

Comparison of Intrinsic Josephson and SIS Tunneling Spectroscopy of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$

L. Ozyuzer, C. Kurter, J. F. Zasadzinski, K. E. Gray, D. G. Hinks, and N. Miyakawa

Abstract—Tunneling spectroscopy measurements are reported on optimally-doped and overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystals. A novel point contact method is used to obtain superconductor-insulator-normal metal (SIN) and SIS break junctions as well as intrinsic Josephson junctions (IJJ) from nanoscale crystals. Three junction types are obtained on the same crystal to compare the quasiparticle peaks and higher bias dip/hump structures which have also been found in other surface probes such as scanning tunneling spectroscopy and angle-resolved photoemission spectroscopy. However, our IJJ quasiparticle spectra consistently reveal very sharp conductance peaks and no higher bias dip structures. The IJJ conductance peak voltage divided by the number of junctions in the stack consistently leads to a significant underestimate of Δ when compared to the single junction values. The comparison of the three methods suggests that the markedly different characteristics of IJJ are a consequence of nonequilibrium effects and are not intrinsic quasiparticle features.

Index Terms—High- T_c superconductors, intrinsic Josephson junctions, tunneling spectroscopy.

I. INTRODUCTION

WITH conventional superconductors, it is relatively easy to fabricate stable superconductor-insulator-normal metal (SIN) and superconductor-insulator-superconductor (SIS) planar type junctions and the tunneling spectra from these have provided a direct measure of the temperature dependent energy gap, $\Delta(T)$, and the electron-phonon interaction of strong-coupled BCS theory [1]. Yet for high temperature superconductors (HTS) no analogous planar junction technology exists. Consequently, our understanding of the quasiparticle density of states (DOS) is obtained from mechanical junction methods. In $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi2212) for example, SIN point contact tunneling (PCT) [2], [3], SIS break junctions [2]–[5] and Scanning Tunneling Microscopy/Spectroscopy (STM/S) vacuum junctions [4], [6], [7] have all been achieved and the results are consistent. The characteristic features of

the Bi2212 junctions are quasiparticle peaks at the gap voltage that are both larger and broader than expected from a d -wave DOS as well as reproducible dip and hump structures at higher voltages which appear to be related to strong coupling effects [4], [7], [8]. The magnitude of the energy gap is consistently around 35–40 meV for optimally doped samples of Bi2212 and decreases (increases) with overdoping (underdoping) [3], [5], [8]. Another type of tunneling configuration has emerged which is unique to the layered, HTS cuprates, namely the intrinsic Josephson junctions (IJJ) between adjacent sets of Cu-O layers (e.g. the bi-layers of Bi2212) within the bulk crystal [9]–[12]. Various techniques have been developed so that stacks with only a few junctions in series can be studied. The near perfect arrangement of such IJJ suggests that they will naturally lead to the intrinsic quasiparticle DOS of HTS, however, we will demonstrate here that the IJJ exhibit markedly different properties compared to the single-junction (SJ) methods and that the quasiparticle spectra of IJJ appear to be exhibiting nonequilibrium effects.

Interest has increased in IJJ because fabrication methods such as the patterning of small area mesas have led to a manageable number of such junctions in series [10]–[12]. These are considered as c -axis tunnel junctions between sets of superconducting CuO_2 planes. In the case of Bi2212, the CuO_2 bi-layers are only 0.3 nm thick and are separated by layers of Sr-O and Bi-O, which are 1.2 nm thick and act as insulating or semiconducting spacers. The measurements on small area mesas have generally been focused on the Josephson currents which can be studied at low bias voltages and the individual switching of each junction from the Josephson branch to the quasiparticle branch can be observed. Surprisingly, this same pattern has also been observed at times using break junction methods which are intended to produce only a single SIS junction between pieces of the HTS crystal. Presumably, in the case of Bi2212, nanoscale sized crystallites can result in the process of breaking the crystal and in some cases the I-V characteristics of these crystallites can be measured.

The temperature and magnetic field evolution of current-voltage and conductance-voltage characteristics of IJJ have been intensively investigated. The study of the quasiparticle branch requires higher voltages and heating effects have been a problem. Various methods have been attempted to minimize such heating effects including intercalation and reduced junction area, although the latter still leads to the same power per unit area. A short current pulse method has been used to minimize self-heating effects [12].

Conventionally, the surface of bulk crystals have been patterned for different sizes and chemically etched or ion milled to

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L. Ozyuzer is with the Department of Physics, Izmir Institute of Technology, Gulbahce Campus, Urla, TR-35430 Izmir, Turkey (e-mail: ozyuzer@iyte.edu.tr).

C. Kurter is with the Izmir Institute of Technology, Izmir, Turkey.

J. F. Zasadzinski is with the Physics Division, Illinois Institute of Technology, Chicago, IL 60616 USA.

K. E. Gray and D. G. Hinks are with the Materials Science Division, Argonne National Laboratory, Argonne, IL USA.

N. Miyakawa is with the Tokyo University of Science, Suwa, Japan.

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obtain stack of junctions. Current and voltage leads have been connected using a passivation technique. This method allows a stable junction geometry so that temperature dependent measurements can be done very easily. This is the most important advantage compared to PCT, break junctions and STM. The disadvantage is that the chemical process steps may alter the intrinsic physical properties of the superconductor, especially the topmost Cu-O layer.

In this paper, we will present a tunneling study of optimally doped and slightly overdoped Bi2212 as obtained from both SJ (SIN and SIS) and IJJ junction types. All three junction geometries are obtained in-situ from a bare crystal at 4.2 K using a point contact method with a blunt Au tip. We will compare results of the three distinct techniques and give an analysis of the quasiparticle tunneling spectra. It is argued that the strong dip/hump features found reproducibly in the SJ spectra are intrinsic and the absence of such features in the IJJ spectra are a consequence of heating or nonequilibrium processes such as quasiparticle injection. Finally, the temperature dependence of SIS and IJJ tunneling conductances are compared and we will argue that this measurement also points toward the SIS junctions as revealing the intrinsic quasiparticle DOS.

II. EXPERIMENT

Single crystals of Bi2212 were grown by a floating zone technique. As grown crystals are slightly overdoped and they are then annealed in Ar gas flow to obtain optimally doped T_c values with an onset at 95 K. Overdoping of samples has been achieved by annealing as grown crystals in O_2 gas flow. The crystals of the present study have a $T_c = 82$ K overdoped, and $T_c = 92$ K near optimally doped with less than 2 K transition width found by ac susceptibility measurements. Tunneling measurements were done using a continuous flow cryostat with a tunneling apparatus described in [13]. Most of the tunneling data are taken at $T = 4.2$ K. Higher temperatures are obtained by adjusting the He gas flow and vaporizer temperature. Samples are cleaved with a scotch tape to obtain clean surface of a-b plane and immediately mounted to the system where the counter electrode, Au tip, approaches along the c-axis of the crystal.

A novel method is used to form SIS break junctions and a detailed description can be found elsewhere [2]. First, SIN junctions are formed when the Au tip touches the nonconducting Bi-O/Sr-O surface bilayer. The SIN junctions provide an independent method for obtaining the superconducting gap. These conductances exhibit quasiparticle peaks at $eV \sim \Delta$ and clearly show the dip and hump features above Δ , most strongly observed in the occupied states of the DOS. With increased pressure the tip breaks through the insulating surface layer forming an ohmic contact to the crystal and this contact forms a strong mechanical bond as well. Further manipulation of the tip leads to a micro cleaving of Bi2212 where a small crystallite is stuck to the Au tip and an SIS junction can be formed between this crystallite and the underlying crystal. The convolution of the experimental SIN data with itself is then compared to the SIS conductances obtained from break junctions. In all cases, the

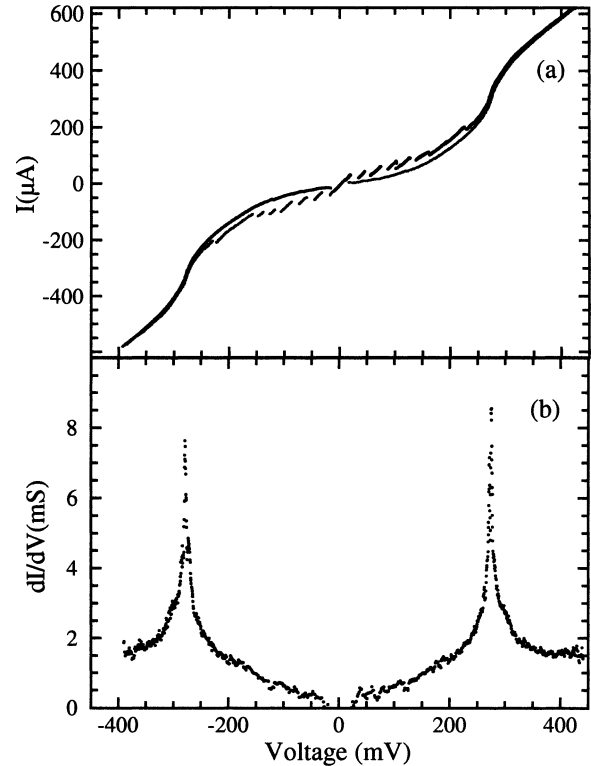


Fig. 1. (a) I-V and (b) dI/dV -V characteristics of IJJ made by point contact method on an overdoped Bi2212 crystal at 4.2 K. Josephson branches have been removed for clarity in dI/dV -V spectra.

SIS conductances show quasiparticle peaks at 2Δ that are consistent with the Δ value obtained from the SIN junction. This rules out the possibility of our SIS conductances being the result of multiple junctions in series. Additionally, the SIS conductances also show a symmetric dip and weak hump feature as well as the presence of a Josephson current. The magnitude of this current generally decreases as the junction resistance increases. Sometimes breaking the sample with Au tip resulted multiple junctions which exhibited the typical IJJ characteristics found on fabricated Bi2212 mesas. The I-V curve show multiple Josephson branches with hysteretic switching properties.

III. RESULTS

Fig. 1(a) shows the I-V characteristics of an IJJ made by the point contact method on the overdoped Bi2212 crystal with $T_c = 82$ K, on which SIN and SIS junctions were also obtained. Multiple Josephson branches and hysteretic behavior are observed. The gray curve represents bias sweeps from positive to negative voltage, and the black line is the reverse. Since it is assumed that the number of branches in the I-V curve is the number of junctions, n , the dynamical conductance peak corresponds to n times Δ_{IJJ} . We will call it Δ_{IJJ} because of the substantially smaller value compared to Δ which is obtained from SIN and SIS junctions. In Fig. 1(a), 7 branches can be counted. In Fig. 1(b), is shown the tunneling conductance of the same junction. Here only the nonhysteretic branches of the I-V curve are shown for clarity. The spectra show very narrow conductance peaks, V_p , at ± 274 mV without any features beyond

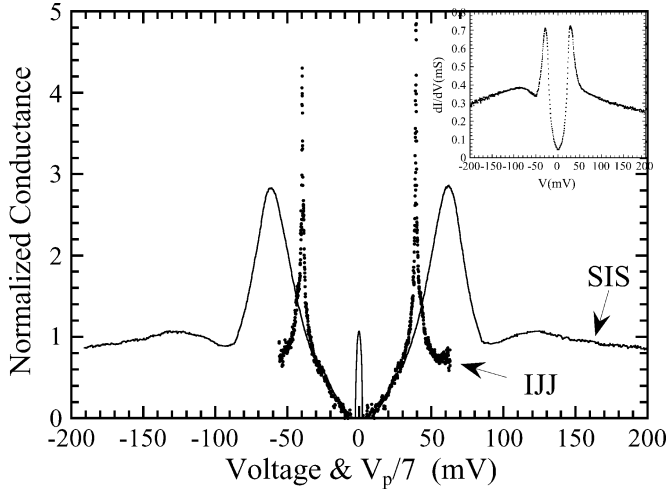


Fig. 2. Comparison of normalized conductance of SIS and IJJ on overdoped Bi2212 at 4.2 K. Inset shows tunneling conductance of a representative SIN spectra. All three spectra are obtained in a single run.

$\pm n\Delta_{IJJ}$. Below the conductance peaks, the tunneling conductance linearly increases up to ± 200 mV, which is similar to observation of [14].

The inset of Fig. 2 shows SIN tunneling conductance characteristics obtained from the same crystal. The main panel of Fig. 2 shows the tunneling conductance of an SIS break junction at 4.2 K compared with the IJJ data. The voltage bias axis of IJJ data is divided by 7 because of 7 branches, so both junction types can be shown on the same figure. The conductance peaks of SIS junction correspond to $\pm 2\Delta$, ± 62 meV. Furthermore, Josephson current peak at zero bias indicates the superconducting nature of the observed energy gap magnitude. An important difference between the two quasiparticle curves is the location and shape of the dynamical conductance peaks. Convolution of the SIN data to generate an SIS conductance leads to a shape consistent with the measured SIS data. IJJ displays sharp peaks at ± 39 mV which corresponds to $2\Delta_{IJJ}$. Thus the IJJ leads to an underestimate of Δ and $\Delta_{IJJ} \neq \Delta$. Excessive heat developed in the IJJ stack is the likely cause of the lower gap value. This leads to an effective temperature of the stack which can be significantly larger than the bath temperature. However, the ratio of Δ_{IJJ}/Δ is around 0.65 which is close to the value of 0.6 found in the theory of Owen and Scalapino [15] that considers the nonequilibrium electron distribution caused by excess quasiparticle injection. Thus such injection effects might be playing a role in the IJJ quasiparticle characteristics. Since the barrier strength is weak between CuO_2 planes, overinjection of quasiparticles may cause a nonequilibrium state [16]. Interestingly, near zero bias both junctions show a similar shape indicative of an anisotropic gap. The IJJ junction characteristics deviate for larger bias, again suggesting the development of a nonequilibrium state.

Another striking difference shown in Fig. 2 involves the higher bias dip/hump features which are completely absent in all of the IJJ studied. Since the dip/hump features are reproducibly observed in the SIN and SIS junctions as well as in STM/S spectra, they must be considered to be intrinsic properties of the electron DOS in the superconducting state.

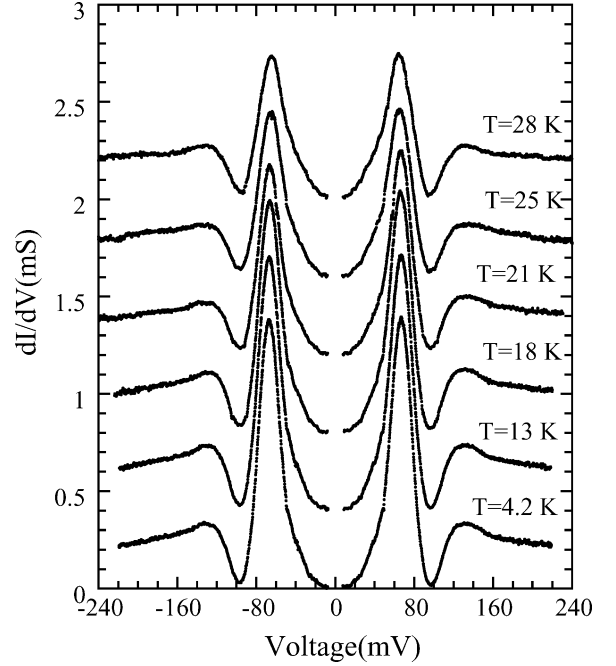


Fig. 3. Temperature dependence of tunneling conductance for a SIS junction. Each spectra is shifted 0.4 mS from each other and Josephson current peaks are deleted for clarity.

Their absence in the IJJ spectra is consistent with the loss of superconductivity in the IJJ at high voltages due to a combination of heating and injection effects [17]. Recently, the local temperature of the IJJ stacks have been obtained using a thin film thermocouple which is evaporated top of the mesa [18]. This technique shows that the local temperature of the mesa can exceed T_c for junction bias voltages near the gap voltage. This is consistent with the absence of dip and hump spectral features except when short-pulse methods are applied [12] or Bi2212 is intercalated with HgBr_2 [11].

We now turn to the temperature evolution of the gap features. Fig. 3 shows temperature evolution of tunneling spectra from an SIS break junction on optimal doped Bi2212 with $T_c = 92$ K. Josephson current peaks are deleted for clarity. In conventional, *s*-wave superconductors, the temperature dependence of the energy gap magnitude, $\Delta(T)$, is well understood, both theoretically and experimentally. $\Delta(T)$ is nearly constant up to $T \sim T_c/4$. In the vicinity of T_c , it closes to zero where it is proportional to $(1 - T/T_c)^{1/2}$. High temperature superconductors exhibit properties which might affect this T dependence. One such property is the presence of a pseudogap well above T_c , up to a characteristic temperature T^* , that has been detected by a number of experimental techniques, such as in-plane resistivity, ARPES, specific heat, and NMR. In addition to these experimental techniques, STM, break junction and planar tunnel junctions have also observed that the pseudogap exists as a depression in the DOS of Bi2212 above T_c . Irrespective of the presence of a pseudogap or its origin, the superconducting gap should exhibit a weak T dependence far below T_c .

Because of the low zero bias conductance and sharp conductance peaks in Fig. 3, we can directly find energy gap from conductance peaks which correspond to $\pm 2\Delta$. For this junction, $2\Delta = \pm 67$ meV. The data also show very strong dip

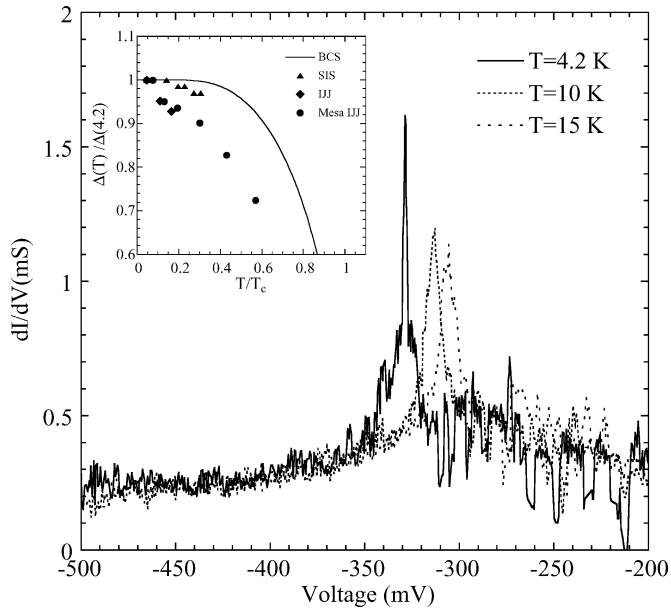


Fig. 4. Temperature dependence of tunneling conductance for IJJ. The inset shows normalized energy gap versus normalized temperature for various junction types.

features (almost zero conductance) which is consistent with a previous break junction tunneling study [8] of similar crystals with $T_c = 95$ K. Tunneling conductance peaks smear when the temperature is raised from 4.2 K to 28 K, however position of peaks hardly moves at all, consistent with a nearly constant superconducting gap as expected from conventional BCS theory. This is illustrated in inset of Fig. 4 where the SIS gap (triangles) is compared with the BCS prediction (line). The Bi2212 gap drops slightly more rapidly than the BCS prediction but this can likely be accounted for by the d-wave gap symmetry in Bi2212. Fig. 4 also shows temperature dependence of tunneling conductance of an IJJ obtained from the same Bi2212 crystal. The hysteretic branches are displayed because the bias sweep direction is a decreasing voltage. The conductance peak voltage rapidly decreases with increasing temperature. In addition, the branches also move and that is seen in previous studies of IJJ [11]. The details of strong temperature dependencies of IJJ conductance peak can be seen in inset of Fig. 4 (diamonds) along with data from $20 \times 20 \mu\text{m}$ mesa (filled circles) fabricated on a similar Bi2212 crystal. What is clearly evident is that IJJ spectra either from point contact generated stacks or patterned mesas show a consistent, rapid decrease of the conductance peak with temperature. This is contrary to the SIS gap evolution (Fig. 3) and to the BCS prediction. This is a further indication that the conductance peak in IJJ is not revealing the true quasiparticle gap.

In summary we have compared the quasiparticle spectra from single junction methods on Bi2212 with those of IJJ, including the temperature dependence. The characteristics of the IJJ spectra are very sharp conductance peaks, a reduced

superconducting gap and the absence of dip/hump features. The conductance peak also exhibits an anomalously rapid decrease with temperature. All of these features indicate the IJJ spectra are affected by nonequilibrium processes.

REFERENCES

- [1] E. L. Wolf, *Principles of Electron Tunneling Spectroscopy*. New York: Oxford Univ. Press, 1985.
- [2] L. Ozyuzer, J. F. Zasadzinski, C. Kendziora, and K. E. Gray, "Quasiparticle and Josephson tunneling of overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystals," *Phys. Rev. B*, vol. 61, pp. 3629–3640, Feb. 2000.
- [3] N. Miyakawa, J. F. Zasadzinski, L. Ozyuzer, P. Guptasarma, D. G. Hinks, C. Kendziora, and K. E. Gray, "Predominantly superconducting origin of large energy gaps in underdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ from tunneling spectroscopy," *Phys. Rev. Lett.*, vol. 83, pp. 1018–1021, Aug. 1999.
- [4] Y. DeWilde, N. Miyakawa, P. Guptasarma, M. Iavarone, L. Ozyuzer, J. F. Zasadzinski, P. Romano, D. G. Hinks, C. Kendziora, G. W. Crabtree, and K. E. Gray, "Unusual strong-coupling effects in the tunneling spectroscopy of optimally doped and overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$," *Phys. Rev. Lett.*, vol. 80, pp. 153–156, Jan. 1998.
- [5] L. Ozyuzer, J. F. Zasadzinski, K. E. Gray, C. Kendziora, and N. Miyakawa, "Absence of pseudogap in heavily overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ from tunneling spectroscopy of break junctions," *Europhys. Lett.*, vol. 58, pp. 589–595, May 2002.
- [6] C. Renner, B. Revaz, J.-Y. Genoud, K. Kadowaki, and Ø. Fischer, "Pseudogap precursor of the superconducting gap in under- and overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$," *Phys. Rev. Lett.*, vol. 80, pp. 149–152, Jan. 1998.
- [7] S. H. Pan, J. P. O'Neal, R. L. Badzey, C. Chamon, H. Ding, J. R. Engelbrecht, Z. Wang, H. Eisaki, S. Uchida, A. K. Gupta, K.-W. Ng, E. W. Hudson, K. M. Lang, and J. C. Davis, "Microscopic electronic inhomogeneity in the high- T_c superconductor $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$," *Nature*, vol. 413, pp. 282–285, Sep. 2001.
- [8] J. F. Zasadzinski, L. Ozyuzer, N. Miyakawa, K. E. Gray, D. G. Hinks, and C. Kendziora, "Correlation of tunneling spectra in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ with the resonance spin excitation," *Phys. Rev. Lett.*, vol. 87, Aug. 2001.
- [9] R. Kleiner, F. Steinmeyer, G. Kunkel, and P. Muller, "Intrinsic Josephson effects in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystals," *Phys. Rev. Lett.*, vol. 68, pp. 2394–2397, Apr. 1992.
- [10] K. Tanabe, Y. Hidaka, S. Karimoto, and M. Suzuki, "Observation of both pair and quasiparticle tunneling in intrinsic junction stacks fabricated on $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystals," *Phys. Rev. B*, vol. 53, pp. 9348–9352, Apr. 1996.
- [11] A. Yurgens, D. Winkler, T. Claeson, S. J. Hwang, and J. H. Choy, "Pseudo-gap features of intrinsic tunneling in $(\text{HgBr}_2)\text{-Bi2212}$ single crystals," *Int. J. Mod. Phys. B*, vol. 13, pp. 3758–3763, Dec. 1999.
- [12] K. Anagawa, Y. Yamada, T. Shibauchi, and M. Suzuki, "60 ns time scale short pulse interlayer tunneling spectroscopy for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$," *Appl. Phys. Lett.*, vol. 83, pp. 2381–2383, Sep. 2003.
- [13] L. Ozyuzer, J. F. Zasadzinski, and K. E. Gray, "Point contact tunnelling apparatus with temperature and magnetic field control," *Cryogen.*, vol. 38, pp. 911–915, Sept. 1998.
- [14] Y. G. Ponomarev, C. S. Khi, K. K. Uk, M. V. Sudakova, S. N. Tchesnokov, M. A. Lorenz, M. A. Hein, G. Muller, H. Piel, B. A. Aminov, A. Krapf, and W. Kraak, "Quasiparticle tunneling in the c-direction in stacks of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ S-I-S junctions and the symmetry of the superconducting order parameter," *Physica C*, vol. 315, pp. 85–90, 1999.
- [15] C. S. Owen and D. J. Scalapino, "Superconducting state under the influence of external dynamic pair breaking," *Phys. Rev. Lett.*, vol. 28, pp. 1559–1561, Jun. 1972.
- [16] K. E. Gray and H. W. Willemsen, "Inhomogeneous state of superconductors by intense tunnel injection of quasiparticles," *J. Low-Temp. Phys.*, vol. 31, pp. 911–925, 1978.
- [17] V. N. Zavaritsky, "Intrinsic tunneling or Joule heating?," *Phys. Rev. Lett.*, vol. 92, p. 259 701, 2004.
- [18] A. Yurgens, D. Winkler, T. Claeson, S. Ono, and Y. Ando, Reply to the Comment on 'Intrinsic Tunneling Spectra of $\text{Bi}_2(\text{Sr}_{2-x}\text{La}_x)\text{CuO}_6$ Auxiliary Information, Sep. 2003.