#### Quasiparticle excitation in a superconducting tunnel junction by $\alpha$ particles

M. Kurakado and H. Mazaki Institute for Chemical Research, Kyoto University, Kyoto, Japan (Received 20 November 1979)

The electric signals induced by  $\alpha$  particles in an S-I-S tunnel junction of Sn were measured in the temperature region of 1.37-4.2 K. From the pulse-height dependence on the electric current through the junction and on temperature, it was found that the impulsive change in the current-voltage (I-V) characteristics in the sample cannot be explained simply by the localized superconducting-normal transition due to the temperature increase. By adopting a simple model, however, it has been revealed that excess quasiparticles are essential to the characteristic change in the junction.

# I. INTRODUCTION

In 1971, Testardi<sup>1</sup> found that a superconducting Pb film irradiated by a laser beam yields an electric resistance even at temperature below the superconducting transition temperature  $T_c$ . In the following year, Owen and Scalapino<sup>2</sup> published a model for a superconductor under an external dynamic pair-breaking influence. In this model, however, the production of electric resistance observed by Testardi cannot be explained, but some interesting predictions are made. Their theory predicted the possibility of the firstorder superconducting transition under an external influence, and showed that the energy gap  $2\Delta$  is a function of the excess quasiparticle density. Motivated by the above prediction, several experimental studies<sup>3-6</sup> were made, by which the decrease in  $\Delta$  was found when superconductors were perturbed by an external influence, but observation of the first-order transition was not successful.

For example, according to the experiment by Sai-Halasz *et al.*,<sup>4</sup> the first-order transition temperature predicted by Owen and Scalapino corresponds to the temperature at which electrical resistance develops in a superconductor. Another interesting work was made by Schuller and Gray,<sup>6</sup> who studied, using a laser beam, the relaxation time of  $\Delta$  at temperatures near  $T_c$ .

Concerning the effect of  $\alpha$ -particle irradiations on superconductors, a few investigations have so far been published. One is the work by Spiel *et al.*<sup>7</sup> who observed the appearance of electric resistance by  $\alpha$ particle irradiations. In a superconducting strip, where an electric current near the critical value is flowing, the irradiation effect appears as the change in the terminal voltage, showing the production of resistance. The other is the work by Wood and White,<sup>8</sup> where a Sn-SnO<sub>2</sub>-Sn tunnel junction was bombarded with  $\alpha$  particles and the resulting electric signals were observed. Based on their observations, they have proposed the possibility of utilizing a superconducting tunnel junction in nuclear spectrometry.

Generally speaking, there are three possibilities which cause the impulsive change in I-V characteristics of a superconducting tunnel junction, when irradiated by  $\alpha$  particles: (a) Ionization spikes are produced in the insulation 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  $\alpha$ -particle irradiations.

Unfortunately, the microscopic mechanism of the production of electric resistance due to an external influence is not clearly understood yet. It is therefore worthwhile to get more information on the nonequilibrium state of superconductors caused by an external effect. In the present work, we performed an experimental study on the effect of  $\alpha$ -particle irradiations on an *S*-*I*-*S* tunnel junction of Sn. Measurements of the spectrum indicate that the excess quasiparticles are essential to the production of the electric signals observed. Details of the experimental procedure are presented here and some qualitative explanations of our results are also attempted.

## II. EXPERIMENTAL

A crossed-film tunnel junction of Sn was prepared by the standard method, vacuum evaporation of Sn (99.999%), and glow discharge oxidation. A Sn film of about 3000 Å was first prepared on a glass plate in a vacuum chamber (initially  $2 \times 10^{-7}$  Torr). Then 0.3 Torr of pure oxygen gas was supplied in the chamber. By applying 600 V to the glow discharge electrode, oxidation of the Sn film was continued for 30 sec. The resultant SnO<sub>2</sub> film was roughly estimated to be 20 Å. After evacuating the chamber, the second vacuum evaporation of Sn was made at the initial pres-

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sure of  $2 \times 10^{-7}$  Torr. The sample thus prepared has a junction area of about  $2.5 \times 10^{-5}$  cm<sup>2</sup> and its total thickness is about 6000 Å. The electric resistance of the sample at 4.2 K,  $R_n$ , is 27  $\Omega$ . Since this kind of superconducting tunnel junction deteriorates rather rapidly at room temperature, the sample was always kept below 80 K.

The block diagram of the measuring system is shown in Fig. 1, where a usual 4-terminal network is used. With the measuring system, two kinds of measurements were made, i.e., observations of the signals produced with  $\alpha$ -particle irradiations [to the multichannel pulse-height analyzer (MCA)], and of the *I-V* characteristics at various temperatures below 4.2 K (to the XY recorder). The  $\alpha$  source was prepared by depositing <sup>210</sup>Po on an Ag film (3 mm diam, ~1000 Å thick), which was previously evaporated on a Mylar substrate. In order to prevent contamination the source was covered by evaporating Ag (5 mm diam, ~1000 Å) on it. Details of the  $\alpha$  source preparation will appear elsewhere.

Since the sample is connected in series with two 100 k  $\Omega$  resistors, the electric current through the sample is reasonably considered to be constant even when some resistive change is induced in the junction. However, as the signal-to-noise ratio of the system is poor even at the optimum bias condition (0.95)mV, 1  $\mu$ A at 1.37 K), it is necessary to deduct, in each experimental run, the background from the spectrum with the  $\alpha$  source. By this reason, two measurements were alternately performed, with and without irradiations of  $\alpha$  particles. All other experimental conditions in a set of measurements were of course kept equal. For measurements of the background, the  $\alpha$  source is mechanically removed from the face of the junction. After the measurement of the background spectrum, the source is shifted to face the junction. The typical pulse-count rate was 600 cpm, which is reasonable from the source strength and the junction-source geometry. Comparisons of two spectra accumulated in the MCA with



FIG. 1. Block diagram of the measuring system.

and without the source, proved that we successfully observed the signals produced in the sample by  $\alpha$  particles (see below).

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

#### **III. RESULTS AND DISCUSSION**

As mentioned in the preceding section, accumulations of the output signals from the junction were made at constant current and temperature. The pulse-height distribution for different values of current *I* and temperature *T* were sequentially measured. In Fig. 2 the typical spectra observed for  $I = 1 \ \mu A$ , T = 1.37 K are shown. The solid circles give the spectrum with the  $\alpha$  source and the open circles are the background obtained without the source. In the figure the signals of  $\alpha$  particles can surely be distinguished. The reproducibility of these spectra were confirmed by repeating the up-down shift of the source position.

The distribution of  $\alpha$ -particle signals does not form a single monoenergetic peak, but spreads to a rather wide region. Analytical explanation cannot be made at the present stage, but taking into consideration the following points, the broadened spectrum may be interpreted: (a) Since the sample thickness is much



FIG. 2. The spectra measured for a constant current  $I = 1.0 \ \mu A$  at T = 1.37 K. Solid circles give the spectrum with the  $\alpha$  source and open circles are obtained without the source. Both spectra were obtained for 1000-sec measurements.  $C_{max}$  is the maximum pulse height of the induced signals and their values are listed in Table I. No induced signals by  $\alpha$  particles were observed at  $T > T_c$ .

smaller than the mean range of  $\alpha$  particles, statistical fluctuation of energy losses by ionization is relatively large even for an equal path length. (b) The pulse height sensitively reflects the span of particle path length. In other words, different incident angles  $(0-45^{\circ})$  of  $\alpha$  particles in the present geometry directly affect the energy deposition in the sample. (c) The number of excess quasiparticles which contribute to the change in the I-V characteristics may depend on the incident position of  $\alpha$  particles on the junction.

For simplicity, 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 conditions 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 without taking into account the effect of *T* on the pulse width.

In Fig. 3 the I-V characteristics of the sample in the equilibrium state are shown, as well as the typical experimental points where the pulse-height distributions were measured. The values of  $C_{\text{max}}$  in each measurement are listed in Table I. It is convenient



FIG. 3. Solid lines are the *I*-*V* characteristic curves of the *S*-*I*-*S* tunnel junction of Sn, in the temperature region of 1.37–4.2 K. Some of the curves at 1.88 < *T* < 4.2 K are not shown in the figure. Solid circles designated by A - M indicate the points where the spectra of signals induced by  $\alpha$  particles were measured.

here to reconsider the three possibilities (see Sec. I) which induce the impulsive change in the electric resistance of the junction.

As to the formation of the ionization spikes in the insulation layer of the sample, there is a report by Klein *et al.*<sup>9</sup> They used a silicon dioxide of 3800 Å thick and  $2 \times 10^{-2}$  cm<sup>2</sup> area. Applying 180 V, they irradiated the sample by fission fragments from <sup>252</sup>Cf,

$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$		Т	N <sub>T</sub> <sup>a</sup> (10 <sup>8</sup> )	$\frac{N^{b}}{N_{T}}$	Ι (μÅ)	<i>V</i> (mV)	$\frac{dV}{dI}$ ( $\Omega$ )	$-\Delta V_q$ ( $\mu$ V)	C <sub>max</sub> <sup>c</sup> (Relative)
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		(K)							
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	A				10	1.24	17.5	21.3	< 0.19
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	В				5	1.17	12.0	14.6	0.22
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	С				4	1.15	14.5	17.6	0.24
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	D	1.37	0.902	0.321	3	1.14	20.0	24.4	0.28
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	E				2	1.11	60.6	38.9	0.40
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	F				1	0.95	313	100	1.00
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	G				0.5	0.65	971	156	~0.93
I 1.64 2.31 0.126 5 1.12 16.0 10.1 0.23 J 10 1.14 15.0 6.5 <0.19 K 5 1.05 35.1 11.9 0.25 L 1.88 4.27 0.0678 3 0.85 150 30.5 0.60	Н				10	1.19	12.8	8.4	< 0.19
J 10 1.14 15.0 6.5 <0.19 K 5 1.05 35.1 11.9 0.25 L 1.88 4.27 0.0678 3 0.85 150 30.5 0.60	I	1.64	2.31	0.126	5	1.12	16.0	10.1	0.23
K      5      1.05      35.1      11.9      0.25        L      1.88      4.27      0.0678      3      0.85      150      30.5      0.60	J				10	1.14	15.0	6.5	< 0.19
L 1.88 4.27 0.0678 3 0.85 150 30.5 0.60	Κ				5	1.05	35.1	11.9	0.25
	L	1.88	4.27	0.0678	3	0.85	150	30.5	0.60
M 1 0.24 451 30.6 0.43	М				1	0.24	451	30.6	0.43

TABLE I. Numerical values of  $-\Delta V_q$  obtained on the basis of the excess quasiparticle model and the observed  $C_{\text{max}}$  at points A - M (see Fig. 3): The values of some parameters are also given.

<sup>a</sup>The number of thermally excited quasiparticles in the equilibrium state at T.

<sup>b</sup>The number of excess quasiparticles produced by an incident  $\alpha$  particle. N is approximated by

 $Q/6\Delta_0$  and the numerical values in the table are obtained for Q = 100 keV.

<sup>c</sup>The maximum channel number in each energy spectrum (normalized to the value at point F).

1.0

0.5

C<sub>max</sub>

and observed the charge flow of about  $1 \times 10^{-5}$  C. In our case, as the electrical potential loaded to the insulation layer is less than 2 mV, the electric field is only 0.2% of the above case. Besides, we used  $\alpha$  particles, of which the specific ionization is only a few percent of fission fragments. From the different experimental conditions, we concluded that ionization spikes rarely took place in the present case.

We proceed on the second possibility, i.e., a localized superconducting-normal transition due to an increase of temperature. In this case, by an incident  $\alpha$ particle, a portion of the junction area makes the phase transition to the normal state and consequently the total resistance of the junction decreases. Considering the parallel combination of a normal region and the highly nonlinear I-V characteristic of the tunnel junction the total current I supplied from a constant current source is approximately expressed by

$$I = \frac{S_n}{S} \left( \frac{IR_s + \Delta V_h}{R_n} \right) + \frac{S - S_n}{S} \left( I + \frac{dI}{dV} \Delta V_h \right) \quad , \tag{1}$$

where S denotes the junction area and  $S_n$  is the area which has made the phase transition to the normal state.  $R_s$  and  $R_n$  are the values of V/I in the equilibrium superconducting and normal states, respectively.  $\Delta V_h$  is the expected change in the terminal voltage. From Eq. (1) we get

$$\Delta V_h = -\frac{IS_n(R_s - R_n)}{S_n + R_n(S - S_n)(dI/dV)} \quad . \tag{2}$$

In this equation  $R_n (= 27 \ \Omega)$  and  $S (= 2.5 \times 10^{-5} \ \mathrm{cm}^2)$  are evidently constant. According to Spiel et al.<sup>7</sup>  $S_n \propto 1/(T_c^2 - T^2)$  if the heat is not shared with the lattice. Therefore,  $S_n$  is constant as far as comparisons of data at the same temperature T are made.

In order to test this heating model, we compare the observed values of  $C_{\text{max}}$  with the calculated values of  $\Delta V_h$  given by Eq. (2). We choose T = 1.37 K. Since the magnitude of  $S_n$  is not known explicitly, calculations of  $\Delta V_h$  were carried out for  $\gamma = 10^{-5}$ ,  $10^{-4}$ ,  $10^{-3}$ ,  $10^{-2}$ ,  $10^{-1}$ , and 1 where  $\gamma = S_n/S$ .  $R_s$  and dI/dV are obtained from the *I*-*V* characteristic curve in Fig. 3. The calculated curves of  $\Delta V_h$  as a function of *I* have a very similar shape for all  $\gamma$ 's except for  $\gamma = 1$ . In the last case  $\Delta V_h$  is almost constant at  $I > 1.5 \ \mu$ A. The typical results (the curves of  $-\Delta V_h$ 



FIG. 4.  $-\Delta V_h$  vs *I* expected from the heating model.  $\gamma$  is defined as  $S_n/S$  where S is the junction area and  $S_n$  is the area in the normal state. The uppermost curve is the observed  $C_{\text{max}}$  at points A-F (see Fig. 3) normalized to the value at point F.

Ι (μA)

for  $\gamma = 10^{-4}$  and  $10^{-1}$ ) are shown in Fig. 4, where the curve of  $C_{\text{max}}$  is also given for comparison.

As seen in the figure  $-\Delta V_h$  in the heating model has the minimum at around  $I = 5 \ \mu A$  and then increases as I increases. On the other hand, the observed  $C_{\text{max}}$  decreases linearly as I increases. From this result, it is difficult to attribute the origin of  $\alpha$ particle signals to a localized superconducting-normal transition.

The third possibility is a change in the I-V characteristics due to excess quasiparticles. In the semiconductor model, the current I flowing through an S-I-Stunnel junction in the thermal equilibrium is given by<sup>10</sup>

$$I = \frac{G}{e} \int_{-\infty}^{\infty} \frac{|E|}{(E^2 - \Delta_T^2)^{1/2}} \frac{|E + eV|}{[(E + eV)^2 - \Delta_T^2]^{1/2}} [f(E) - f(E + eV)] \Theta(|E| - \Delta_T) \Theta(|E + eV| - \Delta_T) dE \quad , \tag{3}$$

where G is the normal-state conductance, V is the applied voltage, and  $2\Delta_T$  is the energy gap in the equilibrium state at T. f(E) is the Fermi-Dirac distribution function and  $\Theta$  is the step function. When the sample is irradiated by an  $\alpha$  particle, f(E) and  $\Delta_T$  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 assumptions: (a) The number of excess quasiparticles N produced by an incident  $\alpha$  particle is much smaller than that of thermally excited quasipar-

T = 1.37 K

ticles  $N_T$  at T. (b) The distribution function of N has the same form as for  $N_T$ . The above two assumptions permit us to adopt an approximate expression for the total quasiparticle distribution as f(E) $+\delta(E)$ , where  $\delta f(E)$  is the fractional change in f(E) caused by an incident particle. Thus, we get the following simple relations:

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

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

where  $2\Delta$  denotes the energy gap in the nonequilibrium state, and  $N_T$  is defined by

$$N_T = 2N(0) U \int_{\Delta_T}^{\infty} \frac{E}{(E^2 - \Delta_T^2)^{1/2}} f(E) dE \quad . \tag{6}$$

In Eq. (6), U is the volume of the junction in question, and N(0) is the density of states at the Fermi level for electrons of one spin orientation. The numerical values of  $N_T$  computed are listed in Table I. From Eqs. (4) and (5) we get

$$\delta f(E) = \begin{cases} \frac{N}{N_T} f(E) & \text{for } E > \Delta \\ N & \end{cases}$$
(7)

$$\left[-\frac{N}{N_T}[1-f(E)] \text{ for } E < -\Delta \right]$$
(8)

$$I_{0} = \begin{cases} \frac{G}{e} \int_{\Delta_{T} - eV}^{-\Delta_{T}} \frac{|E|}{[E^{2} - \Delta_{T}^{2}]^{1/2}} \frac{|E + eV|}{[(E + eV)^{2} - \Delta_{T}^{2}]^{1/2}} dE & \text{for } eV > \\ 0 & \text{for } eV < 2\Delta_{T} \end{cases}$$

In the present measurements a constant current was supplied (instead of a constant voltage) and the signals were measured as changes in voltage  $\Delta V_q$ . Thus,

$$\Delta V_q = -\frac{dV}{dI} \Delta I = -\frac{N}{N_T} \frac{dV}{dI} (I - I_0) \quad . \tag{12}$$

The above equation indicates that in the excess quasiparticle model  $\Delta V_q$  does not directly reflect *I*, but rather the difference between *I* and *I*<sub>0</sub> is important. The physical meaning of *I*<sub>0</sub> is apparent from Eq. (11); i.e., *I*<sub>0</sub> is the current from the energy states under the energy gap in one layer of Sn to the states above the energy gap in another layer of Sn. Hence, *I*<sub>0</sub> is zero for  $eV < 2\Delta_T$  but when eV becomes greater than  $2\Delta_T$ , *I*<sub>0</sub> increases quite rapidly as a function of *V*. This results in the decrease in the relative contribution of excess quasiparticles, and consequently  $-\Delta V_q$  should become smaller. As for the change in  $\Delta_T$ , Owen and Scalapino<sup>2</sup> and Chang and Scalapino<sup>11</sup> found a simple equation for uniform nonequilibrium superconductors

$$\Delta/\Delta_T \simeq 1 - 2n \quad , \tag{9}$$

if the conditions  $n = N/4N(0) U\Delta_0 \ll 1$ ,  $T \ll T_c$ , and  $T \neq T_c$  are satisfied, where  $2\Delta_0$  denote the energy gap in the equilibrium state at T = 0. In the present case, the diffusion length of nonequilibrium quasiparticles during their lifetime is crudely estimated to be 50  $\mu$ m, and thus it is not unreasonable to adopt the relation given by Eq. (9). Analogous to other excitation processes like an electron-hole pair production in a semiconductor, we assume  $N \simeq Q/6\Delta_0$ , where Q is the energy loss of an  $\alpha$  particle in the junction. On this assumption  $n \simeq 8 \times 10^{-4}$  when Q = 100 keV and from Eq. (9) we can neglect the change in  $\Delta_T$  caused by the production of excess quasiparticles. Henceforth,  $\Delta$  is replaced by  $\Delta_T$ .

The above discussion concerning the fluctuation of f(E) and  $\Delta_T$  gives the approximate expression for the change in current  $\Delta I$  for a constant supplied voltage as

$$\Delta I = \frac{N}{N_T} (I - I_0) \quad , \tag{10}$$

 $> 2\Delta_T$ 

(11)

It was not possible to measure  $I_0$  experimentally. Indeed, as seen from the I-V characteristic curve of the present sample, the current flowing through the junction is not precisely expressed by I given in Eq. (2). At the very least, however, it could be possible to approximate  $(I - I_0)$  above the gap voltage as a typical value of I just below the gap voltage. The numerical values of  $-\Delta V_q$  given in Table I are thus obtained. This approximation eventually neglects the effect that  $(I - I_0)$  decreases as V increases over the gap voltage. Nevertheless, the general tendency of  $C_{\text{max}}$  agrees pretty well with  $-\Delta V_q$  evaluated by the quasiparticle model. A little more detailed discussion demonstrating the utility of this model will be given below.

First, we compare the values of  $C_{\text{max}}$  at points A, B, C, D, E, F, and G (T = 1.37 K for all points). The relative values of  $C_{\text{max}}$  for the first four points gradually increase as V decreases. At point E, where V is just below the gap voltage,  $C_{\max}$  steps up and at points F and G,  $C_{\max}$  has the value several times larger than those at points A -D. This relative change in  $C_{\max}$  is similar to the change in  $-\Delta V_q$  and evidently the contribution of  $I_0$  is reflected on the observed  $C_{\max}$ .

Second, a similar explanation can be applied when comparisons of  $C_{\text{max}}$  for points B, I, and K are made. Measurements at these points were carried out at different temperatures 1.37, 1.64, and 1.88 K. Since the bias 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 bias potential V is comparable to or smaller than the energy gap  $2\Delta_{1.64} \simeq 1.12$  mV or  $2\Delta_{1.88} \simeq 1.10$  mV; i.e., the contribution of  $I_0$  is small enough or zero in these cases. The observed values of  $C_{\text{max}}$  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,  $C_{\text{max}}$  becomes greater as T increases. It should be noted that the value of  $-\Delta V_q$  at point B is greater than those at points I and K. This is certainly due to overestimation of  $-\Delta V_q$  at B; i.e.,  $(I - I_0)$  for point B (and also for point A) should be smaller than the used value.

Third, we compare points F(T = 1.37 K) and M(T = 1.88 K) where  $I_0 = 0$  for both points. Our measurements gave that  $C_{\text{max}}$  for F is greater than that for M. From Eq. (12), this is attributed to the dif-

ferent values of  $N_T$ , of which the numerical values are also listed in Table I.

As the last example, we compare points D and Lwhere T = 1.37 and 1.88 K, respectively, and  $I_0 = 0$ for both points. Since  $N_T$  at point D is evidently smaller than that at L, it may be expected (as in the above case) that  $C_{max}$  at D is greater than that at L. However, our observations gave an opposite result, i.e.,  $C_{max}$  at D is smaller than at L. This opposition can be attributed to the fact that dV/dI in Eq. (12) for D is much smaller than that for L.

In conclusion, we have attempted qualitative explanation of the electrical resistance produced in an S-I-S tunnel junction under irradiations of  $\alpha$  particles. As revealed in the above discussion, the excess quasiparticle model is effective for qualitative understandings of the nonequilibrium state in the junction. This leads us to the conclusion that excess quasiparticles are essential for the impulsive change in I-Vcharacteristics. More refined measurements and analysis of the pulse shape are in progress.

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