Nonequilibrium superconducting thin films with sub-gap and pair-breaking photon illumination

T Guruswamy, D J Goldie and S Withington

Quantum Sensors Group, Cavendish Laboratory, University of Cambridge, J J Thomson Avenue, Cambridge CB3 0HE, UK

E-mail: tg307@mrao.cam.ac.uk

Received 12 December 2014, revised 6 February 2015
Accepted for publication 26 February 2015
Published 8 April 2015

Abstract
We calculate nonequilibrium quasiparticle and phonon distributions for a number of widely-used low transition temperature thin-film superconductors under constant, uniform illumination by sub-gap probe and pair-breaking signal photons simultaneously. From these distributions we calculate material-characteristic parameters that allow rapid evaluation of an effective quasiparticle temperature using a simple analytical expression, for all materials studied (Mo, Al, Ta, Nb, and NbN) for all photon energies. We also explore the temperature and energy-dependence of the low-energy quasiparticle generation efficiency $\eta$ by pair-breaking signal photons finding $\eta \approx 0.6$ in the limit of thick films at low bath temperatures that is material-independent. Taking the energy distribution of excess quasiparticles into account, we find $\eta \to 1$ as the bath temperature approaches the transition temperature in agreement with the assumption of the two-temperature model of the nonequilibrium response that is appropriate in that regime. The behaviour of $\eta$ with signal frequency scaled by the superconducting energy gap is also shown to be material-independent, and is in qualitative agreement with recent experimental results. An enhancement of $\eta$ in the presence of sub-gap (probe) photons is shown to be most significant at signal frequencies near the superconducting gap frequency and arises due to multiple photon absorption events that increase the average energy of excess quasiparticles above that in the absence of the probe.

Keywords: superconductors, nonequilibrium, quasiparticle, detectors

1. Introduction
The energy distribution of the quasiparticle excitations of a superconductor determines its electrical [1] and thermal transport properties [2]. A theoretical understanding of devices including kinetic inductance detectors (KIDs) [3–5], superconducting tunnel junctions [6], superconducting qubits [7–9], SQUID based parametric amplifiers and mixers [10], and quantum capacitance detectors [11] requires a description of the quasiparticle system response. Therefore, a method for calculating the excited, possibly nonequilibrium distribution of quasiparticles is central to any device model.

Nonequilibrium superconducting detectors rely on pair-breaking to create their detected signal. We are particularly interested in thin-film superconductors fabricated on a dielectric substrate that is cooled to a sufficiently low temperature (the bath temperature) $T_b$. Typically $T_b \sim 0.1 T_c$ where $T_c$ is the superconducting transition temperature. An interacting signal photon of energy $h \nu_{\text{signal}}$, where $h$ is Planck’s constant and $\nu_{\text{signal}}$ the signal frequency, directly breaks a condensate pair in the superconductor provided $h \nu_{\text{signal}} \geq 2 \Delta$ where $\Delta$ is the superconducting energy gap. Due to the high density of states close to the gap, and for moderate energy photons, the interaction creates a distribution of excited quasiparticles with peaks [12] at $E = \Delta$ and
\[ E = \hbar \nu_{\text{signal}} - \Delta \]  
where \( E \) is the quasiparticle energy. \( E \) is related to the underlying Bloch state energy \( e \) by 
\[ E = \sqrt{e^2 + \Delta^2} \]. Higher energy signal photons will directly release an atomic electron via the photoelectric effect. In this case effects associated with localized heating and gap-reduction or ‘hotspots’ may become important. In this work we concentrate on modelling the detection of moderate energy photons \( \hbar \nu_{\text{signal}} < \Omega_0 \) where these effects are insignificant. The photon energies considered are particularly important to understand the responsivity, sensitivity and signal-to-noise of currently-deployed and planned astronomical instruments performing measurements in the frequency window of 0.1 to 10 THz. In the case of KIDs, the nonequilibrium state created by the interaction is monitored using a probe of energy \( \hbar \nu_{\text{probe}} \ll 2\Delta \), where \( \nu_{\text{probe}} \) is the probe frequency, so an additional drive term needs to be included in the detailed analysis.

An important consideration in any nonequilibrium superconducting detector operating at low reduced temperatures \( T_0/Tc \ll 1 \) is that the detected signal is most influenced by the presence of low-energy quasiparticles because the relaxation of the primary excitation occurs on time-scales that are much shorter than the ultimate relaxation time of the low-energy excess. (Here we define low-energy quasiparticles to have \( E < \Delta \)). We assume the signal photons interact with 100% efficiency i.e. we ignore for example any optical coupling efficiencies. A primary excitation with \( E = \hbar \nu_{\text{signal}} - \Delta \) relaxes towards the gap emitting a phonon. If this emitted phonon has energy \( \Omega \gg 2\Delta \), an additional pair may be broken enhancing the number of low-energy excess quasiparticles and thus the detected signal. But in a thin-film this emitted phonon may be lost into the substrate, reducing the number of low-energy excitations. The average number \( m \) of low-energy quasiparticles resulting from the interaction of a single high-energy photon can be quantified in terms of a quasiparticle generation efficiency (or quasiparticle yield) \( \eta \). If the low-energy quasiparticles have average energy \( \langle E_{\text{qp}} \rangle \) then the efficiency \( \eta = m \langle E_{\text{qp}} \rangle / \hbar \nu_{\text{signal}} \). Often it is assumed \( \langle E_{\text{qp}} \rangle = \Delta \) [13].

Phonon loss means \( \eta \approx 1 \). Most existing work assumes a value of \( \eta \approx 0.6 \) for all materials, photon frequencies and bath temperatures [3]. At high temperatures \( T \approx T_c \) our description of the energy-cascade no longer applies [14, 15] because thermal phonon energies become comparable with \( \Delta \), and scattering and recombination occur on comparable timescales. Then a two-temperature model describing the quasiparticle and phonon systems that assumes \( \eta = 1 \) is most appropriate. Here we show that the full nonequilibrium calculation yields \( \eta \rightarrow 1 \) as \( T_0/T_c \rightarrow 1 \) in agreement with the assumption of the two-temperature model.

Previous work modelled the effect of incoming energy at low temperatures with an effective temperature [16] or as an effective chemical potential [17] for the quasiparticle distribution. To go beyond this analysis, Chang and Scalapino [18, 19] derived a set of coupled nonlinear kinetic equations describing the interactions of quasiparticles and phonons to find their respective energy distributions \( f(E) \) and \( n(\Omega) \). They solved for the nonequilibrium distributions resulting from various drive terms, including photon and phonon injection. A number of investigations have explored the effect of very high energy photons (x-ray or optical photons) on infinite superconductors [20, 21] and thin films [22] calculating a quasiparticle generation efficiency of \( \eta \approx 0.6 \), ignoring \( \Omega \gg 2\Delta \) phonon loss. Another approach [13] considered the time-evolution following local energy deposition into the thin film. Kozorezov et al [23] considered the energy down-conversion process after absorption of a high energy photon for a variety of materials and concluded that the materials can be categorized into three different classes, with the low energy-gap superconductors all having \( \eta \approx 0.6 \).

We have previously reported [24] a numerical approach that solves the coupled kinetic equations describing the quasiparticle and phonon distributions \( f(E) \) and \( n(\Omega) \) for a superconducting thin-film driven by a sub-gap probe [24], and including the effect of an additional pair-breaking signal [25], for Al thin-films at low temperatures \( T \sim 0.1 \ T_c \). We have also reported detailed calculations of KID characteristics (quality factors and quasiparticle lifetimes as functions of readout power and \( T_0 \)) that were compared to precise measurements of Al resonator behaviour finding good agreement [26]. More recent measurements on KIDs [27] of the quasiparticle number and recombination time-dependence on readout power appear to show that they must be explained using nonequilibrium quasiparticle distributions. Other recent measurements with a Ta KID [28] report a detector response that has the same qualitative features as the energy dependence of \( \eta \) for Al described in Guruswamy et al [25]. Here we apply the method to a number of other technologically important low temperature superconductors and discuss the physics involved. We concentrate in particular on the refractory elements and extend the earlier analysis to bath temperatures approaching the superconducting critical temperature \( T_c \). The highest probe powers studied here \( \sim 10^8 \text{W m}^{-2} \text{in} \text{Nb} \) approach those that we estimate were used by Chiu et al [29]. In that work high absorbed (probe) power densities were associated with large induced supercurrents and magnetic fields approaching the critical field. Over most of the range of powers that we study our calculations show that these effects are negligible. Nevertheless we include high absorbed powers in our modelling to enter this regime. We study Mo, Al, Ta (in its higher \( T_c \sim 4.4 \text{K} \) form), Nb and NbN. In each instance we use measured material properties that characterize films that we deposit by sputtering under ultra-high-vacuum. We study thin films with thicknesses in the range 40 to 500 nm that are most commonly used for KIDs. Application of our results to ultrathin films \( (d \sim 10 \text{ nm}) \) must be done with caution, but even so we expect them to be representative. Our results are applicable to many different device designs, as only the phonon escape time to the substrate \( \tau_{\text{e}} \) is device-geometry dependent. The method assumes that phonon pair-breaking, quasiparticle recombination, and electron-phonon scattering are the only significant interaction processes. This assumption is investigated in section 2.2 for typical thin-films intended for detector applications. Section 2 reviews superconducting parameters for
the materials discussed, considers the relative contribution from electron–electron scattering for the different materials, describes the numerical method, and also describes how $\eta$ is calculated for a pair-breaking signal. Section 3 presents an analytical superconductor cooling model that can be used to calculate the effective quasiparticle temperature $T_\phi$ for both sub-gap and pair-breaking photons. Section 4 describes detailed numerical results for both sub-gap and pair-breaking photon interactions including the effect of the effect of changes in the bath temperature. Section 5 summarizes the work done.

2. Methods

2.1. Superconducting parameters

The superconducting parameters needed for modelling are the characteristic quasiparticle lifetime

$$\tau_0 = \frac{Z_1(0) h}{2\pi b (k_B T_c)},$$

and the characteristic phonon lifetime

$$\tau^{\text{ep}}_0 = \frac{\hbar N_{\text{ion}}}{4\pi^2 N(0)\Delta_0},$$

as defined by Kaplan et al [30] $N(0)$ and $N(0)_{\text{ion}}$ are the mass-enhanced and bare single-spin density of states at the Fermi energy $E_F$ respectively, where $N(0) = Z_1(0) N(0)_{\text{ion}}, N_{\text{ion}}$ is the ion density, and $\Delta_0$ the zero temperature superconducting energy gap. $Z_1(0)$ is the electron–phonon renormalization factor $Z_1(0) = 1 + \lambda$, where $\lambda = 2 \int_0^\infty d\Omega \alpha^2(\Omega) F(\Omega) / \Omega$ is the dimensionless electron–phonon coupling strength. $\alpha^2(\Omega) F(\Omega)$ is the Eliashberg function, $\alpha^2(\Omega)$ is the electron–phonon interaction matrix element and $F(\Omega)$ is the phonon density of states. The material-dependent parameter $b$ is calculated within a Debye model such that $b\Omega^2 = \alpha^2(\Omega) F(\Omega)$ for low phonon energies $\Omega \sim 2\Delta_0$. The averaged value of the interaction matrix element is $\langle \alpha^2 \rangle = 1/3 \int_0^\infty d\Omega \alpha^2(\Omega) F(\Omega)$. Values of $b$ and $\langle \alpha^2 \rangle$ for Al, Ta and Nb are already tabulated [30]. For Mo we use the point-contact measurements of Caro et al [31] while for NbN we use the tunnelling data of Kihlstrom et al [32] and the transport property measurements of Semenov et al [33]. We note that the precision of our calculations of $b$ are dependent on the accuracy within which we can interpret the relevant low-frequency parts of the reported $\alpha^2(\Omega) F(\Omega)$ data. We assume here that the phonons of the superconducting film can be described by a 3-dimensional (3-D) model. For ultrathin films, typically of thickness $d < 10$ nm, this assumption fails and measured values of the temperature dependence of the normal-state electron-phonon relaxation time $\tau_{e-p}$ indicate a reduced phonon dimensionality [34].

The characteristic times calculated are listed in table 1. We assume the weak-coupling relationship $\Delta_0 = 1.76 k_B T_c$ for all materials to determine $T_c$ in table 1. The assumption is reasonable for Mo Al and Ta, but gives a slightly higher $T_c$ compared to experiment for Nb (typically $T_c \sim 9.3$ K) and for NbN ($T_c \sim 15$ K). However we do not expect the assumption to significantly affect the physics of the conclusions drawn; the differences introduced by strong coupling are reduced because of the short lifetimes of states with energy $E \gg \Delta$ [18, 30]. $\tau^{\text{ep}}_0$ is the value of $\tau^{\text{ep}}_0$ required for energy conservation calculated from 8 and discussed in more detail in 2.3. The values of $\tau_0$ for Al, Ta and Nb shown in table 1 have already been widely used to interpret measurements, often of quasiparticle recombination times. For NbN there is less available experimental data. Gousev et al [36] measured $\tau_{e-p}$ in ultrathin (3.5 nm) NbN films finding $\tau_{e-p} \approx 35$ ps for $T = 5$ K and with temperature dependence $\tau_{e-p} \sim T^{1.6}$. Scaling this measurement to 16.8 K we estimate $\tau_{e-p} \sim 5$ ps. Identifying [30] $\tau_{e-p} = \tau_0/8.4$, a 3-D phonon spectrum gives a value of 2.4 ps in reasonable agreement.

2.2. Electron–electron versus electron–phonon scattering

Insight into the contribution from electron–electron $e \rightarrow e$ scattering in the down-conversion process is most easily found by considering the relevant scattering rates $T_e$. For the electron energies of interest $e < \Omega_0$, we assume that electron-phonon ($e \rightarrow p$) inelastic scattering dominates over ($e \rightarrow e$) scattering. We previously investigated [25] to what extent this assumption is valid for low-resistivity Al films. Now we consider the other low-$T_c$ superconductors using material parameters typical of those that we measure experimentally for the thin-films we deposit by magnetron-sputtering in ultra-high vacuum.

In the normal-state the $e \rightarrow p$ scattering rate $\tau^{-1}_{e-p}$ is [37]

$$\tau^{-1}_{e-p}(e) = \frac{2\pi}{\hbar} \int_0^e d\Omega \alpha^2(\Omega) F(\Omega).$$

Assuming $\alpha^2(\Omega)$ is constant at low energies, a Debye model for phonons [30] leads to

$$\tau^{-1}_{e-p}(e) = \begin{cases} \frac{e^3}{3\tau_0 (k_B T_c)^3} & \text{if } e < \Omega_0 \\ \Omega_0 & \text{if } e \geq \Omega_0 \end{cases}.$$  

The electron–electron scattering rate in clean films is estimated with the Landau–Pomeranchuk formula [23]

$$\tau^{-1}_{e-e}(e) = \frac{e^2}{\hbar \kappa} \frac{r_e^{1/2}}{7.96},$$

where $r_e$ is the radius containing one electron charge divided by the Bohr radius (approximated as 1). For thin resistive films the $e \rightarrow e$ rate can be significantly enhanced so that [38]

$$\tau^{-1}_{e-e}(e, R_{eq}) = \frac{e^2 R_{eq}}{2 \pi^2 \hbar^2} \ln \frac{\hbar \kappa}{e^2 R_{eq}},$$

where $R_{eq} = \rho / d$ is the sheet resistance of the thin-film, $\rho$ is the resistivity and $d$ is the thickness. These normal-state

1 This differs by a factor of $\pi$ from the expression given in Alshuler and Aronov [46].
for all materials except Mo. Figure 1 shows that $\phi$ V $\phi$ and below this energy range. However, the $e - e$ scattering rate is considerably suppressed in the superconducting state at low $T_c$ [39]. We conclude that there is negligible contribution from $e - e$ scattering for most of these materials, thicknesses, temperatures and signal photon energies. For Mo we note that additional $e - e$ scattering may increase the quasiparticle generation efficiency over the results presented here by providing an additional pair-breaking mechanism especially near $T_c$.

2.3. Numerical method

We solved the coupled kinetic equations [18] describing the quasiparticle and phonon distributions numerically to find the steady-state driven distributions, $f(E)$ and $n(\Omega)$ for the quasiparticles and phonons respectively, using Newton–Raphson iteration. Drive terms for pair-breaking and sub-gap photons [43] were included as necessary and the numeric pre-factors of these terms scaled to match the absorbed powers, $P_{\text{probe}}$ for sub-gap photons and $P_{\text{signal}}$ for the pair-breaking signal. The numerical method was iterated until the solutions conserved energy to within $\xi = 10^{-5}$. This was quantified as $\xi = \sqrt{\xi_{\text{ph}}^2 + \xi_{\text{ph}}^2}$, where $\xi_{\text{ph}}$ is the relative difference between the photon power absorbed and power flow from quasiparticles to phonons, and $\xi_{\text{ph}}$ is the relative difference between the photon power absorbed and the power flow from phonons to substrate (held at a temperature $T_b$). The numerical method is outlined in more detail in Goldie and Withington [24].

For each material, the temperature dependent energy gap $\Delta(T)$ was calculated using the BCS gap equation,

$$\frac{1}{N(0)V_{\text{BCS}}} = \int_{\Delta(b)}^{\Delta(0)} dE \frac{1 - 2f(E, T)}{\sqrt{E^2 - \Delta(T)^2}},$$

where $f(E, T) = (\exp(E/k_B T) + 1)^{-1}$ is the Fermi–Dirac distribution, and $N(0)V_{\text{BCS}}$ is the dimensionless electron–phonon coupling constant, calculated from the zero temperature form of (7) as $N(0)V_{\text{BCS}} = 1/\sinh^4(\Omega_0/\Delta_0)$.

### Table 1. Characteristic quasiparticle and phonon lifetimes, and associated parameters. Data from Gladstone et al [35], Kaplan et al [30] or Zehnder [13] unless otherwise specified. For Mo and NbN we use measurements of $\alpha^2(\Omega) F(\Omega)$ from Caro et al [31] and Kihlstrom et al [32] respectively. $\tau_{ee}^c$ is the value of $\tau_{ee}^c$ required for energy conservation calculated from (8), $\nu_{gap} = 2\Delta_0/h$ is the gap frequency.

| Material | Mo | Al | Ta | Nb | NbN |
|----------|----|----|----|----|-----|
| $\Delta_0$ ($\mu$eV) | 140 | 180 | 700 | 1470 | 2560 |
| $\nu_{gap}$ (GHz) | 68 | 87 | 339 | 711 | 1240 |
| $T_c/\Omega$ | 0.92 | 1.18 | 4.6 | 9.67 | 16.8 |
| $b$ ($10^{-4}$ MeV$^{-2}$) | 2.28 | 3.17 | 17.3 | 40 | 47 |
| $\langle \alpha^2 \rangle$ (MeV$^2$) | 1.62 | 1.93 | 1.38 | 4.6 | 4.99 |
| $\lambda$ | 0.42 | 0.43 | 0.69 | 1.84 | 1.46 |
| $\tau_0$ (ns) | 1.310 | 438 | 1.78 | 0.15 | 0.02 |
| $\tau_{ee}^c$ (ps) | 231 | 260 | 22.7 | 4.17 | 5.98 |
| $\tau_{ee}^c/\tau_{\phi}^c$ | 1.04 | 1.04 | 0.91 | 0.94 | 1.01 |

### Figure 1. Energy dependence of normal state scattering rates in Mo. Electron–phonon rate $\tau_{\phi}^{-1}(\epsilon)$ (solid green), clean limit electron–electron rate $\tau_{ee}^{-1}(\epsilon)$ (dotted blue), and thin-film resistive $\tau_{ee}^{-1}(\epsilon, R_{\text{ph}})$ (dashed red). Calculations are for a 40 nm Mo film with $\rho = 9.2 \times 10^{-4} \Omega \text{m}$. Calculations significantly overestimate the $e - e$ scattering rates in the superconducting state at low temperatures $T_c \ll T_f$ [39]. Figure 1 plots the normal-state scattering rates for a 40 nm thick Mo film and we have used the thin-film resistive $e - e$ scattering rate 6. We identify the following characteristic energies for each material: $\epsilon_b: \Omega_0$; and a low-energy crossover $E_c$, below which the $e - e$ scattering rate exceeds the $e - p$ rate. $E_c/\Delta$ is highest for Mo for the material parameters chosen, as shown in table 2.

During the energy down-conversion, the inelastic scattering rate is only relevant above energies $E \gg 3\Delta$ where additional pair breaking is possible. We find that $E_c$ is below $3\Delta$ for all materials except Mo. Figure 1 shows that $e - p$ scattering dominates in that case for $\epsilon \approx 6\Delta$ to $10^3\Delta$ for the modelled Mo film, and $e - e$ scattering may contribute above
The numerical method converges on a solution that minimizes \( \xi \) provided the material parameters satisfy
\[
\frac{2\pi N(0)\tau_0^0\Delta_0^0}{9N_{\text{ion}}\tau_0(k_B T_c)} = 1, \tag{8}
\]
a consequence of matching the low energy part of the measured Eliashberg function to a Debye-model approximation that determines \( b \). In our previous work we ensured that \( T_c \) satisfied (8), but then the \( T_c \) obtained did not precisely agree with (7) (although the difference is small <1%). In this work, since we are also interested in the temperature dependence of the response we calculated \( T_c \) from (7) and defined an energy-conserving characteristic phonon lifetime \( \tau_0^0 \) to satisfy (8). Given the difficulty of interpreting the low-energy \( a^2(\Omega)F(\Omega) \) data that we and indeed Kaplan et al \[30\] and others have noted we consider this approach entirely reasonable. The ratios \( \tau_0^0/\xi \) are given in table 1. Satisfyingly the ratios are within 10% of unity for all materials studied. In our calculation of \( b \) for Mo, our error would be of this order given the difficulty of determining the low-energy \( a^2(\Omega)F(\Omega) \) from the available data. We note that other estimates of the characteristic times exist (see for example Parlati et al \[44\]) although those estimates do not necessarily satisfy the energy-conserving requirement.

In the numerical code the energy-bin distribution for both the quasiparticles and phonons was also changed when calculating bath temperature dependencies. As \( T_b \) is increased, the phonon-bath power flow \( P(\Omega)|_{b\to b} \) above \( \Omega \approx \Delta \) increasingly resembles the difference between two thermal distributions, and an increasing fraction of power is carried by high energy phonons, as shown in figure 2. Therefore at high temperatures, the phonon distribution must be accurately calculated to these higher energies. The energy of the last phonon bin \( \Omega_{\text{max}} \) significantly affects the energy conservation of the solutions. Figure 2 (inset) shows that to have a relative power flow difference \( \xi|_{b\to b} < 10^{-5} \), required \( \Omega_{\text{max}} > 15 k_B T_b \) for \( T_b/T_c = 0.95 \).

However, the bin width could not be made too large as the energy gap \( \Delta(T) \), readout photon energy \( h\nu_p \), and signal photon energy \( h\nu_b \) had to be rounded to the nearest multiple of the bin width, such that the photon-induced peaks in the distributions occurred within well-defined energy bins. We chose to model the phonons with \( N = 2500 \) uniformly-sized bins with maximum phonon energy \( \Omega_{\text{max},b} \), and similarly the quasiparticles with maximum energy \( E_{\text{max}} = \Omega_{\text{max}} + \Delta(T) \). Bin width was therefore variable, depending on material and temperature. The quasiparticle density of states broadening parameter \( \gamma \) was recalculated for each simulation, changing for different material and bin width combinations. It was chosen to minimize the difference between the quasiparticle number calculated as the integral \( 4N(0)\int_{\Delta}^{\infty} dE \rho_{\text{BCS}}(E, \Delta) f(E, T_b) \), where \( \rho_{\text{BCS}}(E, \Delta) = E/\sqrt{E^2 - \Delta^2} \), and the equivalent sum over the discretized distribution with the broadened density of states calculated with \( E \to E + i\gamma \). In all cases the approach worked well.

2.4. Low-energy quasiparticle generation efficiency for a pair-breaking signal

For a single pair-breaking photon we have defined a low-energy generation efficiency \( \eta = m \langle E_{\text{qp}} \rangle/\hbar \nu \). Here we describe the calculation of \( \eta \) for a constant input signal power \( P_{\text{signal}} \) in a thin-film that may also be driven by a non-direct pair-breaking constant readout power \( P_{\text{probe}} \) (such as used for KID readout). The absorbed power creates a number of excess quasiparticles \( N_{\text{excess}} = N_{\text{signal}} - N_{\text{probe}} \) such that the quasiparticle system total energy has increased by \( E_{\text{excess}} = E_{\text{signal}} - E_{\text{probe}} \), where \( N_{\text{signal}} \) and \( E_{\text{signal}} \) are the quantities for distributions driven by the signal and probe together. In the steady state, photons absorbed at a rate \( T_b \) create low-energy quasiparticles at a rate \( T_{\text{qp}} = mT_b \).

Solving a modified set of Rothwarf–Taylor rate equations \[45\] for \( T_b \) and \( T_{\text{qp}} \) results in \[25\]
\[
\eta = \frac{m \langle E_{\text{qp}} \rangle}{P_{\text{signal}}} \left( \frac{N_{\text{signal}}^2 - N_{\text{probe}}^2}{P_{\text{signal}}} \right) \frac{2R}{1 + \beta R},
\]
where \( R \) and \( \beta \) are the distribution-averaged recombination rate and pair-breaking rates respectively. We define \( \langle E_{\text{qp}} \rangle \) the average energy of the excess low-energy quasiparticles where
\[
\langle E_{\text{qp}} \rangle = \frac{\int_{\Delta}^{\infty} \rho(E)\left( f(E) - f(E, T) \right) dE}{\int_{0}^{\infty} \rho(E)\left( f(E) - f(E, T) \right) dE}.
\]
At low temperatures \( \langle E_{\text{qp}} \rangle \approx \Delta \) as usually assumed: taking account of the detailed energy distributions allows \( \eta \) to be determined at arbitrary temperatures.
Table 2. Characteristic material energies related to scattering. \( E_c \) is the energy at which the electron–phonon scattering rate becomes greater than the thin-film resistive electron–electron scattering rate. Data from Gladstone et al [35], Kaplan et al [30] or Kozorezov et al [23] unless otherwise referenced. The material resistivities and thicknesses used are measured values from thin-films deposited by our group. All data is for polycrystalline films except for Ta where we assume parameters typical of the higher-\( T_c \) bcc form.

| Material | \( \rho \) (\( 10^{-8} \) \( \Omega m \)) | \( d \) (nm) | \( \epsilon_r \) (eV) | \( \Omega_0 \) (meV) | \( E_c / \Delta_0 \) |
|----------|--------------------------------|----------|----------------|----------------|----------------|
| Mo       | 9.2                           | 40       | 9.32           | 39.6           | 6.1            |
| Al [40]  | 0.8                           | 35       | 11.6           | 36.2           | 1.42           |
| Ta (bcc, epi) | 4.1                  | 100         | 9.5             | 20.7           | 0.23           |
| Nb       | 8.8                           | 100      | 6.18           | 23.7           | 0.14           |
| NbN [41, 42] | 250                        | 100      | 15.6           | 28.4           | 0.3            |

3. Superconductor cooling model

We used the steady-state driven distributions that were calculated to determine parameters for the thin-film superconductor cooling model outlined in Goldie and Withington [24]. This model is an analytic expression relating \( T_N \) to the power flow \( P \) between the quasiparticle and phonon systems (the latter assumed to be the substrate phonons) for a given \( T_b \ll T_c \) and material.

\[
P = \frac{1}{\eta(P, \nu)} \sum_i \frac{\eta_i}{\eta + \tau_i / \tau_{pb}} \times \left[ T_N^i \exp \left( -\frac{2N_b (T_N^i)}{k_B T_N^i} \right) - T_b \exp \left( -\frac{2N_b (T_b)}{k_B T_b} \right) \right] (11)
\]

\( \eta \) is a material-dependent constant, \( \tau_{pb} \) is the phonon pair breaking time, \( \sim \tau_0^p \) for \( T_b \ll T_c \). \( T_N^i \) is the effective temperature calculated to characterize the driven nonequilibrium quasiparticle distributions so that \( N_q = 4N(0) \int_0^\infty dE \rho(E)f(E, T_N^i) \eta(P, \nu) \) depends on the drive (probe or pair-breaking signal). Goldie and Withington [24] showed for a sub-gap probe \( \eta(P, \nu) \equiv \eta_2 \), the fraction of phonon-bath power flow carried by excess phonons with energy \( \Omega > 2 \Delta_0 (T_b) \) that depends on \( P, \nu_{probe} \) and \( \tau_l \). Here we show that (11) can also be applied for direct pair-breaking when \( \eta(P, \nu_{signal}) \equiv \eta \) as defined in section 2.4.

4. Results

4.1. Effects of a sub-gap probe

Here we show and discuss results for modelling of the effect of a sub-gap probe. The data-points (crosses) in figure 3(a) show the calculated \( T_N^i \) from the full non-equilibrium solution as a function of \( P_{probe} \). The solid line shows \( T_N^i \) evaluated with (11) and from this we also calculate \( \Sigma_i \). The calculations are for Nb, with \( P_{probe} = 2 \times 10^3 \) W m\(^{-3} \), \( h \nu_{probe} = 16 \) \( \mu \)eV, and \( \tau_l / \tau_0^p = 1 \). The analytical model is an excellent approximation to the full nonequilibrium calculation provided the power-dependence of \( \eta_2 \) shown in figure 3(b) is taken into account. The solid line in figure 3(b) is a piecewise fit such that

\[
\eta_2(P) = \begin{cases} 
\eta_0 \quad & \text{if } P \leq R_0 \\
\eta_1 \ln(P/R_b) + \eta_0 & \text{if } P > R_b 
\end{cases} \quad (12)
\]

\( P_0 \) characterizes a ‘knee’ probe power below which \( \eta_2 \) is constant and \( \eta_1 \) characterizes the energy dependence of \( \eta_2 \) at higher probe powers. Both are material dependent. Figure 3(c) shows that \( \tau_{pb} \sim \tau_0^p \) is a good approximation over the range of interest. Figure 3(d) shows that \( \eta_2 \) has no significant dependence on the phonon escape time \( \tau_l \).

The dependence of the fit parameters \( P_0, \eta_0, \) and \( \eta_1 \) on probe photon energy is shown in figure 4. This includes

![Figure 3](image-url)

![Figure 4](image-url)
calculations for Nb (blue ×), Ta (green □), and Al (red ○). Scaling the energy of the photon by \( \Delta_0 \), demonstrates the monotonic behaviour for all the materials. \( P_0 \) shown in figure 4(a) is scaled by the material-dependent \( \Sigma \) to emphasize the commonality.

Table 3 summarizes parameters derived from the modelling to describe sub-gap photon interactions in all of the materials studied. The table shows \( \Sigma \) and knee parameter \( \tau_0 \) at \( h\nu_{\text{probe}} = 16 \, \mu\text{eV} \) and \( \tau_1/\tau_0^b = 1 \). The values of \( \Sigma \) shown can also be used for estimates of \( T_A^* \) for a direct pair-breaking signal as we will discuss in the next section with an appropriate energy-dependent \( \eta \).

### 4.2. Effects of direct pair-breaking

Here we discuss results for a direct pair-breaking signal with \( 2\Delta < h\nu_{\text{signal}} < 10\Delta \). Figure 5 shows how \( \eta \) varies with signal photon energy. \( \eta \) decreases from unity in the range \( h\nu_{\text{signal}} = 2\Delta \) to \( 4\Delta \), as phonons emitted by quasiparticles scattering towards the gap escape the thin film. Above \( h\nu_{\text{signal}} = 4\Delta \), the phonons emitted in scattering can break additional Cooper pairs, increasing the quasiparticle generation efficiency. The magnitude of the increase depends on the phonon trapping factor [25]. Measurements of the response of a Ta KID [28] at signal frequencies close to the gap energy show these characteristic features. The figure shows that when \( h\nu_{\text{signal}} \), \( T_b \) and \( \tau_1 \) are scaled by the relevant material parameters, the quasiparticle generation efficiency is the same for all materials. Kozorezov et al [23] predicted that the quasiparticle generation efficiency could be categorized into three classes of superconductors. The superconductors included in this work, which fall within either the second (Ta, Nb, NbN) or third (Al, Mo) class, have a material-independent \( \eta \approx 0.6 \). This conclusion differs somewhat from that of Zehnder [13]. We understand this as due to the current work examining the steady-state response to constant incoming power, and scaling all relevant parameters by material characteristics, whereas Zehnder’s work examined the time-dependent response including quasiparticle out-diffusion from a localized hot-spot, and chose a fixed cutoff time of 10 ns to calculate the quasiparticle generation efficiency unscaled by material. We emphasize that for the low energy photons considered in the present work localized hot-spot formation (and consequent gap suppression) does not occur for the signal powers studied.

The other effect shown in figure 5 is the enhancement of \( \eta \) by the readout power due to an increase in the average energy of generated quasiparticles. For \( h\nu_{\text{signal}} \approx 2\Delta \), it can be seen that \( \eta > 1 \) which may seem unreasonable. The
enhancement is due to multiple photon absorption by the quasiparticles i.e. both signal and probe, and the effect is most-pronounced near $h\nu_{\text{signal}} = 2\Delta$. This results in a higher average excess quasiparticle energy when the readout power is present alongside the signal compared to when only the signal is present, and also produces the small variation in $\eta$ between materials. The absorbed readout power $P_{\text{probe}}$ and readout frequency $h\nu_{\text{probe}}$ are not scaled by material parameters in this figure, unlike the signal. When calculated with $P_{\text{probe}} = 0$, the signal quasiparticle generation efficiency is identical for all materials and is unity at $h\nu_{\text{signal}} = 2\Delta$.

Figure 6 shows the variation in $\eta$ as the bath temperature is changed. As $T_b$ increases, the scattering between the more numerous thermal quasiparticles and phonons determines the structure of the driven distribution, rather than scattering between the excess quasiparticles and phonons. This means the power flow from phonons to bath does not have the same peaked structure and instead more closely resembles the difference between two thermal distributions, as earlier shown in figure 2. This type of excess quasiparticle distribution means the average energy of generated quasiparticles increases without increasing the recombination and pair breaking rates. The figure also indicates temperatures for which $k_B T_b = 2\Delta(T_b)$ and $= \Delta(T_b)$, above which we expect thermal and nonequilibrium distributions to interact strongly. As $T_b/T_c \to 1$ we see that $\eta \to 1$ in agreement with the two-temperature model valid in that regime. We find that $T_N$ is well-accounted for by (11), with the value of $\eta$ shown, for

Figure 6. The quasiparticle generation efficiency $\eta$ as a function of reduced temperature $T_b/T_c$. The calculation is for for Al, with $P_{\text{probe}} = 0$, $P_{\text{signal}} = 2$ W m$^{-3}$, $h\nu_{\text{signal}} = 10 \Delta(T_b)$, and two values of the phonon escape time. The inset shows $T_N$ calculated using the full nonequilibrium model (red ) and using (11) (blue line) as a function of $P_{\text{signal}}$ with $T_b = 0.1 T_c$ and $\tau_1/\tau_0^\phi = 1$.

Figure 7. Quasiparticle generation efficiency $\eta$ for materials studied as a function of phonon escape time $\tau_1/\tau_0^\phi$. Calculation uses $T_b = 0.1 T_c$, $P_{\text{probe}} = 0$, $P_{\text{signal}} = 2$ W m$^{-3}$, and $h\nu_{\text{signal}} = 10 \Delta$.

$T_b/T_c \leq 0.7$. The inset shows $T_N$ calculated using (11) for $\tau_1/\tau_0^\phi = 1$ as a function of $P_{\text{signal}}$ with $T_b = 0.1 T_c$.

Figure 7 shows the dependence of $\eta$ on the phonon trapping factor $\tau_1/\tau_{\text{ph}}$ for $T_b = 0.1 T_c$. As the phonon escape time $\tau_1/\tau_0^\phi$ increases, the quasiparticle generation efficiency $\eta \to 0.6$ for all materials examined. This conclusion is in agreement with earlier Monte–Carlo calculations [20, 22] for infinite superconductors or where $\Omega \geq 2\Delta$ phonon loss was ignored.

5. Conclusions

We have described solutions of the coupled kinetic equations that calculate steady-state nonequilibrium quasiparticle and phonon distributions in a number of technologically important superconducting thin-films driven by sub-gap and pair-breaking photons. In particular we consider low energy signal photon interactions $h\nu_{\text{signal}} \leq 10\Delta$, where localized heating and gap-suppression can be ignored. We have calculated numerical parameters for these superconductors that can be used in a simple analytical expression to describe the power-flow between quasiparticles and phonons. This expression allows straight-forward estimates of the effective quasiparticle temperatures for both sub-gap and pair-breaking drives to approximate the nonequilibrium behaviour without resorting to a full numeric solution of the coupled kinetic equations. The analytical expression is shown to give a good account of full solutions of the detailed equations for a wide range of powers and bath temperatures. This is relevant for predicting the behaviour of thin-film nonequilibrium superconducting detectors. We defined a low-energy quasiparticle generation
efficiency for constant absorbed pair-breaking power and calculated detailed numerical solutions as a function of photon energies and bath temperature. We have shown that key parameters determining quasiparticle generation efficiency are the signal frequency and the phonon trapping factor. The enhancement of signal quasiparticle generation efficiency by absorbed readout power is demonstrated and explained as multiple-photon absorption (signal and probe) by the quasiparticles and Cooper pairs. We estimate this effect can result in an increase $\eta$ of up to 20% at signal photon energies of $h\nu_{\text{signal}} = 2\Delta$. The most sensitive detectors currently in development may also be able to distinguish between phonon trapping factors using results presented here. For the low-energy photon interactions studied we also show that the quasiparticle generation efficiency is material independent.

References

[1] Mattis D and Bardeen J 1958 Phys. Rev. 111 412
[2] Bardeen J, Rickayzen G and Tewordt L 1959 Phys. Rev. 113 982
[3] Zmuidzinas J 2012 Annu. Rev. Condens. Matter Phys. 3 169
[4] Baselmans J 2012 J. Low Temp. Phys. 167 292
[5] Vardulakis G, Withington S, Goldie D J and Glodawca D M 2008 Meas. Technol. 19 015509
[6] Friedrich S 2008 J. Low Temp. Phys. 151 277
[7] DiCarlo L, Reed M D, Sun L, Johnson B R, Chow J M, Gambetta J M, Frunzio L, Girvin S M, Devoret M H and Schoelkopf R J 2010 Nature 467 574
[8] Hofheinz M, Weig E M, Ansmann M, Bialczak R C, Lucero E, Neeley M, O’Connell A D, Wang H, Martinis J M and Cleland A N 2008 Nature 454 310
[9] Schoelkopf R J and Girvin S M 2008 Nature 451 664
[10] Irwin K D and Lehnert K W 2004 Appl. Phys. Lett. 85 2107
[11] Bueno J, Shaw M D, Day P K and Echternach P M 2010 Appl. Phys. Lett. 96 103503
[12] Ivey B I, Lisitsyn S G and Eliashberg G M 1973 J. Low Temp. Phys. 10 449
[13] Zehnder A 1995 Phys. Rev. B 52 12858
[14] Semenov A D, Gol’tsman G N and Sobolewski R 2002 Supercond. Sci. Technol. 15 R1
[15] Semenov A D, Nebosis R S, Gousey R P, Heusinger M A and Renk K F 1995 Phys. Rev. B 52 581
[16] Parker W 1975 Phys. Rev. B 12 3667
[17] Owen C and Scalapino D 1972 Phys. Rev. Lett. 28 1559
[18] Chang J-J and Scalapino D J 1977 Phys. Rev. B 15 2651
[19] Chang J-J and Scalapino D J 1978 J. Low Temp. Phys. 31 1
[20] Kurakado M 1982 Nucl. Instrum. Methods 196 275
[21] Rando N, Peacock A, van Dordrecht A, Foden C, Engelhardt R, Taylor B, Gare P, Lumley J and Pereira C 1992 Nucl. Instrum. Methods 313 173
[22] Hjörner R A, Verhoeve P, Martin D D E, Kozorezov A G, Wigmore J K, Venn R, Groot P J and Jerjen I 2009 J. Appl. Phys. 105 123906
[23] Kozorozov A G, Volkov A F, Wigmore J K, Peacock A, Poelae A and den Hartog R 2000 Phys. Rev. B 61 11807
[24] Goldie D J and Withington S 2013 Supercond. Sci. Technol. 26 015004
[25] Guruswamy T, Goldie D J and Withington S 2014 Supercond. Sci. Technol. 27 055012
[26] de Visser P J, Goldie D J, Diener P, Withington S, Baselmans J J A and Klapwijk T M 2014 Phys. Rev. Lett. 112 047004
[27] De Visser P J, Baselmans J J A, Bueno J, Llombart N and Klapwijk T M 2014 Nat. Commun. 5 3130
[28] Neto A, Llombart N, Baselmans J J A, Baryshev A and Yates S J C 2014 IEEE Trans. Terahertz Sci. Technol. 4 26
[29] Chin C, Oates D E, Dresellhaus G and Dresselhaus M 1992 Phys. Rev. B 45 4788
[30] Kaplan S, Chi C, Langenberg D, Chang J-J, Jafarey S and Scalapino D 1976 Phys. Rev. B 14 4854
[31] Caro J, Coehoorn R and de Groot D 1981 Solid State Commun. 39 267
[32] Kihlstrom K, Simon R and Wolf S 1985 Phys. Rev. B 32 1843
[33] Semenov A et al 2009 Phys. Rev. B 80 054510
[34] Gershenson E M, Gershenson M E, Gol’tsman G N, Lyul’kin A M and Sergeev A V 1990 Sov. Phys.-JETP 70 505
[35] Gladstone G, Jensen M and Schriefler J 1969 Superconductivity ed R D Parks vol 2 (New York: Dekker) ch 13, p 713
[36] Gousev Y P, Gol’tsman G N, Semenov A D, Gershenson E M, Nebosis R S, Heusinger M A and Renk K F 1994 J. Appl. Phys. 75 3695
[37] Kozorozov A G 2011 J. Low Temp. Phys. 167 473
[38] Gershenson E, Gol’tsman G, Potapov V and Sergeev A 1990 Solid State Commun. 75 639
[39] Sergeev A V and Yu Reizer M 1996 Int. J. Mod. Phys. B 10 635
[40] de Visser P J, Baselmans J J A, Yates S J C, Diener P, Endo A and Klapwijk T M 2012 Appl. Phys. Lett. 100 162601
[41] Toth L E 1971 Transition Metal Carbides and Nitrides Refractory Materials vol 7 (New York: Academic) p 279
[42] Schwarz K 1975 J. Phys. C: Solid State Phys. 8 809
[43] Eliashberg G M 1972 Sov. Phys.-JETP 34 668
[44] Parlato L, Latempra R, Peluso G, Pepe G, Cristiano R and Sobolewski R 2005 Supercond. Sci. Technol. 18 1244
[45] Rothen S and Taylor B N 1967 Phys. Rev. Lett. 19 27
[46] Altshuler B and Aronov A 1985 Electron–electron interaction in disordered systems Modern Problems in Condensed Matter Sciences vol 10 ed A Efros and M Pollak (Amsterdam: North-Holland) pp 1–153