of larger clusters is not well understood. This behavior may be associated with the higher density of states and lower d-band hybridization at the Fermi level[20] resulting in a higher d-hole density near the occupied portion of the band.

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Experimental Determination of the Quasiparticle Energy Distribution in a Nonequilibrium Superconductor

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We have observed sharp structure in the I-V characteristics of Al-AlO_x-PbBi tunnel junctions induced by tunnel injection of excess quasiparticles into the Al film. Interpretation of this structure provides information on the form of the steady-state quasiparticle energy distribution, showing that it is highly nonthermal. The data also demonstrate the presence of a branch imbalance in the injected Al film.

Nonequilibrium phenomena in superconductors have attracted considerable attention,[1] both because of their intrinsic interest and because the behavior of superconducting devices such as Josephson weak links is often substantially nonequilibrium in nature. An important feature of the problem is the form of the nonequilibrium quasiparticle and phonon energy distributions, $f(E)$ and $n(\Omega)$, respectively. Experiments on phonon generation and detection using superconducting tunnel junctions and phonon fluorescence in superconductors have provided evidence that the phonon distribution can be highly nonthermal.[2-5] Until very recently, however, the most commonly used models of the nonequilibrium state in superconductors have incorporated modified thermal distributions.[6,7] The experimental evidence has so far been insufficient to distinguish among or against these models, or, for that matter, to rule out "simple heating" to the satisfaction of

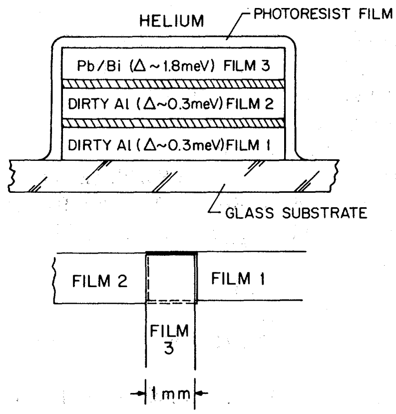

FIG. 1. Asymmetric double–tunnel-junction geometry (cross section and top view) used in these experiments. Quasiparticles were injected into the middle film across the lower oxide layer and were detected by examining the I-V characteristics of the top junction.

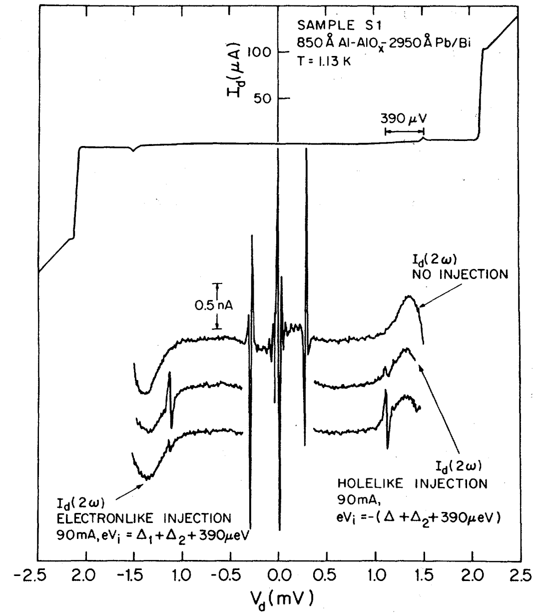

FIG. 2. Detector junction I-V (upper curve) and d^2I/dV^2 (lower curves) characteristics with and without quasiparticle injection.

all. We report here experimental results which show clearly that the form of the quasiparticle distribution in a nonequilibrium superconductor can be extremely nonthermal. The results also provide graphic evidence for the presence of a branch imbalance in the quasiparticle distribution.

Our experiments are similar in concept to those of Miller and Dayem,[8] who first recognized that tunneling can be a sensitive probe of nonequilibrium structure in $f(E)$. Smith[9] has observed such structure using a symmetric double–tunnel-junction, injector-detector system. We have used the asymmetric double-junction system shown in Fig. 1. The Al films were evaporated from alumina crucibles which made them sufficiently dirty to have transition temperatures $T_c \simeq 2.2$ K. A PbBi alloy containing 30% Bi was evaporated to form the top film. The large energy gap ($\Delta_3 \simeq 1.8$ meV) in this film insured that recombination phonons generated in the Al films of the injector junction could not break pairs in, and thus perturb, the PbBi detector film. All three films were dirty enough to reduce substantially the gap-anisotropy contribution to the voltage width of the quasiparticle-tunnel-current rise at the gap-sum voltage in the injector and detector junctions. The voltage width in each was typically 25 μV. The injector junctions typically had resistances of 0.01 Ω, and the detector junctions typically had resistances of 10 Ω. The tunneling-current–voltage characteristic of the detector junction was measured together with the first and second derivatives of the characteristic, with and without a bias current in the injector junction. The derivative traces were made using current modulation techniques very similar to those used in inelastic electron tunneling spectroscopy.[10]

Figure 2 shows the tunneling characteristic of an unperturbed detector junction at $T = 1.13$ K. It shows the well-known current step at voltage $V_d = (\Delta_3 + \Delta_2)/e$ (2.1 mV) and the cusplike peak at voltage $V_d = (\Delta_3 - \Delta_2)/e$ (1.5 mV). The corresponding trace of $I_d(2\omega)$ (the component of the detector-junction current at frequency 2ω due to a voltage modulation of frequency ω, which is closely related to d^2I_d/dV_d^2) shown for the unperturbed detector junction exhibits considerable structure. The two sharp peaks at $V_d = \pm \Delta_2/e$ (± 0.3 mV) were not affected by the application of a small (20 G) magnetic field. These have been previously assigned to a double–particle-tunneling process by other workers.[11,12] The central structure was strongly modulated by a small magnetic field, indicating that it is Josephson related. When holelike excitations were injected into film 2, sharp structure appeared in the second-harmonic signal of the detector junction at voltages $V_d = \pm(\Delta_3 + \Delta_1 - eV_i)/e$, as shown in Fig. 2. V_i here is the injector-junction bias voltage. In this case, larger amplitudes were observed for detector biases corresponding to holelike excitations than for electronlike excitations. When the generator bias was reversed to inject electronlike excitations, the detector structure reversed to show larger electronlike than holelike detected quasiparticles. The ratio of the amplitudes of the holelike and the electronlike structures approached unity as the injector voltage approached the injector-junction gap voltage, and became small at high injector voltages (for electronlike injection). The Josephson-related structure in $I_d(2\omega)$ at low voltages almost completely disappeared with quasiparticle injection of either polarity. In all cases, measurements of the gap decrease during injection showed that the total number of excess quasiparticles present in steady state was less than 10% of the thermal population.

In order to understand this behavior we have extended the theory of Tinkham[13] for the current-voltage characteristic of an S-N tunnel junction in the nonequilibrium state to the S-S case. The result for the detector junction current is

$$I_d = I_{eq} + K\int_0^\infty dE\,\rho_2(E)\left\{\frac{\delta f_<(E)+\delta f_>(E)}{2}\left[\rho_3(E+eV_d)-\rho_3(E-eV_d)\right] + \frac{\delta f_<(E)-\delta f_>(E)}{2\rho_2(E)}\left[\rho_3(E+eV_d)+\rho_3(E-eV_d)\right]\right\}, \quad (1)$$

where $K=G_{NN}/e$, G_{NN} is the normal-state junction conductance, $\rho_n(E)=E(E^2-\Delta_n^2)^{-1/2}$ is the tunneling density of states in film n, and $\delta f_>(E)$ is the excess (above thermal number) distribution of quasiparticles with momentum greater than the Fermi momentum [similarly, $\delta f_<(E)$ is the excess with momentum less than the Fermi momentum].

The first term in Eq. (1) represents the standard equilibrium tunneling integral. The first term in brackets is odd in voltage and is proportional to the sum of the changes in the quasiparticle distribution on both branches of the quasiparticle excitation curve. The second term is even in voltage and proportional to the branch imbalance. This term becomes small for injection energies near the gap edge, implying that the ratio of the holelike to the electronlike structure amplitudes should approach unity as the injection energy is reduced, just as we have observed experimentally.

The top graph in Fig. 3 shows an expanded view of the observed second-harmonic signal at an injector voltage near that shown in Fig. 2. The signal peak-to-peak width is ~ 30 μV, about the width of the current jump at the gap voltage in both the injector and detector junctions. This signal shape corresponds to a peak in dI_d/dV_d (which we have observed directly) and a step in $I_d(V_d)$. The sharpness of this step indicates that little quasiparticle scattering to higher energies occurs after injection at these energies and temperatures. Below the experimental signal, three different assumed distribution functions for the injected quasiparticles in film 2 are shown, together with the associated theoretical signals calculated using Eq. (1). Each distribution function (except the first at the low-voltage end) includes an energy smearing of ~ 30 μeV. For these calculations we assumed that $\delta f_>(E)$ was as shown, and that $\delta f_<(E)=0$. The peak shapes are not strongly dependent on the branch-imbalance ratio assumed. The doubly peaked distribution (bottom) represents contributions from a rounded BCS singularity at $E=eV_i-\Delta_1$ and a second peak at $E=\Delta_2$ due to

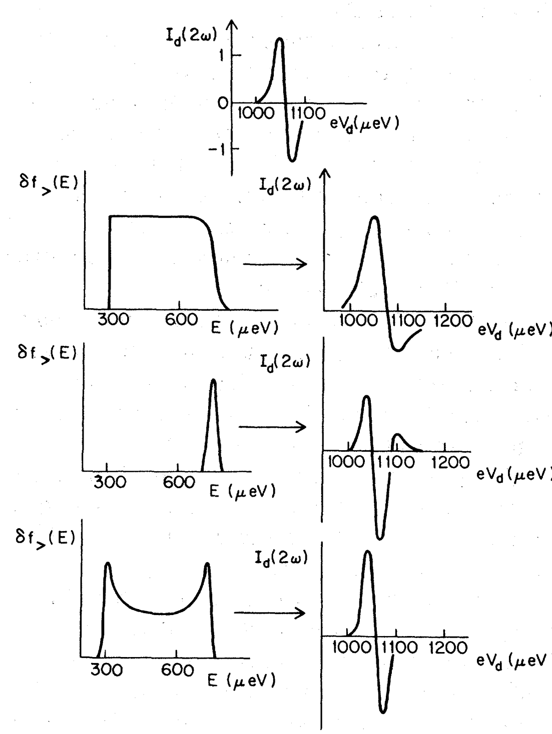

FIG. 3. Comparison of measured second-derivative signal and calculated signals for three different assumed excess quasiparticle distribution functions.

scattering processes. Only this member of the set of assumed distributions matches the shape of the observed signal well. At injector voltages near $(\Delta_1+\Delta_2)/e$, deviations of our calculated signal from that observed were found, indicating that the form of $f(E)$ which we have assumed is incorrect for energies near the gap edge. This is not surprising, since the symmetric distribution we have chosen does not rigorously account for the interplay of quasiparticle scattering, recombination, and branch-mixing processes. We expect that these processes proceed at comparable rates at these temperatures and energies.[14] The distribution functions calculated by Chang and Scalapino[15] with use of a kinetic-equation approach are quite similar to the assumed distribution which gives the best fit to experiment, but show a weaker curvature near the gap edge. We are currently attempting to deconvolute directly the observed curves to obtain quasiparticle distribution functions which can be compared with theoretical predictions. It is abundantly clear from Fig. 3, however, that the nonequilibrium quasiparticle distribution in the injected film 2 in no way resembles a thermal distribution.

We remark that the ratio of electronlike to holelike structure amplitude for electronlike injection (as well as the corresponding ratio for holelike injection) increases more rapidly with injection energy than would be predicted by Eq. (1) if an energy-independent branch-imbalance ratio is assumed. This implies that the branch-imbalance ratio becomes larger (less balanced) as the injection energy is increased, exactly as would be expected from the calculations of Clarke and Paterson.[16] A detailed analysis of this effect will be published elsewhere.

Fuchs *et al.*[17] have recently studied the reduction of the energy gap by tunnel-injected quasiparticles in an asymmetric double-junction structure similar to ours. They find excellent qualitative and quantitative agreement with the predictions of the Owen-Scalapino μ^* model,[6] including an instability of the gap at a critical excess quasiparticle density. While the results of Fuchs *et al.* are certainly important and suggestive, our present work indicates that they should not be taken as support for the detailed validity of the μ^* model, with its modified thermal quasiparticle distribution. We note, as Fuchs *et al.* did, that the observed instability could be associated with a nonequilibrium "intermediate state."[18] The apparent agreement between the experiments of Fuchs *et al.* and the μ^*-model predictions may hence be fortuitous.

In summary, we have detected sharp structure in the I-V characteristic of a superconducting tunnel junction which is due to a highly nonthermal excess quasiparticle energy distribution resulting from tunnel injection of quasiparticles into one of the superconducting films. Analysis of this structure provides a powerful tool for determining the steady-state quasiparticle energy distribution in a nonequilibrium superconductor.

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Orientation Dependence of Free-Carrier Impact Ionization in Semiconductors: GaAs

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The dependence of impact ionization rates upon electric-field orientation is demonstrated in GaAs and is related to specific features of the electronic band structure. This effect has profound implications for the interpretation and implementation of avalanche phenomena in semiconductors.

We demonstrate, for the first time, the existence of orientation dependence of impact ionization by free electrons and holes in semiconductors. Experimental evidence for this effect is given with use of GaAs oriented in the ⟨111⟩ and ⟨100⟩ directions. The results can be understood in terms of calculations[1] which show the strong dependence of the energy threshold for impact ionization on the details of the electronic band structure. In particular, for the ⟨111⟩ direction holes have a higher ionization rate than electrons over the range of electric field 6×10^5 V/cm$>E>3\times10^5$ V/cm. In the ⟨100⟩ direction, however, holes have a higher ionization rate than electrons only for electric fields less than 4.5×10^5 V/cm; for electric fields higher than this value, the electron ionization rate becomes larger than the corresponding rate for holes. Because the performance of a device utilizing the avalanche principle (such as an avalanche photodiode) is a function of the ionization rates, there will always exist at least one preferred direction in any semiconductor which optimizes avalance device performance.[2]

Impact ionization can occur in a semiconductor when a free carrier (electron or hole) can gain sufficient energy, such as from acceleration in an electric field, to create an additional electron-hole pair by excitation of an electron from the valence band to the conduction band. It is now recognized that the impact ionization rate for electrons (α) and for holes (β) are in general different, reflecting the different dynamic properties of electrons and holes. It has been shown recently[3] that $\beta>\alpha$ in GaAs with the electric field in the ⟨100⟩ direction for electric fields in the range 3×10^5 V/cm$>E>2.0\times10^5$ V/cm. We have confirmed the basic features of this result in the same range of electric field using a more sophisticated experimental technique which permits separate injection of pure electron and pure hole currents into the high-field avalanche region. We have extended these measurements to electric fields as high as 6×10^5 V/cm, where the behavior of α and β is quite different. We have given a description of this technique and analysis of experimental measurements in earlier work on impact ionization.[4,5]

Baraff has shown[6] that the impact ionization rates depend strongly on the threshold energy for ionization by the initiating hot carrier. Recently, we have demonstrated[1] that these carrier threshold energies are very sensitive to the details of the energy-momentum relationship for the carrier involved. Threshold energies can be calculated from the electronic band structure with use of a method first described by Anderson and Crowell.[7] However, accurate band structures, now available for the entire Brillouin zone from nonlocal pseudopotential calculations,[8] are required for a proper analysis.

The first experimental evidence indicating the strong dependence of the ionization rates upon the electronic band structure was seen in a study[5] of impact ionization in $GaAs_{1-x}Sb_x$ mixed-crystal alloys. Measurements were made for the elec-