# Modified heating theory of nonequilibrium superconductors*

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Most recent experiments on nonequilibrium superconductors have been interpreted in terms of a theoretical model that assumes the recombination time of quasiparticles acts as a bottleneck in the quasiparticle relaxation process. However, in all experiments to date, the actual bottleneck is the escape time of recombination phonons from the superconductor. We show that all previous experiments can be interpreted in terms of a modified heating theory where the quasiparticles remain in both thermal and chemical equilibrium at an effective temperature $T^*$ greater than the helium-bath temperature. The temperature $T^*$ is determined from a consideration of only those phonons with energy greater than $2\Delta$.

During the past few years, the results of several experiments investigating the properties of superconductors driven into nonequilibrium states by optical radiation have been discussed within the context of a model proposed by Owen and Scalapino[1] and Chang and Scalapino.[2-3] Measurements of both the current-voltage characteristics of tunnel junctions[4,5] and the microwave reflectivity of thin films[6,7] agree remarkably well with calculations based on this model with one exception. This general agreement has been interpreted as providing excellent experimental verification of this model of a nonequilibrium superconductor where the quasiparticles are considered to be in thermal equilibrium with the lattice at temperature $T$ but not in chemical equilibrium with the pair state. One of the most striking predictions of this model is a first-order phase transition to the normal state at a sufficiently large density of excess quasiparticles. This first-order transition is not observed either in tunneling measurements[8] or in dc resistivity and microwave reflectivity[7] measurements. It has been speculated[3,7] that a dynamic intermediate state or simple thermal inhomogeneity[8] may be responsible for the absence of this first-order transition in the experiments.

For many reasons all of the experimental results are inconsistent with the trivial assumption that the optical radiation has simply raised the temperature of the superconductor which then remains in both thermal and chemical equilibrium at this elevated temperature. However, in this paper we show that most of the present experimental data including the absence of the first-order transition are consistent with a modified heating theory where the optical radiation increases the number of phonons with energy greater than twice the superconducting energy gap $\Delta$ but leaves the number of phonons with energy less than $2\Delta$ unchanged. These high-energy phonons are assumed to be characterized by an effective temperature $T^*$, while the phonons of less energy are assumed to remain characterized by the ambient temperature $T$. The properties of the illuminated superconductor are assumed to be the thermal-equilibrium properties of an ordinary superconductor at the temperature $T^*$. The search for reasons to explain the absence of the first-order transition predicted by the Owen-Scalapino model and unobserved in the experiments is, of course, unnecessary within the modified heating model since the transition between the superconducting and normal states is second order as for equilibrium superconductors.

We do not assert that this modified heating theory is *the* explanation for all the experiments on optically irradiated superconductors. Rather, it is our intention to demonstrate that alternative models to that proposed by Owen and Scalapino work equally well and that an accurate understanding of nonequilibrium superconductors will require more discriminating experiments and more detailed theoretical models.

The initial motivation for the assumptions of the modified heating model is found in the rate equations of Rothwarf and Taylor,[9]

$$\frac{dN}{dt}=I_0+\frac{2N_\omega}{\tau_B}-RN^2 \tag{1}$$

and

$$\frac{dN_\omega}{dt}=\frac{RN^2}{2}-\frac{N_\omega}{\tau_B}-\frac{N_\omega-N_{\omega T}}{\tau_\gamma}\ , \tag{2}$$

where $N$ is the number density of quasiparticles, $I_0$ is the volume rate of creation of quasiparticles by an external mechanism, $N_\omega$ is the number density of phonons with energy greater than $2\Delta$, $\tau_B^{-1}$ is the mean rate at which these phonons create quasiparticles, $R$ is a recombination coefficient, $\tau_\gamma^{-1}$ is the rate at which phonons of energy greater than $2\Delta$ disappear by processes other than quasiparticle creation, and $N_{\omega T}$ is the thermal equilibrium number density of phonons with energy greater than $2\Delta$. The steady-state solutions are

$$N_\omega/N_{\omega T} = 1 + (\tau_\gamma/2N_{\omega T}) I_0 \quad (3)$$

and

$$(N/N_T)^2 = 1 + (1 + \tau_\gamma/\tau_B) I_0 \tau_R / N_T\,, \quad (4)$$

where $N_T = (2N_{\omega T}/R\tau_B)^{1/2}$ is the thermal-equilibrium number density of quasiparticles and $\tau_R = (RN_T)^{-1}$ is the intrinsic recombination time. All the experiments on optically irradiated superconductors reported in the literature have been performed under conditions where $\tau_\gamma \gg \tau_B$, i.e., where a recombination phonon is far more likely to be reabsorbed by the superconductor with the creation of two quasiparticles than to escape from the superconductor.[5,7] It then follows that the number of phonons and quasiparticles are related by

$$N_\omega/N_{\omega T} \cong (N/N_T)^2\,. \quad (5)$$

The ratio of the number of phonons with sufficient energy to create quasiparticles in the superconductor to the number of phonons in thermal equilibrium at the ambient temperature $T$ is proportional to the square of the quasiparticle departure from equilibrium. This relation indicates that the high-energy phonons are even more out of equilibrium than the quasiparticles. In the limit of $\tau_\gamma/\tau_B \gg 1$, where the phonons are substantially out of equilibrium, it may be incorrect to use the Owen-Scalapino equations and simply interpret the temperature in their equations as the ambient temperature.

The properties of the nonequilibrium superconductors are determined by the number of excess quasiparticles and their energy distribution. The Rothwarf-Taylor equations provide relations between the number of quasiparticles and phonons but provides no information on their energy distribution. In the absence of detailed calculations of the energy distribution function, a number of simplifying assumptions are possible. If the time for the quasiparticles to thermalize with respect to the low-energy phonons is short compared to the intrinsic recombination time, then the distribution function assumed by Owen and Scalapino at the temperature $T$ is presumably the best approximation. If the thermalization time is long, then the energy distribution of the quasiparticles will depend critically on the distribution of phonons with sufficient energy to create quasiparticles. The simplest assumptions to make in the latter case are that the phonons of energy greater than $2\Delta$ can be described by a thermal distribution and an effective temperature $T^*$ adjusted to produce the correct number of phonons, and that the superconductor is described by an ordinary BCS superconductor at the temperature $T^*$. [Numerical calculations indicate that these two assumptions are consistent, i.e., a specified value for $N/N_T$ determines an effective temperature $T^*$ through Eq. (5) as described below. In turn, $T^*$ determines a value of $N_{T^*}/N_T$ nearly equal to the initial value of $N/N_T$.]

An indication that the thermalization time is longer than the intrinsic recombination is obtained from a calculation of the energy at which the probabilities of quasiparticle recombination and relaxation (the emission of a low-energy phonons) are equal.[10] At energies greater than the solid line in Fig. 1 relaxation dominates, i.e., a quasiparticle is more likely to relax before recombining, while below this line recombination dominates. Also shown in Fig. 1 by the dashed line is the average energy of the quasiparticles calculated from

$$\overline{E} = \int_\Delta^\infty E\rho(E) f(E)\, dE \Big/ \int_\Delta^\infty \rho(E) f(E)\, dE\,, \quad (6)$$

where $\rho(E) = E/[E^2 - \Delta^2(T)]^{1/2}$ and $f(E) = [1 + \exp(E/kT)]^{-1}$. The average energy is essentially constant over the full temperature range $0 < T < T_c$. At low temperature $[kT \ll \Delta(0)]$ the energy is determined by the energy gap, while near $T_c$ the energy is determined by the thermal energy $kT$. From this figure we see that a quasiparticle created at a high energy and relaxing toward the average equilibrium energy will probably recombine with another quasiparticle before reaching the average thermal energy. It is thus unlikely that excess quasiparticles will thermalize before recombining.

As an alternative to the Owen-Scalapino model, we propose the following model. An external quasiparticle creation mechanism such as optical radiation produces a steady-state number density of excess quasiparticles described by the normalized quantity $n = (N - N_T)/4N(0)\Delta(0)$, where $N(0)$ is the single-spin density of states. The number of high-energy phonons $N_\omega$ is determined from $n$ through Eq. (5). Since we have made the simplify-

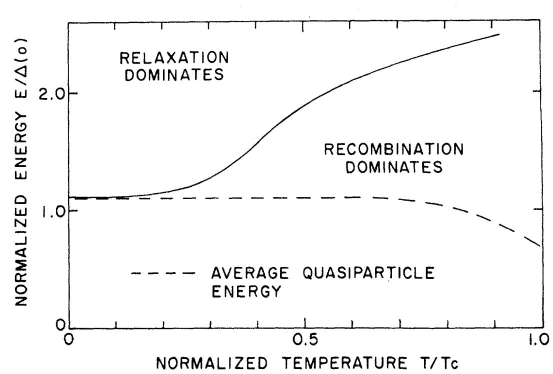


FIG. 1. Solid line indicates the relation between the quasiparticle energy and temperature for which relaxation and recombination are equally probably. The dashed line indicates the average quasiparticle energy of a superconductor in equilibrium.

ing assumption that these phonons are characterized by a thermal distribution at an elevated temperature $T^*$, this effective temperature is given implicitly by the equation

$$\left(\frac{N}{N_T}\right)^2 \cong \frac{N_\omega}{N_{\omega T}} = \left(\frac{T^*}{T}\right)^3 \times \left[ \int_{X_G^*}^{\infty} \left(\frac{x^2\,dx}{e^x - 1}\right) \Big/ \int_{X_G}^{\infty} \left(\frac{x^2\,dx}{e^x - 1}\right) \right], \qquad (7)$$

where $X_G = 2\Delta(T)/kT$ and $X_G^* = 2\Delta(T^*)/kT^*$. Here we have assumed a Debye model to describe the phonon density of states, and used the BCS temperature-dependent energy gap. The integrals can be evaluated to yield the relationship between $T^*/T_c$ and $n$ shown in Fig. 2 for several values of the reduced ambient temperature $t = T/T_c$. Since we have assumed the properties of the illuminated superconductor to be the thermal equilibrium properties at the effective temperature $T^*$, the variation of the energy gap with $n$ is just $\Delta(T^*(n))$, where $\Delta(T^*)$ is the BCS temperature-dependent energy gap. This variation is shown in Fig. 3 together with the results from the Owen-Scalapino model for comparison. Note that for $n \leq 0.1$, the two models are almost identical.

We now have sufficient information to compare this modified heating model to the measurements of Parker and Williams on the current-voltage characteristics of optically irradiated tunnel junctions. They experimentally measured the decrease of the energy gap from which $n$ was determined and the tunneling current $I$ in the range $\Delta(n) \leq eV \leq 2\Delta(n)$. The current in this voltage range is approximately a constant independent of voltage at low temperature. The ratio $I(n)/I(0)$ can be calculated from standard tunneling theory[11] and is given approximately by

$$\frac{I(n)}{I(0)} \cong \exp\left(\frac{\Delta(T)}{kT} - \frac{\Delta(T^*(n))}{kT^*(n)}\right), \qquad (8)$$

where $T^*$ is the effective temperature determined

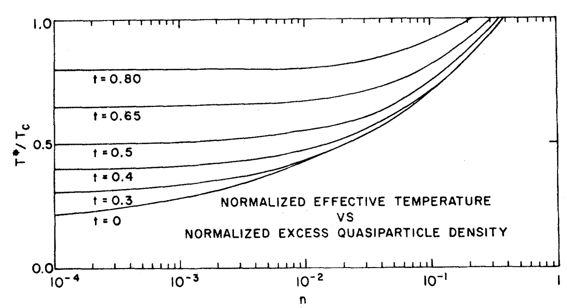


FIG. 2. Normalized effective temperature in the modified heating theory vs the normalized excess quasiparticle density for several values of the reduced ambient temperature $t = T/T_c$.

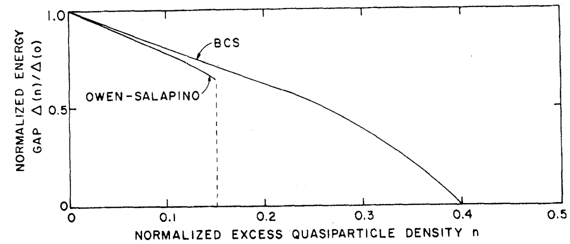


FIG. 3. Normalized energy gap as a function of the normalized excess quasiparticle density for the Owen-Scalapino model at $T = 0$ and the normalized energy gap as a function of the normalized quasiparticle density for a BCS superconductor.

from the value of $n$. All data were obtained with $n \lesssim 0.1$ where the energy gap of both a BCS superconductor and an Owen-Scalapino modified superconductor depend linearly on $n$ through the equation $\Delta(n)/\Delta(0) \cong 1 - 2n$. The original data of Parker and Williams is compared to the modified heating theory in Fig. 4. The agreement with the modified heating model is as excellent as the agreement with the Owen-Scalapino model.

Before we can compare the data of Sai-Halasz *et al.* on the microwave reflectivity of optically illuminated thin films to this modified heating model, we must determine the dependence of the effective temperature $T^*$ on the intensity of the optical illumination. The steady-state solutions to the Rothwarf-Taylor equations can be solved to obtain

$$n = \frac{N_T}{4N(0)\Delta(0)} \left[ \left(1 + \frac{I_0 \tau_{\mathrm{eff}}}{N_T}\right)^{1/2} - 1 \right], \qquad (9)$$

where $\tau_{\mathrm{eff}} \equiv \tau_R(1 + \tau_\gamma/\tau_B)$. In the phonon trapping limit $(\tau_\gamma/\tau_B \gg 1)$, $\tau_{\mathrm{eff}} \cong \tau_R\tau_\gamma/\tau_B = N_T\tau_\gamma/2N_{\omega T}$. If the superconductor is illuminated with monochromatic radiation of frequency $\nu$,

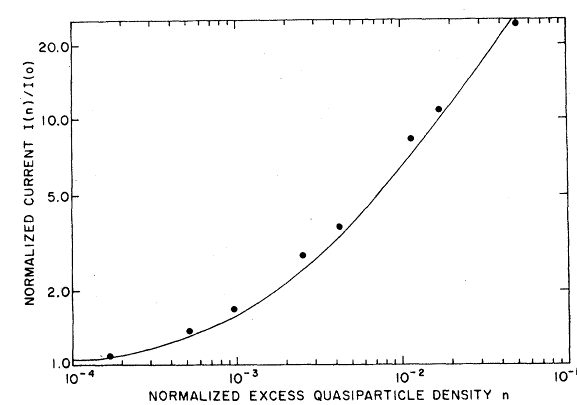


FIG. 4. Normalized tunneling current vs the normalized excess quasiparticle density. The dots are the data from Ref. 4 and the solid line the prediction of the modified heating theory.

$$I_0=(h\nu/\overline{E})(FP/\mathcal{V})(1/h\nu)\ , \tag{10}$$

where $\overline{E}$ is the average energy of an excited quasiparticle, $h\nu/\overline{E}$ is the average number of quasiparticles produced per photon if all the absorbed optical energy is shared by only quasiparticles, $F$ is the fraction of the absorbed optical energy that is shared among the excess quasiparticles, $(h\nu)^{-1}$ is the number of photons per unit energy, $P$ is the absorbed energy flux, and $\mathcal{V}$ is the effective volume of the superconductor. It is assumed that the surface of the superconductor is uniformly illuminated and that the superconductor is sufficiently thin that $n$ is spatially uniform. Equation (9) becomes, after substitution of Eq. (10),

$$n=\frac{N_T}{4N(0)\Delta(0)}\left[\left(1+\frac{\tau_\gamma FP}{2N_{\omega T}\overline{E}\mathcal{V}}\right)^{1/2}-1\right]. \tag{11}$$

In order to obtain an estimate of the fraction of the absorbed energy that is shared among the quasiparticles, we will assume that the absorbed energy is shared among the excess quasiparticles and only those phonons with energy greater than $2\Delta$, i.e., we assume no energy is lost to phonons of energy less than $2\Delta$. An equivalent assumption is that the only means for absorbed energy to leave the superconductor is through the escape from the superconductor of phonons with energy greater than $2\Delta$. With the previous assumption of thermal equilibrium between $N_{\omega T^*}$ and $N_{T^*}$, the fraction $F$ becomes

$$F=\langle N_{T^*}E\rangle_{\rm av}/(\langle N_{T^*}E\rangle_{\rm av}+\langle N_{\omega T^*}\hbar\omega\rangle_{\rm av})=1/(1+R)\ , \tag{12}$$

where $R=\langle N_{\omega T^*}\hbar\omega\rangle_{\rm av}/\langle N_{T^*}E\rangle_{\rm av}$. Using a Debye spectrum for the phonon density of states, we obtain

$$R=\frac{3(kT^*)^4}{8\pi^2 N(0)\Delta(0)(c_s\hbar)^3}\int_{X_G^*}^{\infty}\frac{x^3\,dx}{e^x-1}\times\left(\int_1^{\infty}\frac{x^2\,dx}{(x^2-1)^{1/2}[1+\exp(\frac{1}{2}X_G^* x)]}\right)^{-1}, \tag{13}$$

where $c_s$ is the appropriate average sound velocity and $X_G^*=2\Delta(T^*)/kT^*$. Evaluating the integrals and using values appropriate for Sn [$2\Delta(0)=1.16\times10^{-3}$ eV, $N(0)=1.4\times10^{28}$/eV m$^3$, and $c_s=2\times10^3$ m/sec], the faction $F$ as a function of $T^*$ is obtained. In Fig. 5 the fraction $F$ is plotted against the normalized thermal number of quasiparticles. The fraction $F$ varies between 1 and 0.5 consistent with the value $\frac{3}{4}$ estimated by Sai-Halasz *et al.*

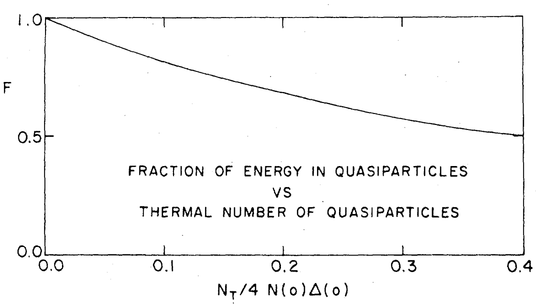


FIG. 5. Fraction of absorbed energy shared among quasiparticles as a function of number of quasiparticles.

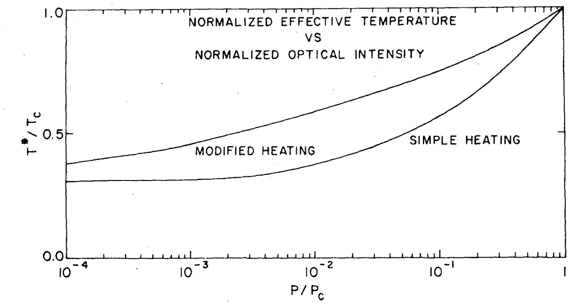


FIG. 6. Normalized effective temperature vs normalized optical intensity for both the simple heating and a modified heating model of an optically irradiated superconductor with ambient temperature $T/T_c=0.30$.

We now have enough information to evaluate Eq. (11). Defining $P_c$ as the optical illumination sufficient to drive the superconductor into the normal state and solving Eq. (11) for $P/P_c$ as a function of $n$, one obtains

$$\frac{P}{P_c}=\frac{2N_{\omega T}V}{\tau_\gamma P_c}\,\frac{\overline{E}(n,T^*)}{F(n,T^*)}\left[\left(\frac{4N(0)\Delta(0)n}{N_T}+1\right)^2-1\right]. \tag{14}$$

$P/P_c$ as a function of $n$ is obtained by evaluating $N_{\omega T}$ at the temperature of the experiments of Sai-Halasz (1.2 K), adjusting $P_c$ so that $P/P_c=1$ when the superconductor becomes normal at $n=0.39$, and using values of $N(0)$ and $\Delta(0)$ appropriate for Sn. Combining this result with $T^*/T_c$ as a function of $n$ from Fig. 2, we obtain $T^*/T_c$ as a function of $P/P_c$. The result is shown in Fig. 6.

For comparison, the result of a simple heating model is also shown in Fig. 6. This model assumes that the only effect of the optical radiation is to raise the temperature while the superconductor remains in complete thermal equilibrium at the elevated temperature. In a thin-film geometry at low temperature, it is reasonable to assume that the steady-state temperature of the superconductor is determined by the heat input and the thermal boundary conductance between the thin film and the substrate and/or the liquid helium. The thermal conductance depends on the difference of the fourth powers of the film and ambient temperatures.[12] Thus the steady-state temperature $T_{ss}$ is given by

$$T_{ss}^4-T^4=aP/P_c\ , \tag{15}$$

where $a$ is adjusted so that $T_{ss}=T_c$ when $P/P_c=1$.

The predicted dependence of the microwave relectivity on the optical power is compared to the

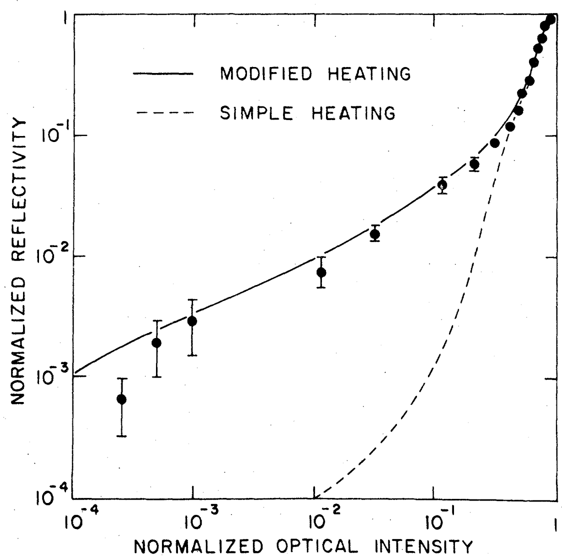


FIG. 7. Normalized reflectivity vs the normalized optical intensity. The dots are the data from Ref. 7 and the solid line the prediction at the modified heating theory.

data of Sai-Halasz in Fig. 7. The curves of normalized reflectivity vs normalized optical intensity are obtained for both the modified and simple heating models by evaluating the microwave reflectivity assuming a BCS superconductor[13] at the appropriate temperature indicated by the curves of Fig. 6. The agreement between the data and the prediction of the modified heating model is as excellent as the agreement between the data and the Owen-Scalapino model. The simple heating model is in obvious disagreement with the data.

The modified heating model also is consistent with many of the original observations on optically illuminated superconductors by Testardi.[14] Testardi observed changes in the dc resistivity of optically illuminated lead films at ambient temperatures well below the temperature range where pure heating effects should be observed. Figure 6 provides a natural explanation for this observation. At a given optical intensity the effective temperature $T^*$ is significantly greater than the increased temperature resulting from pure heating. In the modified heating model the optical energy is concentrated among the high-energy phonons resulting in a greater increase in the number of these phonons, and hence a greater effective temperature, then if the optical energy is distributed among phonons of all energy.

Testardi[14] and others[7] also observed that the dc resistivity of thin superconducting films responds to changes in the optical intensity at a rate significantly faster than any thermal time constant. In the modified heating model, the characteristic response time of the effective temperature $T^*$ is the effective recombination time $\tau_{eff}$. This time is considerably shorter than the rise time of the optical pulses used by Testardi.

The natural expectation when two different models agree equally well with experimental data as do the Owen-Scalapino and the modified heating models is that the phenomenon producing the data is relatively model independent. This is indeed the case with the experimental data on nonequilibrium superconductors. The calculated $I$-$V$ curves of tunnel junctions and the real and imaginary parts of the frequency-dependent conductivity of superconductors primarily depend on the magnitude of the energy gap and are relatively independent of the energy distribution function. It is also possible to show that even the dependence of the energy gap on the excess quasiparticle density $n$ is relatively independent of the energy distribution for $n \lesssim 0.1$. The BCS equations can be solved approximately for many assumed distribution functions and in all cases investigated, $\Delta(n)/\Delta(0) \cong 1 - \beta n$ where $\beta \cong 2$ when $n \lesssim 0.1$. We conclude that the Rothwarf-Taylor equations together with the equation $\Delta(n)/\Delta(0) = 1 - 2n$ are adequate to describe most of the published experimental data on optically illuminated superconductors.

Before one concludes that the modified heating model is consistent with all the experimental data, we hasten to point out that at least three results remain completely unexplained. First is the broadening of the energy gap as measured by the $I$-$V$ curves of tunnel junctions[8]; second is the broad transition of the dc conductivity from infinite to normal[7]; and third is the "delayed" response of the superconductor to short optical pulses.[7,14] These three phenomena make it difficult to discuss simply the behavior of nonequilibrium superconductors at large values of $n$. Nevertheless, it may be that only at large $n$ will data be obtained that will distinguish between the several available models.

In reality, it is probable that neither the Owen-Scalapino nor the modified heating model are complete discriptions of nonequilibrium superconductors. It seems more likely that the distribution function of the excess quasiparticles will be nonthermal and that only a detailed calculation will yield this function. In the absence of such detailed calculations, either of these models can be used as a reference frame to discuss, compare, and interpret experimental data. However, more detailed experiments must be completed before any substantial progress can be made in understanding the details of nonequilibrium superconductors because the gross features of all the available experimental data are essentially dependent only on the number of excess of quasiparticles and the phonon escape time $\tau_\gamma$ and insensitive to the actual quasiparticle energy distribution function.

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