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Excess Quasiparticles in Sn–SnO_2–Sn Tunnel Junction by Charged Particles

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The impulsive response of an S–I–S tunnel junction of Sn has been investigated by means of the α particle irradiation. From the pulse-height dependence of electric signals induced by α particles on the electric current through the junction and on temperature, we have found that the production of excess quasiparticles is essential to the impulsive change in the current-voltage (I–V) characteristics. A simple model is proposed for the qualitative explanation of the observed result.

KEY WORDS: Superconducting tunnel junction / Quasiparticles / Tin / Tunnel current/

INTRODUCTION

The experiment by Testardi[1] in 1971 on the destruction of superconductivity with laser light should be noticed as an important step of investigations to the microscopic behavior of superconductors under an external influence. In the work, he used Pb films of thickness comparable to the optical penetration depth and less than the superconducting coherence length, and found that an electric resistance was produced in the films by the laser light irradiation. The observed rapid changes of state could not account for simple lattice-heating effects, and were considered to be an electronic phenomenon. To explain the observed appearance of electric resistance, he proposed that the electron gas was heated from 3 to 18 K above the lattice temperature.

In the following year, Owen and Scalapino[2] developed a model for a superconductor under an external dynamic pair-breaking influence. In this model, they considered that the recombination time of quasiparticles is much greater than the characteristic scattering time. For this reason, the lattice and electron gas are both treated at the same temperature (lattice temperature), and thus the energy distribution of anomalously large population of free electrons is still characterized by the lattice temperature. By introducing a new parameter which represents the number of excess quasiparticles, they calculated the change in the energy gap as a function of temperature and excess quasiparticles. Their theory predicted the possibility of the first-order superconducting transition under an external influence. The above prediction was followed by several experimental studies,[3~6] by which the decrease in the energy gap was found when superconductors were perturbed by an external influence, but ob-

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servations of the first-order transition were not successful.

For example, Sai-Halasz *et al.*[4] investigated nonequilibrium effects in superconducting Sn films under an external influence. They used microwave reflectivity as a probe of the quasiparticle density. For weak pair breaking, their results were in good agreement with theory and effective quasiparticle recombination time was determined. But for strong pair breaking, instead of the expected first-order transition to the normal state, they found a partially dc-resistive state in the superconductor.

There are only a few studies on the effect of dynamic pair-breaking using α particles, and these are all before Testardi. In 1949, Andrew *et al.*[7] reported the possible use of bulk superconductor as an α detector. Applications of superconducting films to particle detectors were proposed by Sherman.[8] More recently, Spiel *et al.*[9] observed the superconducting-normal transition caused by α particles in thin films of Sn and In. The films used are narrow and an electric current near the critical value is flowing. Therefore, when α particles are irradiated on the strip, the full width of the sample makes transition to the normal state, and this transition is observed by the IR drop. The other is the work by Wood and White,[10] who first observed the electric pulses induced in a superconducting tunnel junction by α particles. They used a crossed-film Sn junction ($\sim$2000 Å) with an insulating oxide layer ($\sim$10 Å), and the junction was bombarded by α particles from a ^{239}Pu source. From an analysis of the induced pulses, they have proposed the possible application of a superconducting tunnel junction to nuclear spectrometry.

The response of a superconducting tunnel junction to incident charged particles is of interest from the view point of microscopic behavior of superconductors under the dynamic pair-breaking influence. In the present work, we measured the impulsive change caused by α particles in I–V characteristics of an S–I–S tunnel junction of Sn. Generally speaking, there are three possibilities for explanations of the impulsive change: (a) Ionization spikes are produced in the insulating oxide layer of the sample when charged particles pass through it. (b) The localized superconducting-normal transition occurs due to the increase of temperature. (c) The excess quasiparticles are produced by α-particle irradiations. Observed spectrum in this work indicates that the excess quasiparticles are essential to the production of electric signals. Details of the experimental procedure are presented here and some qualitative explanations of our results are also attempted.

EXPERIMENTAL

An S–I–S tunnel junction was prepared by means of vacuum evaporation of Sn (99.999%) and glow discharge oxidization. On a glass plate, a Sn film of about 3000 Å was first prepared at the initial pressure of 2×10^{-7} mmHg. During evaporation, the pressure raised to 1×10^{-6} mmHg. After the evaporation, 0.3 mmHg of pure oxygen gas was supplied to the chamber. At 600 V, glow discharge oxidization on Sn film was continued for about 30 sec. The resultant insulating oxide layer was roughly estimated to be 20 Å. The chamber was then again evacuated to 2×10^{-7} mmHg, and the second evaporation of Sn was carried out. The junction thus prepared has total thickness of about 6000 Å and area about 2.5×10^{-5} cm^2. The *I–V* characteristics of the junction measured at 4.2 K gave the electric resistance in the normal state, R_n, as 27 Ω. The sample was always kept below 80 K in order to avoid characteristic deteriorations, probably due to thermal diffusion of the oxide layer.

In Fig. 1 is shown the block diagram of the measuring system, by which two kinds of measurements were performed, *i.e.*, measurements of the spectrum of electric signals induced by α particles, and of the *I–V* characteristics of the junction in thermal equilibrium. A multichannel pulse-height analyzer (MCA) was used for measurement of the spectrum and an X–Y recorder for *I–V* curves. The α source was prepared by depositing ^{210}Po on an Ag film (3 mmϕ, ~1000 Å thick), evaporated on a Mylar substrate. The source was covered by an Ag film (5 mmϕ, ~1000 Å thick) to prevent contaminations. Details of the α source preparation will appear elsewhere.

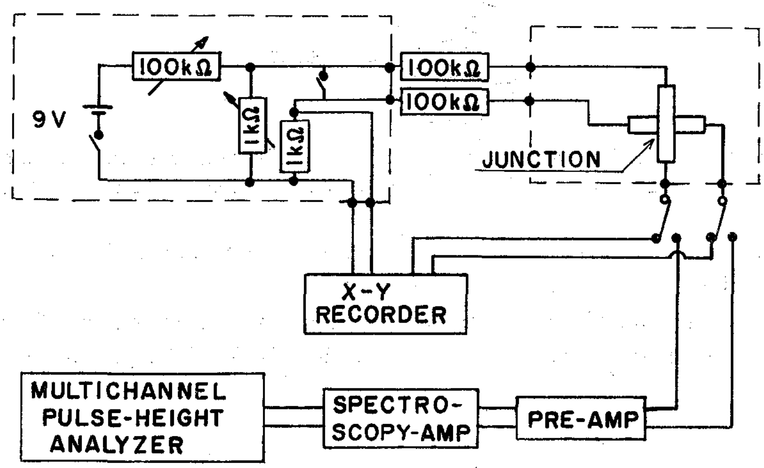

Fig. 1. Block diagram of the measuring system.

In the present experiment, a constant current method was adopted. This means that by supplying a constant current the induced electric signals are observed as changes in the terminal voltage of the junction. In order to ascertain the constancy of current through the junction, two 100 kΩ resistors are connected in series with the sample. These resistors are much larger than that of the junction, it is reasonably considered that the supplied current hold its constancy even when some resistive change is induced in the sample.

In the measurement of induced electric signals, it is necessary to eliminate the background signals. Eliminations of the background contribution from the observed spectrum were carried out by comparing two spectra with and without α-particle irradiations. For measurements of the background, the source is mechanically removed from the face of the junction. After the measurement of the background spectrum, the source is shifted so as to face on the junction. Details of the sample room is shown in Fig. 2. The typical pulse count rate was 600 cpm, which is reasonable from the source strength and the junction-source geometry. Comparisons of

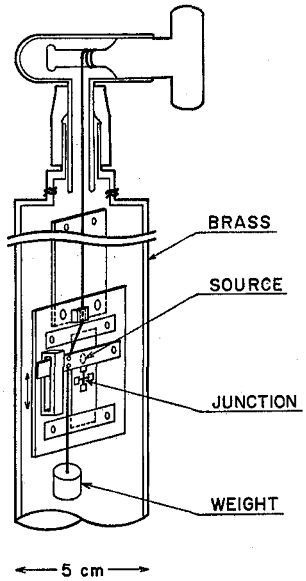

Fig. 2. Details of the sample room. The source (^{210}Po) position is shifted up and down mechanically through a string. The bath temperature is 1.37–4.2 K.

two spectra accumulated in the MCA proved that we successfully observed the signals produced in the junction by α particles (see below).

The I–V characteristics were measured in the temperature region of 1.37–4.2 K, where the X–Y recorder was used instead of the MCA. The results will be given below.

RESULTS AND DISCUSSION

Electric signals induced by α particles were observed with a constant current I supplied through the junction. Some typical pulse-height distributions obtained for several combinations of I and T are shown in Fig. 3. Solid circles give the spectra with α-particle bombardments and open circles are the background obtained after removing the source from the junction face. The distributions apparently depend on I and T, but the signals caused by α particles can surely be distinguished from the background. The reproducibility was confirmed by repeating the up-down shift of the source position.

As seen in the figure, the pulse-height distributions do not form a single mono-energetic peak, but spread over a rather wide region. This broadness may be due to the following reasons: (a) Since the sample thickness ($\sim$6000 Å) is much smaller than the mean range of 5.3-MeV α particles in Sn, the statistical fluctuation of ionization events is relatively large. (b) The incident angle of α particles in the present geometry is 0–45°. This results in different path lengths which are sensitively reflected on the pulse height. (c) The number of quasiparticles which contribute to the signals depends on the incident position of α particles.

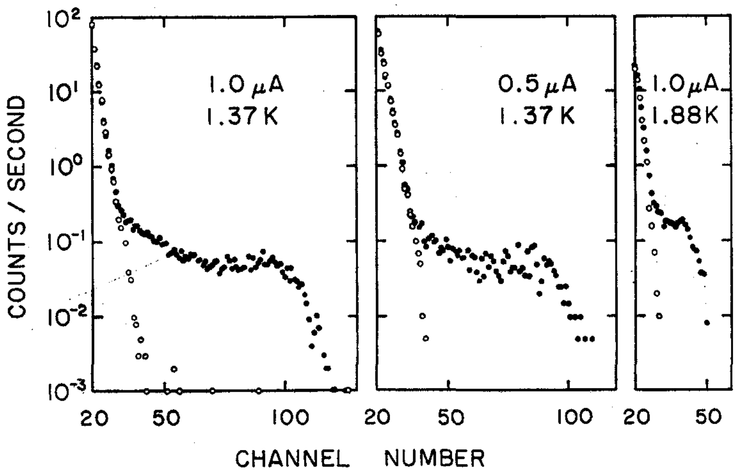

Fig. 3. Typical pulse-height distributions. The measuring periods are, from left to right, 1000, 200, and 500 sec. Solid circles give the spectra with the α irradiation and open circles are without the source.

Taking into account the situation mentioned above, we assume that the maximum channel number C_{max} in each spectrum corresponds to the expected pulse-height of signals under a given condition of I and T. Based on this assumption, comparisons of the results under different combinations of I and T were made. It should be noted here that C_{max} depends on temperature. When temperature goes up, the relaxation time of excess quasiparticles becomes shorter, and consequently the pulse width becomes narrower, resulting in the decrease of gain in the amplifier. Nevertheless, in the present discussion, direct comparisons of C_{max} were made taking no account of the temperature effect on the pulse width.

In Fig. 4 are shown the I–V characteristic curves of the junction in the equilibrium state at T. The points designated by A–M indicate I and V where the pulse-height distributions were measured. Since we concern only with the maximum pulse height in each spectrum, the values of C_{max} are listed in Table I. We reconsider here the three possibilities which may induce the impulsive change in the electric resistance of the junction.

As to the formation of the ionization spikes in the insulating oxide layer of the junction, there is a report by Klein *et al.*[11] They used a silicon dioxide of 3800 Å thick and 2×10^{-2} cm^2 area. Applying 180 V, they irradiated the sample with fission fragments from ^{252}Cf, and observed the charge flow of about 1×10^{-15} Coulomb. In our case, as the electrical potential loaded to the insulating layer is less than 2 mV, the electric field is only 0.2% of the above case. Besides, we used α particles, of which the specific ionization is only a few % of fission fragments. From the different experimental conditions, we concluded that ionization spikes rarely took place in the present case.

There is another possibility which causes electric signals in the junction when irradiated by charged particles. It is the localized superconducting-normal transition

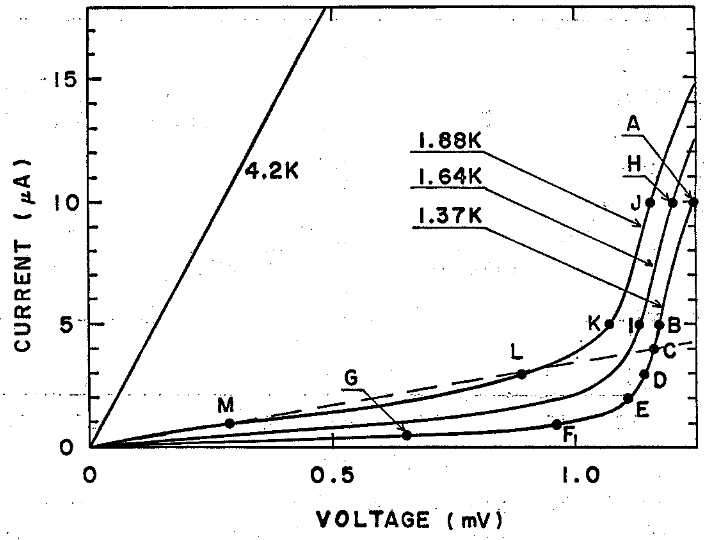

Fig. 4. The I–V characteristic curves of the junction in the equilibrium state at T=1.37–4.2 K. The points A–M indicate the (I, V) combinations where the pulse-height distributions were measured.

Table I. Numerical values of some parameters in data analysis. A–M denote the points where the spectra of induced signals were measured (see Fig. 4).

	T (K)	I (μA)	$\frac{dV}{dI}$ (Ω)	$I\frac{N}{N_T}\frac{dV}{dI}$ (μV)	C_{max}
A	1.37	10	17.5	58.5	<0.19
B		5	12.0	20.0	0.22
C		4	14.5	19.4	0.24
D		3	20.0	20.0	0.28
E		2	60.6	40.4	0.40
F		1	313	105	1.00
G		0.5	971	162	0.93
H	1.64	10	16.5	21.1	<0.19
I		5	17.0	10.9	0.23
J	1.88	10	15.0	9.77	<0.19
K		5	35.1	11.4	0.25
L		3	150	29.3	0.60
M		1	451	29.4	0.43

due to the deposited energy by incident particles. When a small portion of the junction makes the phase transition to the normal state, total electric resistance of the junction decreases (see Fig. 5). For a constant current I, the expected change in the terminal voltage, ΔV, which corresponds to C_{max}, can be expressed by

$$\Delta V = I\frac{R_s S_n (R_n - R_s)}{R_s S_n + R_n (S - S_n)(dI/dV) R_s}, \quad (1)$$

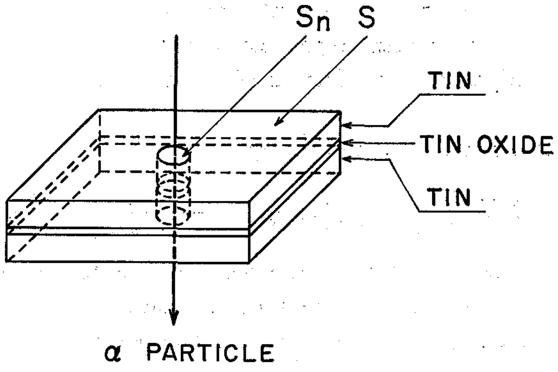

Fig. 5. Schematic figure of the junction bombarded by an α particle. S_n is the area where the phase transition to the normal state occurred, and S is the junction area. The thickness of each tin layer is about 3000 Å and the oxide layer is about 20 Å.

where R_s and R_n are the values of V/I in the equilibrium superconducting and normal states, respectively. S denotes the junction area and S_n is the area which has made the phase transition to the normal state. R_n ($=27\ \Omega$) and S ($=2.5\times10^{-5}$ cm^2) are evidently constant. According to Spiel *et al.*,[9] $S_n\propto 1/(T_c^2-T^2)$, if the heat is not shared with the lattice. In the present experiment, $T<T_c/2$ and besides data compared are for $T=1.37$, 1.64, and 1.88 K. It is therefore reasonably considered that S_n is almost constant for all measurements.

Above discussion suggests that ΔV (or C_{max}) is approximately proportional to $I(\mathrm{d}V/\mathrm{d}I)$ as far as the data at points with an equal value of R_s are compared. As an example, we compare C_{max} for $[I\ (\mu A),\ T\ (K)]=(1,\ 1.88)$, $(3,\ 1.88)$, $(4,\ 1.37)$. All of these three points (M, L, C in Fig. 4) lie on the line of $R_s=280\ \Omega$. From Eq. (1), the ratio of C_{max} should be approximately 1 : 1 : 0.1, if the simple superconducting-normal transition is the case for the production of electric resistance. However, the observed values of C_{max} for these points are 0.43 : 0.60 : 0.24, normalized to C_{max} at point F. From this result, it is difficult to attribute the origin of α-particle signals to the localized superconducting-normal transition.

The third possibility is the change in the I–V characteristics due to excess quasiparticles. In the semiconductor model, the current I flowing through an S–I–S tunnel junction in the thermal equilibrium is given by[12]

$$I=\frac{G}{e}\int_{-\infty}^{\infty}\frac{|E|}{[E^2-\Delta_T{}^2]^{1/2}}\cdot\frac{|E+eV|}{[(E+eV)^2-\Delta_T{}^2]^{1/2}}$$
$$\times[f(E)-f(E+eV)]\Theta[|E|-\Delta_T]\Theta[|E+eV|-\Delta_T]\mathrm{d}E, \qquad (2)$$

where G is the normal-state conductance, V is the applied voltage, $2\Delta_T$ is the energy gap in the equilibrium state at T. $f(E)$ is the Fermi-Dirac distribution function and Θ is the step function. When the sample is irradiated by an α particle, $f(E)$ and Δ_T fluctuate from the equilibrium value, resulting in the change in I–V characteristics.

In order to make rough estimations of the change in $f(E)$, we consider a simple model based on the following two assumptions: (a) The number of excess quasiparticles N produced by an incident α particle is much smaller than that of thermally excited quasiparticles N_T at T. (b) The distribution function of N has the same form as for N_T.

As to the first assumption, N_T is given by

$$N_T = 2N(0)U\int_{\Delta_T}^{\infty} \frac{E}{(E^2-\Delta_T{}^2)^{1/2}} \cdot \frac{1}{\exp(E/k_B T)+1} \mathrm{d}E \tag{3}$$

where $N(0)$ $(=2.12\times10^{22}\ \mathrm{cm^{-3}eV^{-1}})$ is the density of states at the Fermi level for electrons of one spin orientation, U is the junction volume ($50\ \mu\mathrm{m}\times50\ \mu\mathrm{m}\times6000$ Å), and k_B is the Boltzmann constant. The numerical values of N_T in the present sample are

$$\begin{aligned} N_T &= 0.902\times10^8 \text{ at } 1.37\ \mathrm{K} \\ &= 2.31\times10^8 \text{ at } 1.64\ \mathrm{K} \\ &= 4.27\times10^8 \text{ at } 1.88\ \mathrm{K}. \end{aligned}$$

Since the energy loss of an α particle during the passage through the junction is roughly 100 keV, and the average energy expended by the α particle to excite a quasiparticle pair is assumed to be $w=3.5$ meV, N is estimated as 0.3×10^8. Comparisons of the numerical values of N_T and N support that the assumption (a) is reasonable.

As to the second assumption, we have to take into account that the recombination time for quasiparticles to form pairs is much greater than the mean value of scattering time. This fact means, as pointed out by Owen and Scalapino,[2] that the system is in thermal equilibrium although the paired and unpaired electrons are not in chemical equilibrium. So we assume that the distribution function of excess quasiparticles can be considered to have the same form as for the thermally excited quasiparticles.

The above two assumptions permit us to adopt an approximate expression for the total quasiparticle distribution as $f(E)+\delta f(E)$, where $\delta f(E)$ is the fractional change in $f(E)$ due to an incident particle. Thus we get the following simple relations:

$$f(E)+\delta f(E) = \frac{N_T+N}{N_T} f(E) \qquad \text{for} \quad E>\Delta \tag{4}$$

$$1-[f(E)+\delta f(E)] = \frac{N_T+N}{N_T}[1-f(E)] \qquad \text{for} \quad E<-\Delta \tag{5}$$

where 2Δ denote the energy gap in the state with excess quasiparticles. From Eqs. (4) and (5), we get

$$\delta f(E) = \frac{N}{N_T} f(E) \qquad \text{for} \quad E>\Delta \tag{6}$$

$$= -\frac{N}{N_T}[1-f(E)], \qquad \text{orf} \quad E<-\Delta \tag{7}$$

For a uniform nonequilibrium superconductors, Scalapino *et al.*[2,13] found a simple relation between Δ and Δ_T;

$$\frac{\Delta}{\Delta_T} \simeq 1-2n, \tag{8}$$

if the conditions $n=N/4N(0)U\Delta_0\ll 1$, $T<T_c$, and $T\nleq T_c$ are satisfied, where $2\Delta_0$ denote the energy gap in the equilibrium state at $T=0$. Similar to other excitation phenomena like an electron-hole pair production in a semiconductor, we may put $N\simeq Q/w$ and $w=6\Delta_0$, where Q is the energy loss of an α particle in the junction and w is the mean energy loss by the particle per excited charge carrier pair. Thus $n\simeq 8\times 10^{-4}$ for $Q=100$ keV, and from Eq. (8) we find no appreciable change in the energy gap occurs by the production of excess quasiparticles. Henceforth, Δ is replaced by Δ_T.

The above discussion concerning the fluctuation of $f(E)$ and Δ_T gives approximate expression to the change in I given by Eq. (2). We define ΔI as the induced current corresponding to $\delta f(E)$.

Case 1: $eV<2\Delta_T$

$$\begin{aligned}\Delta I=&\int_{\Delta_T}^{\infty} g(E)[\delta f(E)-\delta f(E+eV)]\mathrm{d}E\\ &+\int_{-\infty}^{-\Delta_T} g(E)[\delta f(E)-\delta f(E+eV)]\Theta(|E+eV|-\Delta_T)\mathrm{d}E. \end{aligned}\tag{9}$$

$$g(E)=\frac{G}{e}\cdot\frac{|E|}{[E^2-\Delta_T{}^2]^{1/2}}\cdot\frac{|E+eV|}{[(E+eV)^2-\Delta_T{}^2]^{1/2}}.\tag{10}$$

Using Eqs. (6), (7), and (2), we get

$$\begin{aligned}\Delta I=&\frac{N}{N_T}\left\{\int_{\Delta_T}^{\infty}+\int_{-\infty}^{-\Delta_T-eV}\right\}g(E)[f(E)-f(E+eV)]\mathrm{d}E\\ =&\frac{N}{N_T}\int_{\Delta_T}^{\infty} g(E)[f(E)-f(E+eV)]\mathrm{d}E\\ &+\frac{N}{N_T}\int_{-\infty}^{-\Delta_T} g(E)[f(E)-f(E+eV)]\Theta(|E+eV|-\Delta_T)\mathrm{d}E\\ =&\frac{N}{N_T}I.\end{aligned}\tag{11}$$

Case 2: $eV>2\Delta_T$

$$\begin{aligned}\Delta I=&\left\{\int_{\Delta_T}^{\infty}+\int_{\Delta_T-eV}^{-\Delta_T}+\int_{-\infty}^{-\Delta_T-eV}\right\}g(E)[\delta f(E)-\delta f(E+eV)]\mathrm{d}E\\ =&\frac{N}{N_T}\left\{\int_{\Delta_T}^{\infty}+\int_{\Delta_T-eV}^{-\Delta_T}+\int_{-\infty}^{-\Delta_T-eV}\right\}g(E)[f(E)-f(E+eV)]\mathrm{d}E\\ &-\frac{N}{N_T}\int_{\Delta_T-eV}^{-\Delta_T} g(E)\mathrm{d}E\\ =&\frac{N}{N_T}(I-I_0).\end{aligned}\tag{12}$$

$$I_0=\int_{\Delta_T-eV}^{-\Delta_T} g(E)\mathrm{d}E.\tag{13}$$

From Eqs. (11), (12), and (13), as a general expression, one gets

$$\Delta I=\frac{N}{N_T}(I-I_0).\tag{14}$$

$$I_0 = \begin{cases} \int_{\Delta_T - eV}^{-\Delta_T} g(E)\mathrm{d}E & \text{for } eV > 2\Delta_T \\ = 0 & \text{for } eV < 2\Delta_T . \end{cases} \tag{15}$$

In the present measurements, a constant current was supplied, and the signals were measured as changes in voltage, ΔV. Thus

$$\Delta V = -\frac{\mathrm{d}V}{\mathrm{d}I}\Delta I = -\frac{N}{N_T}\frac{\mathrm{d}V}{\mathrm{d}I}(I - I_0) . \tag{16}$$

In Eq. (16), ΔV (or C_{max}) is not simply proportional to I, but importance is the difference between I and I_0. The physical meaning of I_0 is apparent from Eq. (13), *i.e.*, I_0 is the current from the energy states under the energy gap in one layer of Sn (at $T=0$, but the energy gap holds at $2\Delta_T$) to the states above the gap in another layer of Sn. Hence, for $eV<2\Delta_T$, I_0 is zero, but when eV becomes larger than $2\Delta_T$, I_0 increases quite rapidly as V increases (see Fig. 6). This results in the decrease of the relative contribution of excess quasiparticles, and consequently ΔV becomes smaller.

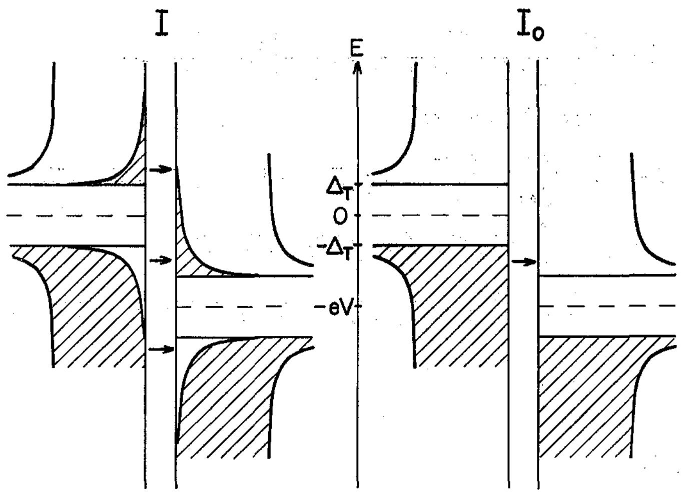

Fig. 6. Density of states in an S–I–S tunnel junction. I is the tunnel current flowing through the junction in the thermal equilibrium, and I_0 is the current from the energy states under the energy gap in one layer of Sn ($T=0$, but the gap is $2\Delta_T$) to the states above the gap in another layer of Sn.

The above situation is well observed in the measurements for points A, B, C, and D in Fig. 4. The relative values of C_{max} for these points are <0.19: 0.22: 0.24: 0.28, which increase as V decreases ($T=1.37$ K for all points). A similar explanation can be applied when comparisons of C_{max} for points B, I, and K are made. Measurements at these points were made at different temperatures, 1.37, 1.64, and 1.88 K. But the current was kept constant, $I=5$ μA. Since the potential V at point B is larger than the estimated energy gap $2\Delta_{1.37} \simeq 1.14$ mV, the contribution of I_0 is significant at this point. On the contrary, at points I and K, the potential V produced by the current of 5 μA, is comparable or smaller than the energy gap $\Delta_{1.64} \simeq 1.12$ mV and $2\Delta_{1.88} \simeq 1.10$ mV, *i.e.*, the contribution of I_0 is small enough or zero in these cases. The observed values of C_{max} for B, I, and K are 0.22, 0.23, and 0.25. As expected from the above discussion, for a constant value of I, C_{max} becomes larger as T increases.

Now we compare points F and M, where $I=1$ μA, but $T=1.37$ and 1.88 K, respectively. It is noted that in these points $I_0=0$. Our measurements gave that C_{max} of F is larger than that of M. From Eq. (16), this is attributed to N_T, of which the numerical values are given before.

As the last example, we compare points D and L, where $I=3$ μA, but $T=1.37$ and 1.88 K, respectively. As in the above case, $I_0=0$ for both points. Since N_T for point D is evidently smaller than that for L, it may be expected that C_{max} for D is larger than that for L. However, our observations gave an opposite result, *i.e.*, C_{max} for D is smaller than for L. This opposition can be attributed to the fact that $\mathrm{d}V/\mathrm{d}I$ in Eq. (16) for D is much smaller than that for L, as given in Table I.

In conclusion, our simple model described in this paper well explains qualitatively the present experimental results. This leads us to the conclusion that excess quasiparticles are essential for the impulsive change in the I–V characteristics.

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