Double peak phenomenon in superconducting tunnel junction x-ray detectors

M. Ohkubo, b) D. Fukuda, b) I. Sakamoto, and N. Hayashi
Electrotechnical Laboratory, Umezono, Tsukuba, Ibaraki 305-8568, Japan

J. Martin, F. Panteleit, and R. P. Huebner
Physikalisches Institut, Lehrstuhl Experimentalphysik II, Universität Tübingen, Auf der Morgenstelle 14, 72076 Tübingen, Germany

(Received 3 November 1997; accepted for publication 30 September 1998)

It has been normally observed that superconducting tunnel junction x-ray detectors produce a double peak originating from different absorption events in two electrodes. The double peak is associated with the dynamics of quasiparticles created by x-ray absorption events. In this study we have found that the double peak phenomenon depends on both bias voltage and magnetic history. On the events in the base and counterelectrodes the detector exhibits dissimilar signal–height versus bias-voltage characteristics, which suffer a large change when a small number of Abrikosov vortices corresponding to even a terrestrial magnetic field are frozen during cooling. These observations are explained by a multiple quasiparticle tunneling model with quasiparticle trapping in the vortices.

© 1999 American Institute of Physics. [S0021-8979(99)09301-9]

I. INTRODUCTION

In superconducting tunnel junction x-ray detectors, which are normally kept at temperatures less than one-tenth of a superconducting transition temperature to suppress thermal excitations, quasiparticles (qp) created by an x-ray absorption event are detected via the quantum tunneling effect through an insulating barrier between two electrodes. Since the superconducting energy gaps ($2\Delta$) are roughly three orders magnitude smaller than semiconductor band gaps, the created qp number is extremely large, so that a high energy resolution is expected for a statistical reason. In fact, it has often been reported that the energy resolutions of the superconducting detectors exceed those of the semiconductor-based detectors, which are about 100 eV for the 5.89 keV x-rays. For example, energy resolutions of 27–29 eV have been reached in Al/AlOₓ/Al or Nb/Al/AlOₓ/Al/Nb junctions, although these values are significantly worse than theoretically expected resolutions of 1.5–4.4 eV.

In addition to the unfulfilled energy resolution prediction, one of the problems in the tunnel junction detectors is that the two electrodes respond differently to monochromatic x-rays, even when the longitudinal junction structure is symmetric. This results in a double peak in energy spectra. The double peak causes a complication in applications like x-ray fluorescence analysis. It is possible to extract the signals due to the only x-ray events in either electrode by employing a qp trapping layer with a $\Delta$ value smaller than that of the absorber electrode, or time discrimination of output pulses. Anyhow, those special treatments are necessary to overcome the double peak problem. On the other hand, since the double peak is indubitably associated with dissimilarities between the counter and base electrodes in such qp dynamic aspects as qp tunneling, qp loss, qp trapping, and phonon decay or escape, there is a chance to derive some of these. However, the actual signal creation is complicated. For example, it has been observed that which electrode produces a higher signal depends on such measurement conditions as bias point. Up to now, there seems to be not enough understanding of the double peak phenomenon.

In this study, we have observed in polycrystalline Nb/Al junctions that the signal heights due to a $^{55}$Fe x-ray source depend on both bias voltage and magnetic history. The x-ray events in the base electrode (base events) intrinsically produced higher signals than those in the counterelectrode (counterevents) in a wide bias–voltage range, when no magnetic flux quanta were frozen during cooling. However, when the vortices corresponding to a terrestrial magnetic field were frozen, the signal height order of the base and counterevents changed with bias voltage. The bias–voltage dependences of the charge outputs and the effects of the frozen vortices are explained by a multiple qp tunneling model with qp trapping by the Abrikosov vortices.

II. EXPERIMENT

Square junctions with a size of 200×200μm² were fabricated by dc-sputtering of Nb and Al targets with intermediate oxidization of the Al surface and by subsequent photolithographic patterning using reactive ion etching. The longitudinal junction structure is practically symmetric: Al(5) /Nb(200) /Al(20) /AlOₓ /Al(20) /Nb(200) /ɛ-Al₂O₃ in nm. The junction parameters are as follows: a normal resistivity of 13.6 $\mu$Ω cm², a gap parameter ($\Delta$) of 1.09 meV at 0 K, which was determined by fitting a Bardeen–Cooper–Schrieffer (BCS) formula to the temperature dependence of subgap qp current at 1.7 mV, and a quality factor (a dynamic resistance at 0.5 mV divided by a normal resistance) of 1500 at 0.42 K. The $\Delta(0)$ value of 1.09 meV is consistent with a $\Delta$ voltage for which a rapid current increase occurs in the current–voltage ($I$–$V$) curve at 0.42 K. The quality factor is

---

a) Electronic mail: ohkubo@etl.go.jp
b) On leave from Tokyo University.
far worse than our best value of $3 \times 10^6$. However, in such high quality junctions Fiske steps are pronounced, so that stable biasing was impossible in our present instrument setup. We plan to improve the instrumentation.

The junction response to a $^{55}$Fe radiation source, which emits the x rays of 5.89 keV (~88%) and 6.49 keV (~12%), was measured at 0.36–0.40 K under a parallel magnetic field of 60–80 G to suppress the dc Josephson current. Before the x-ray measurements, cooling the junction was performed with or without a magnetic shield having an attenuation factor of more than 1000 to study the effects of a terrestrial magnetic field. It has been confirmed that all experimental data are reproducible without meaningful variations. The junction signals were fed to a charge sensitive preamplifier and a shaping amplifier, and then the spectra were stored in a multichannel analyzer. Such parameters as signal heights, rise times, and fall times of the preamplifier output pulses were also analyzed by a digital oscilloscope.

III. EXPERIMENTAL RESULTS

First, the bias–voltage dependence of x-ray spectra for the $^{55}$Fe radiation source was measured after cooling without the magnetic shield, and is shown in Fig. 1 with the signal heights as a function of bias voltage in the inset. The peak in channel 930, of which the full width at half maximum (FWHM) value is 300 eV, is due to a pulser. At 0.52 mV the single x-ray peak with a FWHM of 330 eV is observed only. The x-ray peak is primarily due to the 5.89 keV x rays, and the 6.49 keV x-ray peak is buried in the tail. The x-ray peak has a perfect Gaussian shape except for the shoulder and tail, reflecting that the intrinsic resolution of 140 eV is considerably better than the electronic noise. On the other hand, the double peaks appear at other bias voltages.

The assignment of the two curves in the inset of Fig. 1 to the base and counterevents was performed by employing low temperature scanning electron microscopy (LTSEM).6 The base and counterelectrodes of another junction on the same wafer were separately stimulated by short electron beam pulses, and the decay times of the junction response were measured at a bias voltage of 1.3 mV and at 2.1 K. The details of LTSEM can be found in other publications.7,8 The decay time constants were 0.21 and 0.15 μs for the base and counterelectrodes, respectively. On the other hand, in the scatter plots of the signal heights against the charge sensitive preamplifier rise times, the x-ray events were classified into two groups having mean rise times (~20%–80%) of 0.15 and 0.11 μs.6 When we take a qp loss time and a tunneling time, which are described in Sec. IV, a tunneling probability should be as small as less than 35% at this high bias voltage. Therefore, it is reasonable that both the LTSEM decay times and the preamplifier rise times distinguishably reflect the qp loss times in the base and counterelectrodes. Accordingly, the long and short risetime groups in the scatter plots are assigned to the base and counterevents, respectively. A comparison of the absolute values between the x-ray and LTSEM experiments is inappropriate because of different experimental condition. A signal height measurement by LTSEM was also consistent with the x-ray results. Consequently, the two curves in the inset of Fig. 1 are assigned to the base events for the closed circles and the counterevents for the open circles.

As can be seen in the inset of Fig. 1, the counterevents produce higher signals than the base events in a bias voltage range less than 0.52 mV, and vice versa above that bias point. The crossover of the two curves appears at 0.52 mV, and there the double peak emerges into the single peak. The
signal height order changing with bias voltage is compatible with the other observation. The thin and thick solid lines are based on a multiple quasiparticle tunneling model. The higher base outputs are consistent with the longer qp length. The proximity layer thickness in the Nb absorber and the Al trapping layer is 1.09 meV, which is increased from the bulk value by the proximity effect. In the Al layer the thermal qp density is given by $n_{th} = 4N(0)\Delta(\pi k_B T/2\Delta)^{0.5}\exp(-\Delta/k_B T)$, where $N(0)$ is the single spin density of states at the Fermi energy. The $\Delta$ value of the Nb absorber is extrapolated at 1.55 meV, which is equal to the bulk value, from the Al thickness dependence of $\Delta$ in our experiment. The measured $\Delta$ value of the Al trapping layer is 1.09 meV, which is increased from the bulk value by the proximity effect. In the Al layer the $\Delta$ value should be constant because of a long coherence length. The proximity layer thickness in the Nb absorber adjacent to the Al trapping layer is on the order of the Nb coherence length. The effective temperatures corresponding to the 5.89 keV x-ray absorption are between 1 K for a uniform qp distribution throughout the electrode and 2.5 K for a spread within a diffusion length of 10 $\mu$m. There was no meaningful variation of the tunneling times obtained from the $I$–$V$ curves in this temperature range. We took the $I$–$V$

![FIG. 2. Comparison of charge–output vs bias–voltage characteristics between after cooling with and without a magnetic shield. The charge outputs were evaluated by correcting the signal heights. The closed and open circles denote the experimental data for the base and counterevents, respectively. The thin and thick solid lines are based on a multiple quasiparticle tunneling model.](image)

**IV. DISCUSSION**

**A. Multiple quasiparticle tunneling model**

We adopt a multiple qp tunneling model with the Gray effect to explain the bias–voltage dependences of the charge outputs. The qp tunneling has been often treated as a qp loss process, providing that once qps tunnel through a tunnel barrier they promptly disappear from the vicinity of the barrier. However, this is invalid for most cases. In this study, since our junction has the symmetric structure with the Al qp trapping layers, the tunneling is not a loss process because of the back tunneling. In the multiple tunneling model, the qps can tunnel forth and back until they disappear by recombination into Cooper pairs or other loss processes.

The detected charge $Q_c$ for an x-ray event in the counterelectrode is expressed by

$$Q_c = Q_0 \frac{P_c(1 + P_b)}{1 - P_c P_b},$$

where $Q_0$ is the initial qp charge created by an x-ray absorption, and $P_c$ and $P_b$ are the probabilities of the qp tunneling from the counterelectrode and from the base one to the other, respectively. We omit qp creation by phonon coupling between the two electrodes. When an x-ray event occurs in the base electrode, the subscripts $c$ and $b$ should be exchanged. The tunneling probability is given by

$$P_c = \frac{\tau_{tun,c}}{\tau_{tun,c} + \tau_{r,c}},$$

where $\tau_{tun,c}$ is the electrical tunneling time from the counterelectrode to the base one, and $\tau_{r,c}$ is the qp loss time in the counterelectrode. For the base electrode the subscript $c$ should be changed to $b$.

The bias dependence of the tunneling times can be derived from an $I$–$V$ curve, that is a thermal qp current given by $I_{th} = 2eN_{th}/\tau_{tun}$, where $N_{th}$ is the thermal qp number in an electrode. We assume that the $\tau_{tun,c}$ and $\tau_{tun,b}$ are equal, and that the $N_{th}$ is the sum of the qp numbers in the Nb absorber and the Al trapping layer. The thermal qp density is given by $n_{th} = 4N(0)\Delta(\pi k_B T/2\Delta)^{0.5}\exp(-\Delta/k_B T)$, where $N(0)$ is the single spin density of states at the Fermi energy. The $\Delta$ value of the Nb absorber is extrapolated at 1.55 meV, which is equal to the bulk value, from the Al thickness dependence of $\Delta$ in our experiment. The measured $\Delta$ value of the Al trapping layer is 1.09 meV, which is increased from the bulk value by the proximity effect. In the Al layer the $\Delta$ value should be constant because of a long coherence length. The proximity layer thickness in the Nb absorber adjacent to the Al trapping layer is on the order of the Nb coherence length. The effective temperatures corresponding to the 5.89 keV x-ray absorption are between 1 K for a uniform qp distribution throughout the electrode and 2.5 K for a spread within a diffusion length of 10 $\mu$m. There was no meaningful variation of the tunneling times obtained from the $I$–$V$ curves in this temperature range. We took the $I$–$V$...
curve at 1.88 K. At this high temperature the qp loss is dominated by the fast thermal recombination so that the effects of local qp trapping or other loss processes can be neglected. As a result, the qp number in an electrode is estimated at $10^8$. The tunneling time versus bias–voltage characteristic is shown in Fig. 3 with the $I–V$ curve in the inset.

We calculate the charge outputs as a function of bias voltage by Eqs. (1) and (2) with the data of Fig. 3. The thin and thick solid lines in Fig. 2 denote the calculation curves for $\tau_{r,c} = 0.18 \mu s$ and $\tau_{r,c} = 0.4 \mu s$ in the cooling without the magnetic shield, and $\tau_{r,c} = 0.4 \mu s$ and $\tau_{r,c} = 0.18 \mu s$ with the shield. The experimental data are more peaked near 0.5 mV, and lower above 1.0 mV than the calculation curves. In addition, the experimental base curves are closer to the counter ones below 0.5 mV than the model calculations. Some of the possibilities of these discrepancies are unequal tunneling times, a qp nonequilibrium distribution, or a phonon coupling. Nevertheless, the agreement with the experimental data is reasonable.

It is considered that the properties after cooling with the shield are intrinsic without the effects of the Abrikosov vortices. The intrinsic qp loss times in the base and counterelectrodes are different. They may be caused by microstructural dissimilarity between the counter- and base electrodes or a possible qp trapping layer on the counter surface. The electrode microstructure can affect effective qp lifetimes. For example, it has been pointed out that the grain boundaries of polycrystalline Nb films may cause fast phonon decay and thus shorten the effective qp lifetime. The surface trapping layer formation during the fabrication seems to have taken place, since in our junctions it has generally been observed that counterevents produce smaller signals than base events. It is expected that the thin Al cap layer on the counterelectrode, which is likely oxidized in the air, prevents oxidization or contamination of the counter Nb surface. In fact, the countersignal of the present junction is much larger than that of a junction with no Al cap layer. Nevertheless, it is possible that the contamination prevention was not perfect.

**B. Quasiparticle trapping by Abrikosov vortices**

When the junction is cooled in a terrestrial magnetic field of 0.46 G with a dip of 49°, which corresponds to a vortex density of $0.02 \Phi_0 / \mu m^2$ in the vertical direction, several hundred vortices can be frozen in the junction.

It is known that the vortices trap qps, and reduce charge outputs. The qp loss time including the qp trapping by the vortices may be given by

$$\tau_{r,c}^{-1} = \tau_{r,c}^{-1} + \tau_{AV}^{-1},$$

where $\tau_{r,c}$ is the qp loss time with no vortices, and $\tau_{AV}$ is the qp trapping loss time inside the vortices. The $\tau_{AV}$ values are calculated from the above qp loss times after cooling with and without the shield. The calculated $\tau_{AV}$ values are 0.33 $\mu s$ for the base electrode and 0.36 $\mu s$ for the counterelectrode. When we assume that the qp diffusion length is much longer than the distance between the vortices, the trapping qp loss time is given by

$$\tau_{AV}^{-1} = \frac{1.82 n \pi R_{eff}^2}{\tau_0},$$

where $n$ is the vortex density, $R_{eff}$ is the effective vortex radius, and $\tau_0 (= 100$ ps for Nb) is the material dependent characteristic time appearing in Refs. 12 and 14. The effective vortex radius is proportional to the coherence length $\xi$ and expressed by $R_{eff} = C \xi$ with the material independent coefficient $C$. Theoretical estimations of the $C$ coefficient are rather scattered between 2.7 and 5. In the present case the $\xi$ value, which is defined as $\xi = (l_0 \xi_0 / 3)^{0.5}$ with the electron mean free path $l$ and the clean limit coherence length $\xi_0$, is equal to 11 nm for $l = 10$ nm that was obtained from a residual resistivity of $3.1 \mu \Omega cm$ of our Nb films. The $R_{eff}$ value is then expected to be between 31 and 57 nm. By Eq. (4), the frozen vortex densities are calculated: 0.016–0.06 $\Phi_0 / \mu m^2$ for the base electrode and 0.014–0.05 $\Phi_0 / \mu m^2$ for the counterelectrode. The vortex densities in both electrodes seem to be the same, although the vortex line pattern is unknown.

The terrestrial field of 0.02 $\Phi_0 / \mu m^2$ is within the above vortex density ranges. Moreover, our results are consistent with an LTSEM experiment on the effects of the frozen vortices. We therefore conclude that the vortices frozen in the terrestrial field cause the reduction of the charge outputs and the qp loss times. The charge output reduction is more pronounced in the base events, because the base electrode has the longer qp loss time. In the present calculations, an inversion of the charge output order with bias voltage never happens. However, the experimental data of the intrinsic base charge outputs are closer to those of the counter ones below 0.4 mV than the calculations. Therefore, the charge output reduction results in the inversion of the signal heights.
below 0.52 mV as observed in the inset of Fig. 1. This is the reason for the crossover, which appears in the raw x-ray spectra, after cooling without the shield.

V. CONCLUSIONS

We have reasonably revealed that the double peak phenomenon is induced by the qp loss time dissimilarity between the counter- and base electrodes. In the present junction, the base electrode has the longer qp loss time than the counterelectrode. Correspondingly, the intrinsic charge outputs of the base events are higher than those of the counterevents at any bias voltages, when the junction is cooled in the magnetic shield. On the other hand, after cooling in the terrestrial magnetic field, the Abrikosov vortices are frozen in the junction. The vortices cause the reduction of the qp loss times and thus the charge outputs because of the qp trapping in the vortex cores. The effects of the vortex qp trapping are more pronounced in the base electrode, since the base electrode has the longer qp loss time. This results in the crossover of the raw curves of the signal–height versus bias–voltage characteristics.

Our results show that even if a density of the frozen Abrikosov vortices is as small as that corresponding to a terrestrial magnetic field, the change in the signal creation is significantly large. Further experiments at different vortex densities are necessary.

ACKNOWLEDGMENTS

The authors thank H. Pressler, M. Ukibe, F. Hirayama, M. Koyanagi, A. Shoji, and N. Kobayashi of ETL, M. Katagiri and M. Kishimoto of JAERI, M. Nakazawa and H. Takahashi of Tokyo University, M. Kurakado of Nippon Steel Corporation, and the members of the superconducting electronics group at ETL for fruitful discussion and experimental assistance.

5 S. E. Labov (private communication).