PHYSICAL REVIEW B VOLUME 9, NUMBER 11 1 JUNE 1974

Transport properties of nonequilibrium superconductors*

Jhy-Jiun Chang and D. J. Scalapino
Department of Physics, University of California, Santa Barbara, California 93106
(Received 31 January 1974)

The model developed by Owen and Scalapino for superconductors with excess quasiparticles at thermal equilibrium with the lattice was adapted to investigate the transport properties of the nonequilibrium superconductors. Specifically, we have calculated within this model the frequency and temperature dependence of the ultrasonic-attenuation coefficient, the dynamic electric conductivity, and the temperature dependence of the nuclear-spin relaxation time. It is found that in some cases even a small number of excess quasiparticles can change the transport property drastically.

I. INTRODUCTION

A short time ago, Owen and Scalapino (OS)[1] developed a simple model to describe superconductors with excess quasiparticles. The quasiparticles can be created by external disturbance such as the injection of optical photons or high-frequency phonons ($\hbar\omega > \Delta$, where ω and Δ are, respectively, the phonon frequency and the energy gap of the superconductor) into the superconducting film. In this model, the quasiparticles are treated as if they are in thermal equilibrium with respect to the lattice, although not in chemical equilibrium with respect to the superconducting pairs. This is based on the assumption that the recombination time τ_R, the average time for the quasiparticles to recombine into pairs, is much longer than the time for them to thermalize with respect to the lattice, τ_{th}, so that the quasiparticles spend a long time in the thermalized state, hence one can ignore whatever happened during the short time of thermalization.

These relaxation times have long interested physicists,[2-9] both theorists and experimentalists. It is known that both the thermalization and the recombination of the quasiparticles are mainly due to the emission of phonons. The contributions to τ_{th} and τ_R by the processes shown in Fig. 1 were calculated for Al, Sn, and Pb. It was found that $\tau_R > \tau_{\text{th}}$ except for Pb, for which $\tau_{\text{th}} \gtrsim \tau_R$ when $T - T_c > 0.4T_c$.

Recently there has been interest in the relaxation of branch imbalance too.[9-12] The branch imbalance is measured by the quantity $(N_{k>k_F} - N_{k<k_F})$, where $N_{k \gtrless k_F}$ is the total number of quasiparticles with wave vector $k \gtrless k_F$ and k_F is the Fermi wave vector. The imbalance occurs, for example, when one injects electrons into superconductors. Since the coherence factors of this process are different for the two branches $(u_{k>k_F} > u_{k<k_F})$, we have $N_{k>k_F} > N_{k<k_F}$. However, when photons or phonons are used to create quasiparticles, the coherence factors $(uv' \pm vu')^2$ (+ for photon, − for phonon) are the same for both branches and therefore one does not have to worry about the branch imbalance. Furthermore, even with the presence of the branch imbalance, its effects can be small if τ_Q, the branch mixing time, is much smaller than τ_{th}.

Within the OS model, the gap equation for the nonequilibrium system is

$$[N(0)\,V]^{-1} = \int_{-\omega_c}^{\omega_c} \frac{d\epsilon_k}{E_k} \tanh \tfrac{1}{2}\beta(E_k - \mu^*) \ . \tag{1}$$

This is identical to the BCS gap equation except for the presence of an effective quasiparticle chemical potential μ^*. The value of μ^* can be different from the chemical potential for the paired electrons and depends on temperature and the concentration of the excess quasiparticle. The potential μ^* is determined by the equation

$$n\Delta_0 = \int_0^\infty d\epsilon_k \left(\frac{1}{e^{\beta(E_k - \mu^*)} + 1} - \frac{1}{e^{\beta E_k} + 1} \right) \ . \tag{2}$$

Here n is the excess quasiparticle number measured in units of $4N(0)\Delta_0$ and Δ_0 is the zero-temperature gap for the equilibrium superconductor. Combining Eqs. (1) and (2), the nonequilibrium energy gap Δ and the potential μ^* can be calculated (at least numerically) as functions of n and T. Results are shown in Figs. 2 and 3. The behavior of $\Delta(n, T)$ and $\mu^*(n, T)$ was recently investigated experimentally by Parker and Williams.[8] Their results obtained from tunneling measurements on a superconductor-insulator-nonequilibrium-superconductor junction are in surprisingly good agreement with the theory.

The purpose of this work is to calculate the transport coefficients using the simple model proposed by OS. Specifically, we calculated the ultrasonic attenuation coefficient α, the nuclear-spin relaxation time T_1, and the dynamic electric conductivity coefficient. These transport properties are interesting in their own right and we also hope that later, with more experimental data available for comparison, one will have a better understanding of the relaxation processes in such nonequilib-

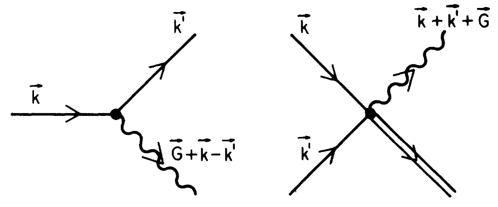

FIG. 1. Diagramatic representations of processes used for calculating τ_{th} (a) and τ_R (b). In diagram (a), a quasiparticle is scattered from state $\vec{k}$ to $\vec{k}'$ with the emission of a phonon of wave vector $\vec{k}'-\vec{k}+\vec{G}$, where $\vec{G}$ is a reciprocal-lattice vector. The umklapp process as well as the normal process is important. The initial electron is "cooled" down by giving energy to phonons. Diagram (b) shows that two quasiparticles combine into a pair and emit a phonon. This is the major contribution to τ_R.

rium systems.

The effects of the excess quasiparticles on the ultrasonic attenuation coefficient α is large when n is large. In fact, at $T=0.08$ when the number of the excess quasiparticles is large enough, the sign of the jump in α at $\hbar\omega=2\Delta(n,T)$ can change from positive to negative and for $n\gtrsim 0.15$ we find that α can become negative over a narrow-frequency region just above $2\Delta/\hbar$ (see Figs. 4 and 5). In order to understand this, consider the low-temperature limit where the excited quasiparticles have relaxed to the gap edge. Then a phonon with energy just larger than twice the nonequilibrium gap will not be able to break pairs because the final quasiparticle states will be already occupied. However, they can stimulate the quasiparticles to recombine into pairs with the emission of a phonon. Thus it is possible to have phonon amplification rather than attenuation.

For the nuclear spin relaxation time T_1, the effects are even more drastic, especially for $T/T_c \ll 1$ (Fig. 6). At $T=0$, the ratio of the relaxation time in the normal state to that in the superconducting state is zero, while for the nonequilibrium case this ratio is approximately $4\mu^{*2}/(\mu^{*2}-\Delta^2)$. Note this becomes larger for *smaller* n in this $T=0$ limit. When the temperature increases the ratio drops; the smaller the n, the faster the decrease in the ratio. However, even for $n=0.01$, the ratio will not drop close to its equilibrium value until $T/T_c\sim 0.5$. The physics behind this is simple. We note that the nuclear Zeeman energy is small, hence the initial and final energies of the scattered electron are essentially the same. Therefore, only quasiparticles come into play. In the equilibrium state, there are simply no quasiparticles at $T=0$, therefore, the ratio is zero. For a superconductor in the nonequilibrium state; however, a small amount of excess quasiparticles changes the chemical potential μ^* from zero to the bottom of the quasiparticle band where both the occupied and unoccupied quasiparticle density of states are large. This results in the large value for the ratio of the relaxation times.

The effects on the dynamic electric conductivity are much less dramatic than that on the ultrasonic attenuation coefficient. Naturally, the additional quasiparticles increase the low-frequency electromagnetic absorption; however, the nonequilibrium changes induced for $\hbar\omega\sim 2\Delta$ are much less than that for α. This is simply a reflection of the difference in coherence factors between the electromagnetic and ultrasonic processes. Detailed calculations are shown in Sec. II followed by some further discussions.

II. TRANSPORT COEFFICIENTS

A. Ultrasonic attenuation

The method which Privorotskii[13] used to calculate the ratio of the attenuation coefficients in equilibrium superconducting and normal states can be trivially generalized for the nonequilibrium case. The expression for this ratio becomes

$$\frac{\alpha_s}{\alpha_N}=\frac{2}{\hbar\omega}\left\{\int_\Delta^\infty dE\frac{E(\hbar\omega+E)-\Delta^2}{(E^2-\Delta^2)^{1/2}[(\hbar\omega+E)^2-\Delta^2]^{1/2}}[f(E-\mu^*)-f(\hbar\omega+E-\mu^*)]\right.$$
$$\left.+\theta(\tfrac{1}{2}\hbar\omega-\Delta)\int_\Delta^{\hbar\omega/2} dE\frac{E(\hbar\omega-E)+\Delta^2}{(E^2-\Delta^2)^{1/2}[(\hbar\omega-E)^2-\Delta^2]^{1/2}}[1-f(E-\mu^*)-f(\hbar\omega-E-\mu^*)]\right\}\ . \qquad (3)$$

This is of the same form as that for the equilibrium case except for the appearance of μ^* in the Fermi functions. We recall that both μ^* and Δ are functions of n and T. The first term is due to the quasiparticle phonon scattering, while the second term comes from the pair breaking and recombination. The two integrations to the right-hand side of Eq. (3) were worked out numerically. Figure 4 shows the ratio for $n=0.01$, $T/T_c=0.08$, 0.50, 0.75, and 0.95. The ratio for $n=0.15$ and $T/T_c=0.08$, 0.25, and 0.46 are shown in Fig. 5.

The results differ from those obtained for the case of an equilibrium superconductor[14] in several respects. For low phonon frequencies where only

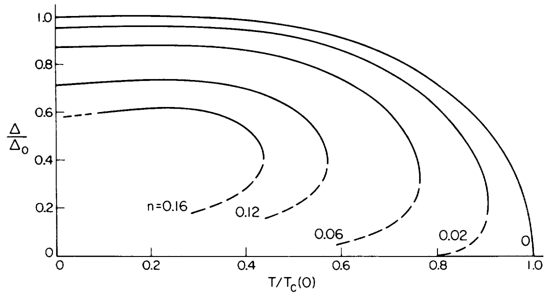

FIG. 2. Temperature dependence of the energy gap for various concentrations of excess quasiparticles. The concentration n is measured in units of $4N(0)\Delta_0$, while Δ and T are normalized, respectively, by the zero-temperature energy gap Δ_0 and critical temperature T_c of the equilibrium state.

quasiparticle phonon scattering contributes to α, the attenuation coefficient for the nonequilibrium state is larger than that for the equilibrium state at the same temperature. This is, of course, simply due to the fact that for nonequilibrium systems there are more quasiparticles available to scatter phonons. When ω is increased, α_s/α_n shows a jump at $\hbar\omega = 2\Delta(T,n)$ which is smaller than $2\Delta(T, n=0)$ for the equilibrium case. Figure 5 shows clearly the strange behavior of $\Delta(T,n)$ at low temperature mentioned by OS, namely, for large fixed n, Δ increases when the temperature increases from $T=0$ and decreases when T is further increased. The magnitude of the jump can be easily calculated from Eq. (3) to be

$$\tfrac{1}{2}\pi[1-2f(\Delta-\mu^*)] \ .$$

The presence of μ^* in the Fermi function always reduces the amount of the jump. In fact, for $\mu^* > \Delta$, the jump changes sign. The physical reason for this has been discussed in Sec. I. For the same reason, the sharp structure in α_s/α_n just above the jump is rounded off at lower temperatures. When $\hbar\omega \gg \Delta$, the ratio reduces to unity as expected.

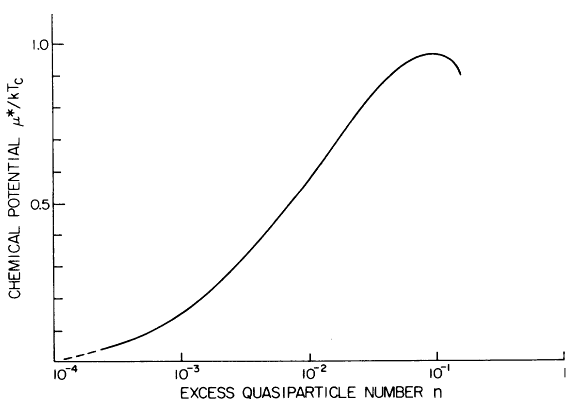

FIG. 3. n dependence of μ^* for $T/T_c = 0.3$.

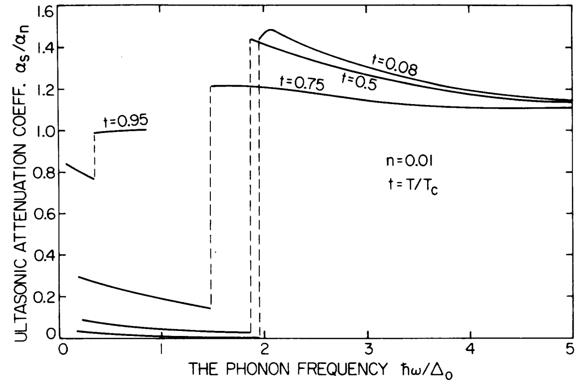

FIG. 4. Frequency dependence of α_s/α_n for $n = 0.01$ and various temperatures $T/T_c = 0.08$, 0.5, 0.75, and 0.95.

B. Nuclear spin relaxation

The nuclear spin relaxation time for equilibrium superconductors has been extensively investigated.[15] In fact, the early measurements by Hebel and Slichter provided an important confirmation of the BCS pairing theory. In the same manner, we believe that the relaxation time for the nonequilibrium state can be measured and should provide insight into the nature of the nonequilibrium state. The ratio of the nuclear relaxation rate in the nonequilibrium superconducting state to that in the normal state is given by

$$\frac{T_{1s}^{-1}}{T_{1n}^{-1}} = 2\int_0^\infty \frac{dE}{k_B T}\rho^2(E)\left(1+\frac{\Delta^2}{E^2}\right) \times f(E-\mu^*)[1-f(E-\mu^*)] \ . \qquad (4)$$

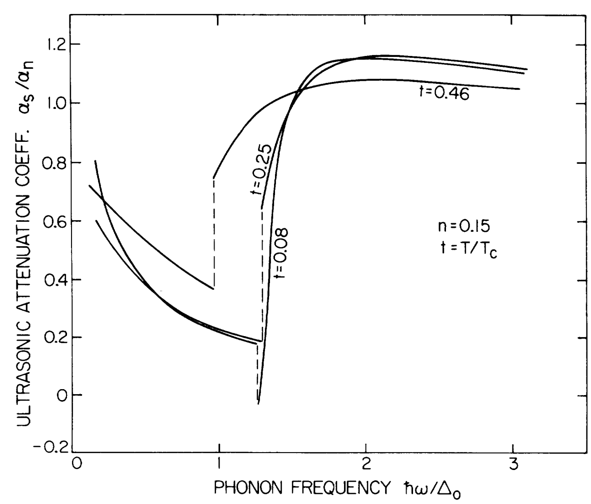

FIG. 5. Frequency dependence of α_s/α_n for $n = 0.15$ and $T/T_c = 0.08$, 0.25, and 0.46.

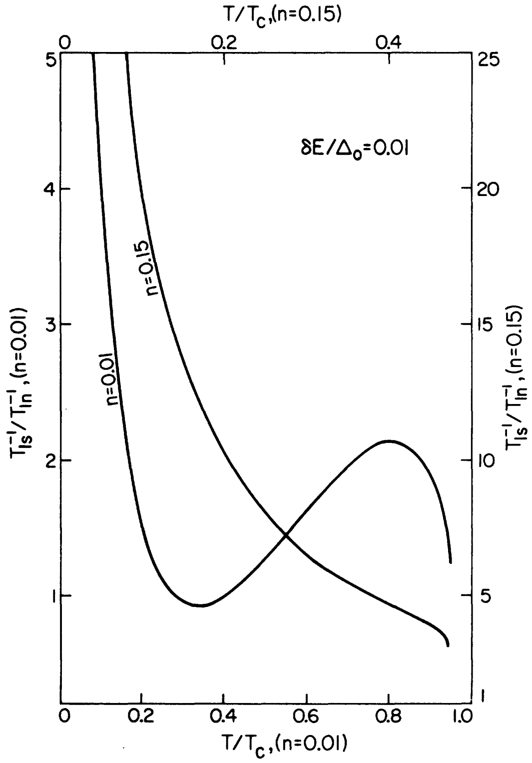

FIG. 6. Temperature dependence of the nuclear relaxation-rate ratio T_{1s}^{-1}/T_{1n}^{-1} for $n=0.01$ and 0.15.

Here $\rho(E)=E/(E^2-\Delta^2)^{1/2}$ is the quasiparticle density of states normalized to $N(0)$ with the nonequilibrium gap and μ^* is the effective potential. To write down Eq. (4), we have neglected the small nuclear Zeeman energy. As pointed out previously by Hebel and Slichter,[15] the integral in Eq. (4) diverges logarithmically at $E=\Delta$. However, this can be healed by introducing an effective width for the quasiparticle states reflecting the gap anisotropy, spatial inhomogeneity, and/or dynamic damping. Following Hebel and Slichter we fold the density of states with a square function of width $2\delta E$ and obtain

$$\rho(E)=0 \qquad \text{if } E\le\Delta-\delta E$$

$$=(1/2\delta E)[(E+\delta E)^2-\Delta^2]^{1/2} \qquad \text{if } \Delta-\delta E<E\le\Delta+\delta E$$

$$=(1/2\delta E)\{[)E+\delta E)^2-\Delta^2]^{1/2} - [(E-\delta E)^2-\Delta^2]^{1/2}\} \quad \text{if } \Delta+\delta E<E \ .$$

Using this density of states, with $\delta E/\Delta_0=0.01$, the integral in Eq. (4) was evaluated numerically. The results for $n=0.01$ and $n=0.15$ are shown in Fig. 6. The most interesting feature is the fast rise of the relaxation time as the temperature decreases at low temperatures. In the equilibrium state, T_{1s}^{-1}/T_{1n}^{-1} drops to zero as $T\to 0$, because there are no quasiparticles available to flip the nuclear spins. While in the nonequilibrium state, at low temperature the excess quasiparticles will pile up at the gap edge where the density of state is large. The Fermi factor there is large too, because μ^* is about the gap energy at sufficiently low temperatures. Equation (9) of Ref. 1 gives

$$\mu^*=\Delta+\tfrac{1}{2}(\Delta_0^2/\Delta)n^2+\text{higher-order terms}$$

and we have $n<0.2$ in the range of our interest. In fact, as $T\to 0$, $f(E-\mu^*)[1-f(E-\mu^*)]\to kT\delta(E-\mu^*)$ and Eq. (4) becomes

$$T_{1s}^{-1}/T_{1n}^{-1}=2\rho^2(\mu^*)\,(1+\Delta^2/\mu^{*2})$$
$$\sim 2(\mu^{*2}+\Delta^2)/(\mu^{*2}-\Delta^2) \quad \text{if } \mu^*-\Delta\gg\delta E \ . \tag{5}$$

Thus the ratio is larger for smaller n as long as n is sufficiently big to satisfy the inequality. The ratio does not drop below unity until $T/T_c(0)\sim 0.3$ even for n as small as 0.01. It is a monotonically decreasing function of temperature for $n=0.15$. Also, the fact that the superconductor is driven normal by the excess quasiparticles before it reaches the critical temperature of the equilibrium state is clearly indicated by the fact that $T_{1s}^{-1}/T_{1n}^{-1}\to 1$ for $T/T_c(n=0)<1$.

C. Dynamic electric conductivity

Following Mattis and Bardeen[16] the dynamic electric conductivity for the nonequilibrium system is found to be

$$\frac{\sigma_s}{\sigma_n}=\frac{\sigma_{1s}}{\sigma_n}-i\frac{\sigma_{2s}}{\sigma_n} \ , \tag{6a}$$

with

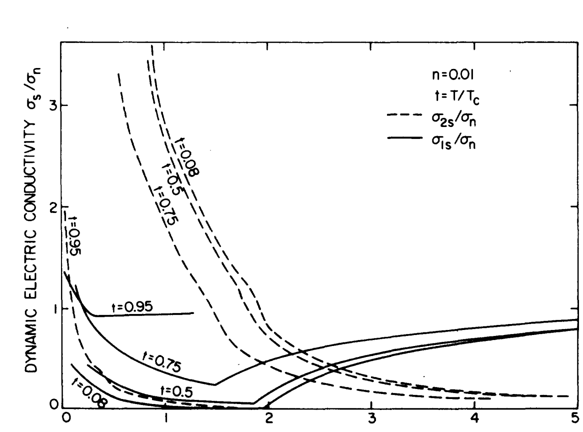

FIG. 7. Dynamic electric conductivity for a nonequilibrium superconductor with $n=0.01$ and $T/T_c=0.08$, 0.5, 0.75, and 0.95.

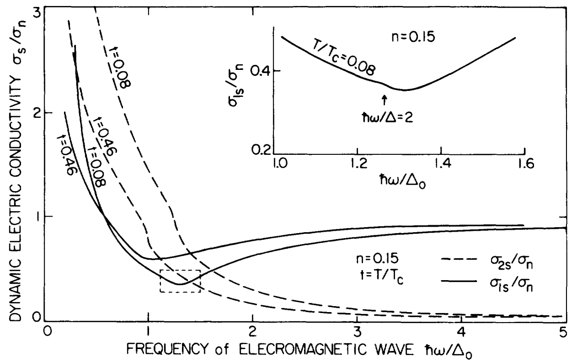

FIG. 8. Dynamic electric conductivity for a nonequilibrium superconductor with $n=0.15$ and $T/T_c=0.08$ and 0.46.

$$\frac{\sigma_{1s}}{\sigma_n}=\frac{2}{\hbar\omega}\int_{\Delta}^{\infty}dE\frac{E(E+\hbar\omega)+\Delta^2}{(E^2-\Delta^2)^{1/2}[(E+\hbar\omega)^2-\Delta^2]^{1/2}}$$
$$\times[f(E-\mu^*)-f(E+\hbar\omega-\mu^*)]+\frac{1}{\hbar\omega}\theta(\hbar\omega-2\Delta)$$
$$\times\int_{\Delta-\hbar\omega}^{-\Delta}dE\frac{E(E+\hbar\omega)+\Delta^2}{(E^2-\Delta^2)^{1/2}[(E+\hbar\omega)^2-\Delta^2]^{1/2}}$$
$$\times[1-2f(E+\hbar\omega-\mu^*)] \tag{6b}$$

and

$$\frac{\sigma_{2s}}{\sigma_n}=\frac{1}{\hbar\omega}\int_{(-\Delta,\Delta-\hbar\omega)_{\max}}^{\Delta}dE$$
$$\times\frac{E(E+\hbar\omega)+\Delta^2}{\{(\Delta^2+E^2)\,[(E+\hbar\omega)^2-\Delta^2]\}^{1/2}}$$
$$\times[1-2f(E-\hbar\omega-\mu^*)]\ . \tag{6c}$$

Values of σ_{1s}/σ_n and σ_{2s}/σ_n for $n=0.01$, $T/T_c=0.08$, 0.5, 0.75, and 0.95 and $n=0.15$, $T/T_c=0.08$ and 0.46 are shown in Figs. 7 and 8. Their general behaviors are very much like that of the equilibrium case. However, we notice that for large n ($n=0.15$) σ_{1s}/σ_n decreases as the frequency of the E-M wave increases from $\hbar\omega=2\Delta$. It is easy to show that the change in slope for σ_{1s}/σ_n at $\hbar\omega=2\Delta$ is given by

$$\tfrac{1}{2}\pi[1-2f(\Delta-\mu^*)]\ ,$$

which is the same as the jump in α_s/α_n at $\hbar\omega=2\Delta$. As mentioned in Sec. I, the reason that σ does not show the same structure exhibited by the ultrasonic attenuation coefficient for $\hbar\omega\sim2\Delta$ arises because of the difference in the coherence factors between the two processes.

III. CONCLUSION

The results for the various transport coefficients presented here were obtained within the context of a special model. As noted in this model the nonequilibrium quasiparticles are taken to be in thermal equilibrium with the lattice, which leads to a Fermi distribution for the quasiparticles. This may well turn out not to give a sufficiently accurate representation of the actual quasiparticle distribution. The deviation of the distribution from $f(E-\mu^*)$ will be reflected in deviations of the observed transport coefficients from those we have calculated. It will be interesting to look for such deviations and analyze them to obtain further information on the nonequilibrium state.

We expect that the general features which we have discussed here will be observed. For example, the decrease in ultrasonic attenuation for $\hbar\omega\gtrsim2\Delta(n)$ relative to the equilibrium state should be observable. Whether it is possible to experimentally sustain conditions such that α becomes negative remains to be seen.

Another area which should be investigated is the question of stability of the excess quasiparticle gas. In the present treatment we have neglected interactions between quasiparticles. In this case, as shown by OS, the system undergoes a first-order phase transition back to the normal state at a certain critical value of $n(T)$. If interactions are included, there may be other possible instabilities. For example, under certain conditions it may happen that spontaneous phonon emission sets in at some critical value of n. In addition, the question of instabilities in the quasiparticle-quasihole or quasiparticle-quasiparticle scattering amplitudes remains to be studied. A particle-hole-like instability would lead to a "droplet" condensation of the quasiparticles such as has been observed in some semiconductors.[17] A particle-particle-like instability would lead to the novel effect of superconductivity in the quasiparticle gas of the nonequilibrium state.

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*Research supported in part by the U. S. Army Research Office, Durham, North Carolina.

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