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# Non-equilibrium effects due to charge fluctuations in intrinsic Josephson systems

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## Abstract

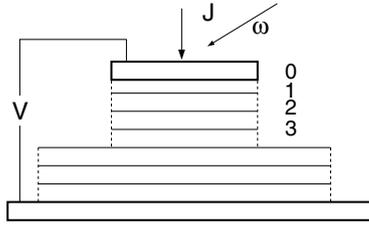
Non-equilibrium effects in layered superconductors forming a stack of intrinsic Josephson junctions are investigated. We discuss two basic non-equilibrium effects caused by charge fluctuations on the superconducting layers: (a) the shift of the chemical potential of the condensate and (b) charge imbalance of quasi-particles, and study their influence on  $I$ – $V$  curves and the position of Shapiro steps.

## 1. Introduction

For the strongly anisotropic cuprate superconductors  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (BSCCO) and  $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$  (TBCCO) the electromagnetic transport perpendicular to the  $\text{CuO}_2$  layers can be described well by a model where the  $\text{CuO}_2$  planes with the intermediate material form a stack of Josephson junctions. This intrinsic Josephson effect can be seen in the multibranch structure of the  $I$ – $V$  curves, but also in the behaviour of the material in external magnetic fields and under high-frequency irradiation [1–4]. Here also, Shapiro steps have been observed [2, 5–7].

In the presence of a bias current each junction of the stack can be either in the resistive or the superconducting state. In the resistive state a finite dc voltage  $V$  appears together with voltage oscillations with a frequency  $\Omega$  given by the Josephson relation  $\hbar\Omega = 2 eV$ . Both effects are accompanied by static and oscillating charge fluctuations on the layers. In a system of weakly coupled very thin atomic layers, such charge fluctuations may lead to non-equilibrium effects described by a shift of the chemical potential of the condensate and a charge imbalance between electron- and hole-like quasi-particles [8–14]. This is different to the case for classical Josephson systems with massive superconducting electrodes. Here similar charge fluctuations occur, but they are restricted to the surface of the superconducting electrode and do not affect the superconductor as a whole, and in particular do not reach the normal electrode.

It is a non-trivial question whether such non-equilibrium effects have measurable consequences. In an experiment like that of Clarke [16] several contacts are necessary to



**Figure 1.** A typical mesa structure used for two-point measurements, consisting of a stack of superconducting layers  $n = 1, 2, \dots$ , a normal electrode  $n = 0$  on top, and a base electrode.

measure the difference between the chemical potentials of the condensate and the quasi-particles. Experiments on cuprate superconductors are mostly done on mesa structures (see figure 1), where the top layer is covered with one gold electrode, and the same electrode is used to inject the current and measure the voltage (for a four-probe measurement see [3]). Also in such situations non-equilibrium effects should be observable for instance through their influence on the  $I$ - $V$  curves: there should be a difference in total voltage depending on whether two resistive junctions are neighbours in the stack or separated by a junction in the superconducting state. Irregularities in the brushlike structure of the  $I$ - $V$  curves, however, can also be produced by other effects, e.g. fluctuations in the critical current density. Therefore in this paper we investigate in particular the possible influence of non-equilibrium effects on the voltage position of the Shapiro steps, which should be more precise. We also add some theoretical considerations for measurements on systems with multiple contacts, where it is possible to measure the charge imbalance relaxation directly.

In a Shapiro-type experiment, high-frequency radiation is applied to the sample, for example to an antenna connected to the contact on top of the mesa. For sufficiently strong radiation, the internal Josephson oscillation frequency of a junction in the resistive state is locked to the external frequency  $\omega$ , fixing the voltage in a small current interval. For a single junction in the resistive state, one expects a set of Shapiro steps in the  $I$ - $V$  curve at voltages  $V = V_{\text{cont}} + m\hbar\omega/(2e)$ . Here  $V_{\text{cont}}$  is the voltage due to the contact resistance between the normal electrode and the first superconducting layer. It is determined by measuring separately the  $I$ - $V$  curve in the superconducting state. A similar result is expected if  $m$  junctions are in the resistive state and we restrict ourselves to the first Shapiro step in each branch. In this paper we will show that the positions of the steps may be changed into  $V = V_{\text{cont}} + m\hbar\omega/(2e) - \delta V$ , where again  $V_{\text{cont}}$  is the contact resistance in the superconducting state. This happens if the first resistive junction is close to the normal electrode. The apparent reduction of the Shapiro step voltage is indirectly due to a change of the contact voltage in the presence of a non-equilibrium charge distribution on the first superconducting layer, and  $\delta V$  is proportional to the relaxation time of the quasi-particle charge. A similar result is obtained for the position of the  $m$ th Shapiro step on the first branch.

In a recent experiment on a BSCCO sample, a downshift of the first Shapiro step on the first branch has been observed [6]. Experiments on other samples show more complicated structures which could not be interpreted completely. Therefore in this paper we concentrate on the theoretical aspects of non-equilibrium effects in layered superconductors. In the following section we summarize the basic results of a theory developed by one of us (DAR) [13] which is based on a kinetic equation for the distribution function describing charge imbalance. First, this theory is used to study the influence of non-equilibrium effects on the  $I$ - $V$  curves. Then these results are applied to the calculation of Shapiro steps and finally to the analysis of systems

with multiple contacts. In all these applications of the non-equilibrium theory, we restrict ourselves to stationary effects. We also neglect the influence of high-frequency oscillations on the dc component of the supercurrent, which is a reasonable approximation for weakly coupled superconducting layers (large values of the McCumber parameter). Some results from the numerical simulation of the time-dependent kinetic equations can be found in [13].

Let us briefly mention here also the relation of the present theory to other theoretical approaches. As mentioned above, there are two non-equilibrium effects connected with charge fluctuations: (a) the shift of the chemical potential of the condensate; (b) the charge imbalance of quasi-particles (there is also an influence of non-equilibrium effects on the amplitude of the superconducting order parameter, which will not be considered here). If we take into account the shift of the chemical potential but neglect charge imbalance, we essentially obtain the results of the theory developed by Koyama and Tachiki [8]. In the approach taken by Artemenko and Kobelkov [10] and by our group [12, 14], a systematic perturbation theory in the gauge-invariant scalar potential is used. In these theories, charge imbalance is considered only indirectly as far as it is induced by fluctuations of the gauge-invariant scalar potential. The present theory is more general. Here charge imbalance is taken into account as an independent degree of freedom, and therefore the results are different from those of earlier treatments. A detailed comparison of the different approaches will be given in a future publication.

## 2. Theory of stationary non-equilibrium effects in layered superconductors

### 2.1. Generalized Josephson relation

We consider a system of superconducting layers  $n$  with superconducting order parameter  $\Delta_n(t) = |\Delta| \exp(i\chi_n(t))$  with time-dependent phase  $\chi_n(t)$ , neglecting a possible time dependence of the amplitude of the order parameter. The basic quantity which enters all the Josephson relations is the gauge-invariant phase difference between layer  $n$  and  $n+1$  given by

$$\gamma_{n,n+1}(t) = \chi_n(t) - \chi_{n+1}(t) - \frac{2e}{\hbar} \int_n^{n+1} dz A_z(z, t) \quad (1)$$

where  $A_z(z, t)$  is the vector potential in the barrier. For the time derivative of  $\gamma_{n,n+1}$ , we obtain the generalized Josephson relation:

$$\dot{\gamma}_{n,n+1} = \frac{2e}{\hbar} (V_{n,n+1} + \Phi_{n+1} - \Phi_n). \quad (2)$$

Here

$$V_{n,n+1} = \int_n^{n+1} dz E_z(z, t) \quad (3)$$

is the voltage and  $\Phi_n(t)$  is the so-called gauge-invariant scalar potential defined by

$$\Phi_n(t) = \phi_n(t) - \frac{\hbar}{2e} \dot{\chi}_n(t) \quad (4)$$

where  $\phi_n(t)$  is the electrical scalar potential. In this paper,  $e$  denotes the elementary charge. The charge of the electron is  $-e$ . The quantity  $\hbar\dot{\gamma}_{n,n+1}$  is the total energy required to transfer a Cooper pair between neighbouring layers  $n$  and  $n+1$ . The quantity  $e\Phi_n$  can be considered as the shift of the chemical potential of the superconducting condensate with respect to an average chemical potential. It determines the minimum in  $k$ -space of the quasi-particle excitation energy.

In the following we have to consider fluctuations of the charge densities on the superconducting layers. For this purpose let us briefly mention some basic concepts for the description of non-equilibrium superconductors as introduced by Tinkham and Clarke [16]. In the BCS theory one may split the total electronic charge density on each layer into  $\rho = \rho^s + \rho^q$  with

$$\rho^s = \frac{2}{\text{Vol}} \sum_k v_k^2 \quad \rho^q = \frac{2}{\text{Vol}} \sum_k (u_k^2 - v_k^2) f_k.$$

In equilibrium the latter vanishes because there are equal numbers of electron- and hole-like excitations. In a non-equilibrium state of the superconductor a finite quasi-particle charge may exist, which is called charge imbalance. The quantity  $\rho^s$ , sometimes called condensate charge, should not be confused with the superfluid charge density, which vanishes at the transition temperature. In contrast,  $\rho^s$  approaches the total density in the normal state. Fluctuations of the two charge densities are caused not only by shifts in the chemical potential entering the quasi-particle excitation energy and the functions  $u_k, v_k$ , but also by fluctuations in the distribution functions  $f_k$ . In a quantum-kinetic theory based on quasi-classical Green functions, the same distinction between the two types of charge density can be made. The  $k$ -dependent functions are then replaced by corresponding energy-dependent quantities.

A fluctuation in the (two-dimensional) charge density  $\rho_n^s$  is directly related to the shift of the chemical potential in the layer  $n$ :

$$\rho_n^s(t) = -2e^2 N(0) \Phi_n(t). \quad (5)$$

Here  $N(0)$  is the two-dimensional density of states of the conduction electrons in the layer. Unlike a true chemical potential, which is defined only in equilibrium, the quantity  $\Phi_n(t)$  may be time dependent. In the following we will, however, restrict ourselves to stationary processes where  $\Phi_n$  is independent of time.

It is convenient to express also fluctuations in the charge imbalance with the help of a quasi-particle potential  $\Psi_n$  by writing formally

$$\rho_n^q = 2e^2 N(0) \Psi_n(t). \quad (6)$$

Then we obtain for the total charge-density fluctuation

$$\rho_n(t) = -2e^2 N(0) (\Phi_n(t) - \Psi_n(t)). \quad (7)$$

The calculation of the charge imbalance  $\rho_n^q$  will be discussed in section (2.3) for the stationary case.

With the help of (7) and the Maxwell equation ( $d$  is the distance between the layers)

$$\rho_n = \frac{\epsilon_0}{d} (V_{n,n+1} - V_{n-1,n}) \quad (8)$$

the generalized Josephson relation now reads

$$\frac{\hbar}{2e} \dot{\gamma}_{n,n+1} = (1 + 2\alpha) V_{n,n+1} - \alpha (V_{n-1,n} + V_{n+1,n+2}) + \Psi_{n+1} - \Psi_n \quad (9)$$

with  $\alpha = \epsilon_0 / (2e^2 N(0) d)$ . This shows that the Josephson oscillation frequency is determined not only by the voltage in the same junction, but also by the voltages in neighbouring junctions. Furthermore, it is influenced by the quasi-particle potential  $\Psi$  on the layers. If we neglect the latter, we obtain for  $\dot{\gamma}_{n,n+1}$  the same result as in [8].

For a Josephson junction in the presence of sufficiently strong high-frequency irradiation, the average Josephson oscillation frequency is locked to the external frequency  $\omega$ . From the generalized Josephson relation (9) we obtain for the  $m$ th Shapiro step

$$m \frac{\hbar \omega}{2e} = \frac{\hbar}{2e} \langle \dot{\gamma}_{n,n+1} \rangle = (1 + 2\alpha) V_{n,n+1} - \alpha (V_{n-1,n} + V_{n+1,n+2}) + \Psi_{n+1} - \Psi_n \quad (10)$$

where the dc components of the quantities on the rhs have to be used. This relation can also be used to describe the voltage for a barrier in the superconducting state; then the lhs is zero:

$$0 = (1 + 2\alpha)V_{n,n+1} - \alpha(V_{n-1,n} + V_{n+1,n+2}) + \Psi_{n+1} - \Psi_n. \quad (11)$$

## 2.2. Current equation

In order to calculate the electronic transport in layered superconductors we need expressions for the current and charge density. The results are derived with help of a Keldysh technique for non-equilibrium Green functions. In particular, the quasi-particle charge is calculated from a kinetic equation for the corresponding quasi-particle distribution function. For the current density the following approximate result is obtained:

$$j_{n,n+1} = j_c \sin \gamma_{n,n+1} + \frac{\sigma_{n,n+1}}{d} \left( \frac{\hbar}{2e} \dot{\gamma}_{n,n+1} + \Psi_n - \Psi_{n+1} \right). \quad (12)$$

The first term is the current density of Cooper pairs. The rest is the quasi-particle current density (we neglect here the interference term proportional to  $\cos \gamma$ ). The physical meaning of the latter becomes more apparent if we express these terms, with help of (7), (8), using the charge densities:

$$j_{n,n+1} = j_c \sin \gamma_{n,n+1} + \frac{\sigma_{n,n+1}}{d} \left( V_{n,n+1} + \frac{\rho_n}{2e^2 N(0)} - \frac{\rho_{n+1}}{2e^2 N(0)} \right) \quad (13)$$

which can also be written as

$$j_{n,n+1} = j_c \sin \gamma_{n,n+1} + \frac{\sigma_{n,n+1}}{d} \left( (1 + 2\alpha)V_{n,n+1} - \alpha(V_{n-1,n} + V_{n+1,n+2}) \right). \quad (14)$$

Then we see that the quasi-particle current is driven not only by the voltage, but also by a diffusion term proportional to the difference in charge between the two layers. The expression (13) for the current density is valid for temperatures close to  $T_c$ . For  $T \ll T_c$  the conductivities are different for the two contributions of the quasi-particle current (they correspond to the quantities  $\sigma_0$  and  $\sigma_1$  introduced by Artemenko and Kobelkov [10]). For simplicity, we neglect this difference in the following.

Note that (13), (14) also stay meaningful in the normal state for a system of junctions with different conductivities. Then  $\alpha$  describes the shift of the chemical potential due to charge accumulation in the normal state.

In the stationary state the dc density  $j_{n,n+1}$  is the same for all barriers and is equal to the bias current density  $j$ , i.e. there is no contribution from the displacement current. If the Josephson junction is in the resistive state and in the absence of external high-frequency irradiation we may neglect the dc component of the supercurrent density in (14) and obtain

$$\frac{j d}{\sigma_{n,n+1}} = (1 + 2\alpha)V_{n,n+1} - \alpha(V_{n-1,n} + V_{n+1,n+2}). \quad (15)$$

This approximation is valid as long as the effect of high-frequency charge oscillations in the phase difference  $\gamma_{n,n+1}(t)$  and their influence on the dc density  $j_{n,n+1}^s := \langle j_c \sin \gamma_{n,n+1}(t) \rangle$  can be neglected. In general, the time dependence of the phase difference in the resistive state is of the form  $\gamma_{n,n+1}(t) = \Omega t + \delta\gamma_{n,n+1}(t)$  where  $\delta\gamma_{n,n+1}(t)$  oscillates with the Josephson frequency  $\Omega$  (plus higher harmonics) determined by the corresponding dc voltage  $\hbar\Omega \simeq 2eV_{n,n+1}$ . The amplitude of the high-frequency oscillation  $\delta\gamma_{n,n+1}(t)$  and its influence on  $j_{n,n+1}^s$  can be estimated along the lines of [15]. In the limit where the Josephson frequency  $\Omega$  is large compared to the Josephson plasma frequency  $\omega_p = \sqrt{2edj_c/(\hbar\epsilon_0)}$  and furthermore the quasiparticle conductivity  $\sigma$  is low,  $\sigma/(\Omega\epsilon_0) \ll 1$ , the direct-supercurrent density  $j_s \simeq (\Omega_p^4/\Omega^4)j^{\text{qp}}/2$  is a small correction to the quasiparticle current density  $j^{\text{qp}}$ . These conditions

are well fulfilled for strongly anisotropic BSCCO (with high values of the McCumber parameter) at low temperatures and intermediate voltages (but not for YBCCO). In this limit this dynamic effect leads to a further weak coupling of dc voltages in neighbouring layers which is investigated numerically in [12] and is neglected here in comparison with the stationary effects due to non-equilibrium quasi-particle charges.

For a correct determination of the total voltage of a stack as shown in figure 1, an investigation of the influence of non-equilibrium effects on the contact voltage between the normal electrode and the first superconducting layer is necessary. The physical nature of this contact is not known precisely. It may vary between a tunnelling contact and a metallic contact with superconductivity partially suppressed on the first superconducting layer by the proximity effect. In the following, we assume that this contact can be described by a tunnelling contact between a thick normal electrode and a thin superconducting layer. Then we may neglect the shift of the chemical potential in the normal layer and obtain for the current density between the normal electrode ( $n = 0$ ) and the first superconducting layer ( $n = 1$ ) (compare with (15))

$$\frac{jd}{\sigma_{0,1}} = (1 + \alpha)V_{0,1} - \alpha V_{1,2}. \quad (16)$$

There might be a small shift of the chemical potential on a thin layer at the surface of the normal electrode. This can be included in the theory, but has no influence on the total voltage. For notational simplicity, we have assumed here that the width  $d$  of the barrier between the normal electrode and the first superconducting layer is the same as that between the two superconducting layers. This can easily be generalized but does not change the content of the final results. Note that the quasi-particle potentials appear explicitly only in the equations derived from the generalized Josephson equation and not in the current equations (15), (16).

### 2.3. Relaxation of the quasi-particle charge

In addition to the current equation, the microscopic theory provides us with an equation for the charge density. As the quasi-particle charge is only part of the total density, an additional mechanism for the charge imbalance relaxation has to be introduced. In the stationary case the following result for the quasi-particle charge is obtained [13]:

$$\rho_n^q = -2e^2 N(0) \Psi_n = \tau_q (j_c \sin \gamma_{n,n+1} - j_c \sin \gamma_{n-1,n}) \quad (17)$$

where  $\tau_q$  is the charge imbalance relaxation time.

This result can be made plausible in a simple way. Let us start from a balance equation for the superfluid charge density:

$$0 = j_c \sin \gamma_{n-1,n} - j_c \sin \gamma_{n,n+1} - \left. \frac{\partial \rho_n^q}{\partial t} \right|_{\text{conv}}. \quad (18)$$

The first two terms on the rhs describe the change of the superfluid density due to supercurrents between the neighbouring layers. In the stationary state this is balanced by the conversion of quasi-particle charge into condensate charge (the last term in (18)). We assume that in the stationary case this conversion can be described by a relaxation process

$$\left. \frac{\partial \rho_n^q}{\partial t} \right|_{\text{conv}} = -\frac{1}{\tau_q} \rho_n^q. \quad (19)$$

Using this expression in (18), the result (17) is obtained.

The relation (17) can also be written in another way which will be useful in the following. Using the continuity equation for the total current density in the stationary case, we find for the quasi-particle current density

$$0 = j_{n-1,n}^{\text{qp}} - j_{n,n+1}^{\text{qp}} + \left. \frac{\partial \rho_n^q}{\partial t} \right|_{\text{conv}}. \quad (20)$$

Then,

$$\Psi_n = d(j_{n-1,n}^{\text{qp}} - j_{n,n+1}^{\text{qp}})/\sigma_q \quad (21)$$

with  $\sigma_q = 2e_0^2 N(0)d/\tau_q$ .

For the later discussion it will be useful to introduce a relaxation parameter  $\eta_{n,n+1} \equiv \sigma_{n,n+1}/\sigma_q$ . In the following we will assume that non-equilibrium effects are small, i.e.  $\alpha \ll 1$ ,  $\eta_{n,n+1} \ll 1$ . In first order in  $\alpha$  and  $\eta$  we may then use the approximation  $j_{n,n+1}^{\text{qp}} = \sigma_{n,n+1} V_{n,n+1}/d$  for the calculation of  $\Psi_n$  and obtain

$$\Psi_n \simeq \eta_{n-1,n} V_{n-1,n} - \eta_{n,n+1} V_{n,n+1} \quad (22)$$

or for a barrier in the resistive state we may replace  $j_{n,n+1}^{\text{qp}}$  in (21) by the total current density  $j$ .

The relaxation parameter  $\eta$  can also be written as  $\eta = \tau_q/\tau_t$ , where  $\tau_t$  is a typical tunnelling time. In the case of weak non-equilibrium,  $\eta \ll 1$ , considered here, the charge imbalance has relaxed before quasi-particles have tunnelled to the next layer.

The charge imbalance potential  $\Psi_n$  vanishes for a layer between two junctions in the superconducting state, when the current is carried exclusively by Cooper pairs. It vanishes also for a layer between two junctions in the resistive state, when the current is carried primarily by quasi-particles. A non-zero value of  $\Psi_n$  can be found, in particular, for a layer between one junction in the superconducting and one junction in the resistive state. As  $\tau_q$  is proportional to  $1/\Delta$  close to  $T_c$ , but  $j_c$  is proportional to  $\Delta^2$ , the charge imbalance vanishes at  $T_c$ .

These results are derived here for the stationary case, which will be needed for the discussion of dc properties in the following. In the presence of a time-dependent chemical potential  $\Psi_n(t)$ , an additional term proportional to  $d\Psi_n(t)/dt$  has to be added in (19). The results for the time-dependent case are discussed in [13].

### 3. Influence of non-equilibrium effects on the $I$ - $V$ curves

As a first application of the theory outlined above, we study the influence of non-equilibrium effects on the  $I$ - $V$  curves, i.e. we calculate the total voltage for a stack of junctions, where one or more Josephson junctions are in the resistive state in the absence of high-frequency irradiation. Special attention is paid to the contact voltage. Here we consider explicitly only one contact. The effect of the other contact can be added easily to the final result.

#### 3.1. All junctions in the superconducting state

Let us start the discussion with the case where all junctions are in the superconducting state. Then the voltages are determined by the following set of equations. Equation (16) is used for the contact with the normal electrode and (11) with  $n \geq 1$  for the junctions in the superconducting state. Adding up these equations we obtain for the total voltage, which is the contact voltage in the superconducting state,

$$V = V_{\text{cont}} = \frac{j d}{\sigma_{0,1}} + \Psi_1. \quad (23)$$

For the quasi-particle potentials we have

$$\Psi_1 = \eta_{0,1}jd/\sigma_{0,1} = jd/\sigma_q \quad \Psi_n = 0 \quad \text{for } n \geq 2. \quad (24)$$

Then the contact voltage is

$$V_{\text{cont}} = V_{\text{cont}}^0 + \delta V \quad (25)$$

with  $\delta V = jd/\sigma_q$  and the ‘bare’ contact voltage  $V_{\text{cont}}^0 = jd/\sigma_{0,1}$ . We see that the contact voltage is changed by the charge imbalance induced by the quasi-particle current on the first superconducting layer.

### 3.2. One or two junctions in the resistive state

If the first Josephson junction (between layers 1 and 2) is in the resistive state, but all the other junctions are in the superconducting state, then the contact with the normal electrode is again described by (16). For the resistive junction we use the current relation (15) with  $n = 1$ , for the superconducting junctions (11) we use that with  $n \geq 2$ .

Adding up these equations, we find for the total voltage

$$V = \frac{jd}{\sigma_{0,1}} + \frac{jd}{\sigma_{1,2}} + \Psi_2 \quad (26)$$

where we have used  $\Psi_n = 0$  for  $n \geq 3$ . The non-equilibrium effect now comes from the second layer, where  $\Psi_2 = \eta_{1,2}jd/\sigma_{1,2} = jd/\sigma_q$ . Here and in the following we assume that the charge imbalance relaxation rate is the same on all superconducting layers. Then the total voltage can also be written as

$$V = V_{\text{cont}} + \frac{jd}{\sigma_{1,2}}. \quad (27)$$

The  $I$ – $V$  curve of the first branch appears at the usual voltage after subtracting the contact voltage. The case is different if the Josephson junction in the resistive state is not close to the normal contact. Then one obtains for the total voltage

$$V = V_{\text{cont}} + \frac{jd}{\sigma_{n,n+1}}(1 + 2\eta_{n,n+1}) = V_{\text{cont}} + \frac{jd}{\sigma_{n,n+1}} + \delta V \quad (28)$$

with  $\delta V = 2jd/\sigma_q$ . The first branch has a slightly larger voltage for the same current density. The voltage shift  $\delta V$  is due to the charge imbalance potentials generated by the quasi-particle currents on the two layers  $n$  and  $n + 1$  between the resistive junction and the neighbouring superconducting junctions.

Of special interest is the case where two junctions are in the resistive state, if we compare the total voltage for the case where the resistive junctions are neighbours with the situation where they are separated by one or more barriers in the superconducting state. In the latter case the total voltage is again larger by  $\delta V = 2jd/\sigma_q$ . This shift is due to the charge imbalance potential on the two additional boundaries separating the resistive junctions from the junctions in the superconducting state. This result can be easily generalized to the case of several resistive junctions.

In the present theory the shift of the chemical potential described by the coefficient  $\alpha$  has no influence on the total voltage. This is a consequence of the special form of the quasi-particle current density used here (compare with (12), (14)). This result is different from those of earlier treatments [17], where for the quasi-particle current density a simple ansatz of the form  $j_{n,n+1}^{\text{qp}} = \sigma V_{n,n+1}/d$  has been used.

#### 4. Influence on Shapiro steps

For a Josephson junction in the presence of strong high-frequency irradiation, the current relation (15) is not useful, since on a Shapiro step the current is not fixed by the voltage. Instead, the Josephson oscillation frequency is locked to the external radiation. Then we have to use the generalized Josephson relation (10) to determine the voltage. Let us assume that the first Josephson junction (between layers 1 and 2) is in the resistive state and all other junctions are in the superconducting state; then the voltages for the different barriers are again determined by (16, 11), but for the resistive junction with a first Shapiro step we have to use

$$\frac{\hbar\omega}{2e} = (1 + 2\alpha)V_{1,2} - \alpha(V_{0,1} + V_{2,3}) + \Psi_2 - \Psi_1. \quad (29)$$

With  $\Psi_n = 0$  for  $n \geq 3$  we obtain for the total voltage

$$V = \frac{jd}{\sigma_{0,1}} + \frac{\hbar\omega}{2e} + \Psi_1. \quad (30)$$

In this case the quasi-particle potential  $\Psi_1$  is given by

$$\Psi_1 = d(j_{0,1}^{\text{qp}} - j_{1,2}^{\text{qp}})/\sigma_q. \quad (31)$$

As  $j_{0,1}^{\text{qp}} = j$  and  $j_{1,2}^{\text{qp}} \simeq j$  at the centre of the Shapiro step,  $\Psi_1$  vanishes in this case. Therefore we find for the total voltage

$$V = V_{\text{cont}}^0 + \frac{\hbar\omega}{2e} \quad (32)$$

where  $V_{\text{cont}}^0 = jd/\sigma_{0,1}$  is the ‘bare’ contact voltage. If we introduce instead the contact voltage which is measured in the superconducting state, we have

$$V = V_{\text{cont}} + \frac{\hbar\omega}{2e} - \delta V \quad (33)$$

with  $\delta V = jd\eta_{0,1}/\sigma_{0,1} = jd/\sigma_q$ .

The apparent shift of the Shapiro step for the first Josephson junction is due to the difference of the charge imbalance potential  $\Psi_1$  on the first superconducting layer in the pure superconducting state and in the resistive state (the effect of the charge imbalance  $\Psi_2$  on the second layer drops out in the total voltage). In order to get such an experimental shift of the Shapiro step voltage it is essential that the Josephson junction in resonance with the external radiation is close to the normal electrode. Otherwise the quasi-particle potentials  $\Psi_n$  drop out in the total voltage.

The result can easily be extended to the case where the first  $m$  Josephson junctions are in the resistive state and in resonance with the external radiation. In that case we find

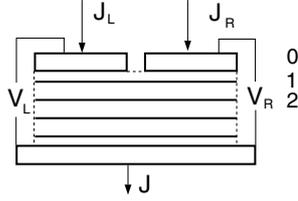
$$V = V_{\text{cont}} + m \frac{\hbar\omega}{2e} - \delta V \quad (34)$$

with  $\delta V = jd\eta_{0,1}/\sigma_{0,1} = jd/\sigma_q$ . Again it is assumed that the first Josephson junction in the resistive state is close to the normal electrode; otherwise  $\delta V = 0$ . Note that only the voltage of the Shapiro step on the first branch is shifted; the difference between the other Shapiro steps is still at the ‘right’ voltage  $\hbar\omega/(2e)$ .

A similar result is obtained for higher-order Shapiro steps on the first branch. Then the voltage of the  $m$ th Shapiro step is given by

$$V = V_{\text{cont}}(j) + m \frac{\hbar\omega}{2e} - \delta V(j) = V_{\text{cont}}^0(j) + m \frac{\hbar\omega}{2e}. \quad (35)$$

As the Shapiro steps appear here at different current values, we have to take into account the current dependence of the contact voltage explicitly. Then all Shapiro steps appear to be shifted if the contact voltage in the superconducting state is subtracted.



**Figure 2.** A stack with two normal electrodes used to measure the charge imbalance relaxation rate.

## 5. Four-point measurements

In the preceding section we have investigated the influence of charge imbalance on the shift of Shapiro steps. This effect is indirectly due to a change of the contact voltage between the normal electrode and the first superconducting layer caused by charge imbalance. With the help of a four-point measurement it is possible to determine the contact voltage separately. Furthermore, a direct determination of the charge imbalance and its relaxation rate will be possible.

We consider a stack with two normal electrodes on top (see figure 2). Then one contact can be used to inject a current and the other contact to measure the voltage in the absence of a current.

Let us denote the current densities and voltages of the left and right electrode (with length  $l_{L,R}$ ) by  $j_{L,R}$  and  $V_{L,R}$ . We assume that the currents injected through the left and right electrode are different; in particular, we are interested in the case  $j_R = 0$ .

First we consider the case where all inner junctions are in the superconducting state. Then the individual voltages are determined by the following set of equations:

$$\frac{j_{L,R}d}{\sigma_{0,1}} = (1 + \alpha)V_{0,1}^{L,R} - \alpha V_{1,2}^{L,R} \quad (36)$$

$$0 = (1 + 2\alpha)V_{n,n+1}^{L,R} - \alpha(V_{n-1,n}^{L,R} + V_{n+1,n+2}^{L,R}) + \Psi_{n+1} - \Psi_n \quad \text{for } n \geq 1. \quad (37)$$

The injected currents induce a charge imbalance potential  $\Psi_1$  on the first superconducting layer controlled by the quasi-particle currents. We assume that the diffusion length of quasi-particles along the layer is large compared to the width of the stack. Then we obtain a uniform charge imbalance potential:

$$\Psi_1 = \left( \frac{l_L}{l} j_L + \frac{l_R}{l} j_R \right) \frac{d}{\sigma_q}. \quad (38)$$

Note that the total charge on the first superconducting layer in general is not uniform and the voltages  $V_L$  and  $V_R$  are different, but the electrochemical potential which is controlled by  $\langle \gamma_{n,n+1} \rangle$  is constant along the layers.

For the voltages at the left and right contact we then obtain

$$V_{L,R} = \frac{j_{L,R}d}{\sigma_{0,1}} + \Psi_1. \quad (39)$$

In the case  $j_R = 0$  we find in particular

$$V_L - V_R = \frac{j_L d}{\sigma_{0,1}} \quad V_R = V_L \frac{l_L}{l} \eta_{0,1} \quad (40)$$

which allows us to measure the relaxation parameter  $\eta_{0,1}$  and the ‘bare’ contact voltage  $j_L d / \sigma_{0,1}$  separately.

Now we consider the case where the barrier between the first two superconducting layers is in the resistive state. Then a similar calculation leads to

$$V_{L,R} = \frac{j_{L,R}d}{\sigma_{0,1}} + \frac{jd}{\sigma_{1,2}} + \Psi_2 \quad (41)$$

with  $\Psi_2 = jd\eta_{1,2}/\sigma_{1,2} = jd/\sigma_q$ . Now the non-equilibrium effect comes from the second superconducting layer. Note that the current density flowing through the barrier (1), (2) is the total current density  $j$ . In the special case  $j_R = 0$  a comparison between  $V_L$  and  $V_R$  allows us to determine the ‘bare’ contact voltage of the left electrode.

Finally let us discuss the results for applied high-frequency radiation. If the barrier between the first two superconducting layers is in the resistive state and is in resonance with the radiation frequency  $\omega$ , the first Shapiro step appears at the voltage

$$V_{L,R} = \frac{j_{L,R}d}{\sigma_{0,1}} + \frac{\hbar\omega}{2e} + \Psi_1 \quad (42)$$

with  $\Psi_1 \simeq 0$  in the centre of the Shapiro step. In particular, for  $j_R = 0$  the voltage at the right electrode measures the Shapiro step voltage directly without the contact voltage. The same is true for the voltage of the  $m$ th Shapiro step on the first branch.

## 6. Summary

In this paper we have discussed the influence of non-equilibrium effects on stationary properties of Josephson contacts in layered superconductors. In particular, we have studied the influence on the  $I$ - $V$  characteristics and on the voltage position of the Shapiro steps. We find that of the two basic non-equilibrium effects, (a) shift of the chemical potential of the condensate and (b) charge imbalance of quasi-particle charges, only the latter has an influence on these properties, while the chemical potential shift drops out in the total voltage. This result depends crucially on the form of the quasi-particle current density and the correct treatment of the contact resistance. Note that for time-dependent processes, like collective modes, the coupling between superconducting layers due to the chemical potential shift is also relevant [8].

In our theory the charge imbalance relaxation plays an important role. Microscopically this relaxation is caused by inelastic scattering processes within the layer. In the case of d-wave superconductors, elastic scattering may also contribute. In the present treatment a phenomenological relaxation rate has been introduced.

In our investigation of Shapiro steps we find a shift of the voltage of the first Shapiro step on the first resistive branch of the  $I$ - $V$  curve if the Josephson junction in the resistive state is close to the normal electrode. This apparent shift, which is not a violation of the basic Josephson relations, can be traced back to a change of the contact voltage between the normal electrode and the first superconducting layer due to charge imbalance on the first superconducting layer in the presence of a finite quasi-particle current. The distance between higher-order Shapiro steps on the first branch is also changed due to the current dependence of the contact voltage.

Recently, Shapiro step experiments have been performed in the THz regime on mesa structures of BSCCO [6] with normal gold electrodes. The voltage position of the Shapiro step has been determined by subtracting the contact voltage in the superconducting state in the absence of radiation. In one experiment a 3% downshift of the step position on the first branch has been observed, i.e. the ratio between charge imbalance lifetime and tunnelling time is  $\eta = \tau_q/\tau_t = 0.03$ . In other samples, Shapiro steps on higher branches could also be detected. Their positions showed a more complicated structure which up to now is not fully understood. Therefore it is not yet possible to present a detailed comparison with experimental data. These experimental results should be compared with very recent measurements of Shapiro steps in

BSCCO double-step mesa structures [7] with large superconducting electrodes, where no shift of Shapiro steps is observed or expected.

The difficulty with the proper subtraction of the contact voltage can be avoided in four-point measurements. Therefore we have investigated the influence of charge imbalance on the Josephson effect for an experimental set-up where the first superconducting layer is in contact with two normal electrodes, and only one electrode is used to inject the current. In this case the contact voltage is also influenced by the charge imbalance produced by the quasi-particle currents, but the contact voltage can be determined separately by comparing the voltages measured at the two electrodes. Taking that into account, the Shapiro steps appear again at the right position.

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### References

- [1] Kleiner R, Steinmeyer F, Kunkel G and Müller P 1992 *Phys. Rev. Lett.* **68** 2394
- [2] Kleiner R and Müller P 1994 *Phys. Rev. B* **49** 1327  
Müller P 1994 *Festkörperprobleme/Advances in Solid State Physics* vol 34, ed R Helbig (Braunschweig: Vieweg) p 1
- [3] Yurgens A, Winkler D, Zavaritsky N V and Claeson T 1996 *Phys. Rev.* **53** R8887
- [4] Yurgens A A 2000 *Supercond. Sci. Technol.* **13** R85
- [5] Doh Y J, Kim J, Kim K T and Lee H J 2000 *Phys. Rev. B* **61** R3834
- [6] Rother S and Müller P 2001 private communication
- [7] Wang H B, Wu P H and Yamashita T 2001 *Phys. Rev. Lett.* **87** 107002
- [8] Koyama T and Tachiki M 1996 *Phys. Rev. B* **54** 16 183
- [9] Bulaevskii L, Dominguez D, Maley M, Bishop A and Ivlev B 1996 *Phys. Rev. B* **53** 14 601
- [10] Artemenko S and Kobelkov A 1997 *Phys. Rev. Lett.* **78** 3551
- [11] Ryndyk D 1998 *Phys. Rev. Lett.* **80** 3376
- [12] Preis C, Helm C, Keller J, Sergeev A and Kleiner R 1998 *Proc. SPIE* **3480** 236
- [13] Ryndyk D 1999 *Zh. Eksp. Teor. Fiz.* **116** 1798 (Engl. transl. 1999 *JETP* **89** 975 )
- [14] Helm C, Keller J, Preis C and Sergeev A 2001 *Physica C* **362** 43
- [15] Helm C, Preis C, Walter C and Keller J 2000 *Phys. Rev. B* **62** 6002
- [16] Clarke J 1972 *Phys. Rev. Lett.* **28** 1363  
Tinkham M and Clarke J 1972 *Phys. Rev. Lett.* **28** 1366
- [17] Matsumoto H, Sakamoto S, Wajima F, Koyama T and Machida M 1999 *Phys. Rev. B* **60** 3666