Non-equilibrium spectroscopy of high-$T_c$ superconductors

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V.M. Krasnov
Department of Physics, Stockholm University, AlbaNova University Center, SE-10691 Stockholm, Sweden

E-mail: Vladimir.Krasnov@physto.se

Abstract. In superconductors, recombination of two non-equilibrium quasiparticles into a Cooper pair results in emission of excitation that mediates superconductivity. This is the basis of the proposed new type of “non-equilibrium” spectroscopy of high $T_c$ superconductors, which may open a possibility for direct and unambiguous determination of the coupling mechanism of high $T_c$ superconductivity. In case of low $T_c$ superconductors, the feasibility of such the non-equilibrium spectroscopy was demonstrated in classical phonon generation-detection experiments almost four decades ago. Recently it was demonstrated that a similar technique can be used for high $T_c$ superconductors, using natural intrinsic Josephson junctions both for injection of non-equilibrium quasiparticles and for detection of the non-equilibrium radiation. Here I analyze theoretically non-equilibrium phenomena in intrinsic Josephson junctions. It is shown that extreme non-equilibrium state can be achieved at bias equal to integer number of the gap voltage, which can lead to laser-like emission from the stack. I argue that identification of the boson type, constituting this non-equilibrium radiation would unambiguously reveal the coupling mechanism of high $T_c$ superconductors.

1. Introduction

For more than twenty years High Temperature Superconductivity (HTSC) in cuprates remains one of the most acute unresolved problems of modern physics [1,2]. To realize how challenging the HTSC problem is, it is instructive to make comparison with a non-cuprate high temperature superconductor MgB$_2$, discovered in 2001. Despite anomalous properties, superconductivity in MgB$_2$ was understood in a very short time [3], largely based on new experimental and theoretical techniques developed for HTSC. But for cuprates the colossal puzzle formed by collected experimental data and theoretical ideas does not yet fall into a simple coherent picture.

Why explanation of HTSC takes so long time? Perhaps, a simple explanation of HTSC does not exist because a number of different phenomena coexist in those very complex materials [2]; or may be the difficulty with understanding HTSC highlights “a major intellectual crisis in the quantum theory of solids” [4]. Or may be the main obstacle is caused by immense difficulty in obtaining unambiguous information about electronic structure of HTSC: d-wave symmetry of the order parameter requires angular resolved spectroscopy [4]. The situation is further complicated by ill-defined quasiparticles [1,4]; possible inherent inhomogeneity [2]; rapid chemical deterioration of surfaces and extremely short coherence length of HTSC, which may preclude determination of bulk electronic properties in
surface sensitive experiments (e.g. photoemission spectroscopy of YBa$_2$Cu$_3$O$_7$ is overshadowed by the surface state [4]).

Furthermore, the necessity to “decipher” HTSC data may cause a methodological problem: because it shifts the roles of theory and experiment. Usually experiment has an authority to disprove theory (I. Giaever in his Nobel lecture considered that as a “dream” of an experimentalist). However, since HTSC data often require theoretical interpretation, theory comes in position to “validate” experiment. This may lead to unproductive “locking” of experiment to theory. Such situation is not new, but occurs often at crossover points when experimental data break out of the conventional pattern. Recall the aphorism of Niels Bohr made at the early stage of quantum mechanics [5]: “if there is a finite amount of experiments and an infinite amount of theories, then there is an infinite amount of theories that agree with a finite amount of experiments”.

Of course the point is not in collecting an infinite amount of experimental data, but to develop new unambiguous experimental techniques for HTSC, which would put experiment back in position to “cull the vast herd of possible explanations” [1]. Such experiments are few in HTSC but they do exist. One of them was observation of flux quantization, which unambiguously proved that both in low and high $T_c$ superconductors the condensate consists of paired electrons [2].

For low $T_c$ superconductors (LTSC) and MgB$_2$ the pairing is mediated by phonons [3], but for HTSC the binding “glue” for electrons is not yet confidently known [1,2]. Here I propose a new type of “non-equilibrium” spectroscopy, which has an ability to unambiguously disclose the pairing mechanism of HTSC. The idea of the experiment is illustrated in Fig. 1a). When non-equilibrium quasiparticles are injected into the superconductor through a tunnel junction, they relax to the ground state eventually recombining into a Cooper pair. This is accompanied by emission of radiation, as shown in Fig. 1 b). Importantly, those bosonic excitations which are mediating superconductivity are

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**Figure 1.** a) A schematic energy diagram of a superconductor. Non-equilibrium quasiparticles are injected at $E_0 - \Delta$. Relaxation of the quasiparticles typically follow a two-step process: first a quasiparticle fall to the bottom of the band emitting bremsstrahlung radiation; then two quasiparticles at the bottom of the band recombine into a Cooper pair, emitting a recombination radiation. Fig. 1 b) shows the spectrum of radiation caused by relaxation of non-equilibrium quasiparticles. Importantly, recombination radiation consists of bosonic excitations which are mediating superconductivity. Therefore identification of those excitations will provide unambiguous information about the coupling mechanism in HTSC. Fig 1 c) shows a sketch of a triple mesa structure micro/nano-patterned on top of Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ single crystal, which was employed in Ref.[7] for generation and in-situ detection of non-equilibrium recombination radiation in HTSC. Two current biased stacks of intrinsic tunnel junctions (Gen.1 and Gen.2) generated, and an unbiased stack (Det.) detected the non-equilibrium radiation.
emitted upon recombination of two quasiparticles. For example, phonons are emitted in LTSC. This was clearly demonstrated in phonon generation-detection experiments [6]. In that case two LTSC tunnel junctions were deposited on opposite sides of a single crystal. Relaxation of non-equilibrium quasiparticles in the biased junction generated non-equilibrium phonons, which propagated to the opposite side of the crystal and were absorbed and detected by the second junction.

Recently a similar experiment was performed with Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ HTSC [7]. Interlayer tunneling was employed for creation of non-equilibrium quasiparticle distribution in CuO$_2$ planes within small mesa structures micro/nano-fabricated on top of a Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ single crystal, as shown in Fig 1 c). Such mesas represent natural stacks of atomic scale “intrinsic” tunnel junctions. Strong recombination radiation was detected by the detector stack when the voltage per junction in the generator stacks exceeded $2\Delta/e$, where $\Delta$ is the superconducting energy gap.

This brings us to the main point of this paper. Non-equilibrium radiation consists of excitations that mediate superconductivity. Therefore, to determine the mechanism of HTSC one needs to identify the content of recombination radiation: whether it consists of phonons, spin waves or something else. Discrimination can be done by forcing the radiation into an intermediate media, which filters away certain modes (e.g. phonons can propagate through single crystals [6] but not through the open space); or through the time-of-flight [6] experiments: phonons (ionic excitations) are slow, while electronic excitations are fast. The main advantage of the proposed non-equilibrium spectroscopy is that it may provide a direct, and, potentially, interpretation-free answer about the coupling interaction that causes the high temperature superconductivity in cuprates.

**Figure 2.** Simulated non-equilibrium quasiparticle a) and boson b) distribution in a stack of five SIS junctions, as a function of the voltage per junction. Note that two jumps in the distributions functions occur at $eV = 2\Delta$ and $4\Delta$. The former corresponds to the onset of tunnelling, while the latter to the secondary quasiparticle band, due to pairbreaking by bremsstrahlung bosons. c) Suppression of the gap due to non-equilibrium quasiparticle distribution. d) Simulated $dI/dV(V)$ characteristics at $eV=4\Delta$. A peculiarity, caused by appearance of the secondary quasiparticle band is seen. For comparison, a curve without taking into account gap suppression is shown by the red line.
2. Theoretical analysis on non-equilibrium quasiparticle relaxation, due to electron-boson interaction in superconductors.

Relaxation of non-equilibrium quasiparticles occurs via emission of bosons, due to electron-boson interaction. For LTSC, electron-phonon interaction is the main mechanism of relaxation [6]. Such interaction is described by Eliashberg’s electron-phonon spectral function. The non-equilibrium distributions can be obtained by solving a system of kinetic balance equations [8], taking into account electron relaxation with absorption and emission of bosons, pair breaking and recombination, particle injection and escape rates. Those equations have to be supplemented by the self-consistency equation for \( \Delta \) [8]. The detailed description of the formalism, used in numerical simulations presented below, can be found in the supplementary to Ref. [7]. In Fig. 2 I present results of numerical solution of a complete system of non-linear kinetic balance equations with the self-consistency equation for the case of voltage-biased stack of five SIS junction, at different voltages per junction and at \( T = 0.5 T_c \).

Fig. 2 a) and b) show simulated non-equilibrium parts of distribution functions at the lowest energies for electrons and bosons, respectively. At \( eV > 2\Delta \), corresponding to the onset of tunnel current through the SIS junction, primary non-equilibrium quasiparticles appear. Relaxation of those creates primary bosons (note that at \( \Omega = 0 \) there is a net absorption of bosons by non-equilibrium quasiparticles [6b]), which are divided in the bremsstrahlung band, \( 0 < \Omega < eV - 2\Delta \), and the recombination band \( \Omega > 2\Delta \), as shown in Fig. 1 b). At \( eV > 4\Delta \), the bremsstrahlung and recombination bands collapse [7], i.e., the top energy of the bremsstrahlung bosons becomes larger that \( 2\Delta \), sufficient for pair breaking. The latter creates secondary non-equilibrium particles, as shown in Figs. 2 a,b). The appearance of non-equilibrium quasiparticles suppresses the superconducting gap, as shown in Fig. 2 c). This in turn leads to appearance of peculiarities in \( dI/dV \) characteristics at \( eV \) equal to even-integer number \( \Delta \), as shown in Fig. 2 d), qualitatively consistent with peculiarities observed experimentally in stacked Bi-2212 intrinsic Josephson junctions [7].

As noted in Ref. [7], stacking of junctions can lead to dramatic amplification of non-equilibrium phenomena due to cascade amplification of boson population in the stack (note approximately factor five larger boson, than electron, population in Figs. 2 a and b). This may lead to novel non-linear phenomena, such as quantum cascade-like lasing [7] due to enhanced stimulated emission upon collision of bremsstrahlung and recombination boson bands at \( eV = 2(n+1)\Delta \) (n-integer).

To conclude, recombination on non-equilibrium quasiparticles in superconductors is accompanied by emission of the excitation that couples the Cooper pair. This is the basis for the proposed non-equilibrium spectroscopy of high \( T_c \) superconductors. This new technique should operate in the following way: non-equilibrium quasiparticles should be injected in HTSC and the outgoing non-equilibrium radiation (which was already observed [7]) must be identified qualitatively. Quasiparticle injection in HTSC can be easily achieved by employing natural intrinsic Josephson junctions, which also provide cascade amplification of non-equilibrium radiation [7], advantageous for the proposed technique. Here I have also demonstrated that the complete system of non-linear kinetic balance and the self-consistency equations can be solved, which is necessary for quantitative understanding of non-equilibrium phenomena.

[1] 2005, Science 310, 1271 and 1885;
[2] 2006, Nature Physics 2, 134-136, 138-143, and 159-168
[3] J.Akimitsu, et al., 2005, Prog.Theor.Phys. Suppl. 159, 326
[4] A.Damascelli, Z.Hussain and Z.X.Shen, 2003, Rev. Mod. Phys. 75, 473
[5] according to L.D.Landau, see M.Bessarab “Landau” ISBN 5-239-00143-X
[6] W. Eisenmenger and A.H. Dayem, 1967, Phys. Rev. Lett. 18, 125; A.H.Dayem and J.J.Wiegand, 1972, Phys. Rev. B 5, 4390; R.C.Dynes and V.Narayanamurti, 1972, Phys. Rev. B 6, 143.
[7] V.M.Krasnov, 2006, Phys.Rev.Lett. 97, 257003.
[8] J.J. Chang and D.J. Scalapino, 1977, Phys. Rev. B. 15, 2651.