion ought to solve the observed discrepancies at lower temperatures and for x rays absorbed outside of the tunneling region.

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