Intrinsic Josephson Effect and Violation of the Josephson Relation in Layered Superconductors

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Equations describing the resistive state of a layered superconductor with anisotropic pairing are derived. The similarity with a stack of Josephson junctions is found at small voltages only, when current density in the direction perpendicular to the layers can be interpreted as a sum of the Josephson superconducting, the Ohmic dissipative, and the interference currents. In the spatially uniform state differential conductivity at higher voltages becomes negative. Nonuniformity of the current distribution generates branch imbalance and violates the Josephson relation between frequency and voltage. [S0031-9007(97)03075-5]

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Layered high-T_c superconductors are known to exhibit the intrinsic Josephson effect when the current is flowing across the conducting layers (see, e.g., [1] and recent measurements [2]). Though such materials can be considered as a stack of 2D superconductors linked by the Josephson coupling [3,4], the theory of the ac Josephson effect in tunnel junctions cannot be directly applied to describe the resistive state of layered superconductors. In a system of series connected junctions the electric field is located mainly in the insulating barriers due to screening by the electrons in the metal, and the superconducting banks are in the equilibrium state. This results in many important consequences including the Josephson relation between voltage and frequency. But in the layered materials the superconducting layers are of atomic thicknesses and one must not ignore nonequilibrium effects which are related to perturbations of the quasiparticle distribution in the superconductor. On the other hand, many experimental evidences for d-wave or nearly d-wave symmetry of the superconducting order parameter in layered high-T_c superconductors were given in the last few years, and a compatibility of the experimental data with a d-wave scenario was shown in many theoretical works (see, e.g., [5,6], and references therein). In this case the superconducting order parameter has nodes; i.e., the quasiparticle density is never exponentially small and the nonequilibrium effects due to quasiparticles become especially important. Thus, to understand the intrinsic Josephson effect in high-T_c superconductors one must take into account the nonequilibrium distribution of the quasiparticles and the relaxation processes in the resistive state.

In this study we calculate current and charge densities in superconductors with anisotropic pairing as functions of the phase differences of the order parameter in neighboring layers and of the nonequilibrium scalar potential related to the quasiparticle branch imbalance, i.e., the difference between densities of electronlike and holelike quasiparticles [7,8]. We find that the direct analogy with a stack of Josephson junctions is limited by nonequilibrium effects and scattering processes, the difference with Josephson junctions being the most pronounced at lower temperatures. This results in the negative differential conductivity and in the violation of the Josephson relation. Similar effects are expected in layered superconductors with isotropic pairing, too, but in the latter case the quasiparticle density drops down exponentially when temperature decreases and generation of branch imbalance violating the Josephson relation would be negligible in the most interesting region of low temperatures.

In our calculations we use the quasiclassical theory of nonequilibrium superconductivity [9] modified for the case of layered superconductors [10,11]. We solve the equations in the discrete Wannier representation for the Keldysh [12] matrix propagator, $\hat{G}_{nm}$. Its diagonal components are the retarded and advanced Green's functions, g^R and g^A, and its upper off-diagonal component, g^K, is related to the electron distribution function. We consider the hopping conductivity regime between the layers, $t_\perp \tau \ll \hbar$, which corresponds to the case of Josephson interlayer coupling. Here $t_\perp$ is the overlap integral describing the electron spectrum in the perpendicular direction, $\epsilon_\perp = 2t_\perp \cos dk_\perp$, d is the lattice constant in the perpendicular direction, and τ is the momentum scattering time along the layers. This approach bears some similarity to the interlayer diffusion model [13] in which the interlayer coupling is mediated through incoherent hopping processes, $t_\perp$ being neglected. We assume that a symmetry of the superconducting order parameter is imposed by the symmetry of the coupling potential in the self-consistency condition: thus we do not address the question of the microscopic nature of the interaction resulting in such a symmetry. Then the equation for $\hat{G}_{nm}$ has the form

$$-i\hbar\left(\sigma_z \frac{\partial}{\partial t}\hat{G}_{nm} + \frac{\partial}{\partial t'}\hat{G}_{nm}\sigma_z + \mathbf{v}\nabla\hat{G}_{nm}\right) + t_\perp \sum_{i=\pm 1}(A_{nn+i}\hat{G}_{n+im} - \hat{G}_{nm+i}A_{m+im}) + h_n(t)\hat{G}_{nm} - \hat{G}_{nm}h_m(t') = \frac{i\hbar}{2\tau}(\langle\hat{G}_{nn}\rangle\hat{G}_{nm} - \hat{G}_{nm}\langle\hat{G}_{mm}\rangle), \quad (1)$$

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where ϕ is the angle of the in-plane electron momentum, $\langle\cdots\rangle$ means averaging over ϕ, and $h_n = -i\sigma_y\Delta_n(\phi) + \mu_n + \sigma_z \mathbf{v}\mathbf{p}_n$. Furthermore, $\mu_n = (\hbar/2)\partial\chi_n/\partial t + e\Phi_n$ is the gauge invariant scalar potential in layer n, χ_n is the order parameter phase in the nth layer, Φ_n is the electric potential, $\mathbf{p}_n = (\hbar/2)\nabla\chi_n - (e/c)\mathbf{A}_n$ is the superconducting momentum parallel to the layers, $\mathbf{A}_n$ is the vector potential, and $A_{nm} = \cos[(\chi_n - \chi_m)/2] + i\sigma_z \sin[(\chi_n - \chi_m)/2]$. The Pauli matrices introduced above act on the spin indices of Green's functions. Each $\hat{G}_{nm}$ depends on two times (or on two energies in Fourier representation), and on ϕ.

The right-hand side (rhs) of (1) is an elastic-collision integral in Born approximation. Using Born approximation we neglect low-energy quasiparticle bound states created by impurities (see [14], and references therein), and, hence, our results are applicable provided typical energies of quasiparticles are larger than the bandwidth of the impurity induced bound states, $T > \sqrt{\hbar T_c/\tau}$.

We calculate current and charge densities assuming the clean limit, $T_c\tau \gg \hbar$, and neglect, where possible, the pairbreaking due to elastic scattering. We assume also the case of frequencies $\hbar\omega$ much smaller than typical electronic energies: temperature and the amplitude of the gap, Δ_0.

Consider a current flowing between the layers of a quasi-two-dimensional superconductor, $t_\perp \ll \Delta_0$. When the current exceeds its critical value it produces nonequilibrium perturbations the scale of which is determined by the interlayer coupling, and for small $t_\perp$, the value of the nonequilibrium potential μ is small as well. Then we may solve Eq. (1) perturbatively, considering $t_\perp$ and μ as small values. We calculate the off diagonal in layer number components of g^K in the linear approximation in $t_\perp$; these components are related to the current density in the direction perpendicular to the layers. The diagonal in layer numbers component of $\mathrm{Tr}g^K$ determines perturbations of the charge density; we calculate it up to the second order in $t_\perp$ neglecting μ in comparison to Δ. The structure of the solution can be demonstrated by the combination $g_j = A_{nn-1}g^K_{n-1n} - g^K_{nn-1}A_{n-1n}$ the integral of which over energies and ϕ yields the current density across the layers [10]. In the Fourier representation we get

$$g_j = -\,it_\perp \tanh\frac{\epsilon}{2T}\left(\frac{\Delta^2}{(\xi^R)^3} - \frac{\Delta^2}{(\xi^A)^3}\right)S_\omega - \sum_{\omega_1}\frac{dn_F}{d\epsilon}\,\frac{4it_\perp\theta(|\epsilon| - |\Delta|)}{a(\omega_1 + i\tilde{\nu})}$$
$$\times\left[\omega_1(a^2 s_{\omega_1}c_{\omega-\omega_1} - c_{\omega_1}s_{\omega-\omega_1}) + \sum_{\omega_2}\frac{\omega_2(\mu_{n-1} - \mu_n)_{\omega_2}}{\hbar(\omega_2 + i\tilde{\nu})\langle(\omega_2 + i\nu_b)/(\omega_2 + i\tilde{\nu})\rangle}(s_{\omega-\omega_1}s_{\omega_1-\omega_2} + c_{\omega-\omega_1}c_{\omega_1-\omega_2})\right], \tag{2}$$

where S, s, and c are Fourier components of $\sin\varphi_n$, $\sin\varphi_n/2$, and $\cos\varphi_n/2$, respectively, and $\varphi_n = \chi_n - \chi_{n-1}$ is the phase difference between the layers. The first term in (2) describes perturbations of the retarded and advanced propagators; it is related to the supercurrent, $\xi^{R(A)} = \pm\sqrt{(\epsilon \pm i0)^2 - |\Delta|^2}$. The last terms describe perturbations of the distribution function and contribute to the quasiparticle current, n_F is Fermi distribution function, and $a = \epsilon/\xi$, $\xi = \sqrt{\epsilon^2 - |\Delta|^2}$. Furthermore, $\tilde{\nu} = (1/\tau)[\langle a\rangle - (\Delta/\epsilon)\langle\Delta/\xi\rangle]$ is the effective momentum scattering rate for quasiparticles. Finally, $\nu_b = (1/\tau)[\langle a\rangle - a^{-1} - (\Delta/\epsilon)\langle\Delta/\xi\rangle]$ is the effective branch imbalance relaxation rate. The branch imbalance is related to the nonequilibrium potential μ [7,8,15]. In the case of isotropic pairing elastic scattering does not contribute to the relaxation of the branch imbalance, and the latter relaxes via energy scattering processes. We shall consider the opposite case of the order parameter close to d-wave symmetry, when the gap has nodes: $\langle\Delta(\phi)\rangle^2 \ll \langle\Delta(\phi)^2\rangle$. In this limit we may simplify the equations because both $\tilde{\nu}$ and ν_b do not depend on ϕ, $\tilde{\nu} = (1/\tau)\langle a\rangle$, and $\nu_b = (1/\tau)\langle\Delta^2(\phi)a/\epsilon^2\rangle$. Then the expression for the current density between layers n and $n-1$ acquires the form

$$j_\perp = j_c\sin\varphi_n + \frac{1}{ed}\int_{-\infty}^{t}\frac{dt_1}{\tau}\left[\left(\int_{-\infty}^{t_1}\frac{dt_2}{\tau}\,\hat{\sigma}_b(t,t_1,t_2)[\mu_{n-1}(t_2) - \mu_n(t_2)] + \hat{\sigma}(t,t_1)\frac{\hbar}{2}\frac{\partial\varphi_n}{\partial t_1}\right)\right.$$
$$\left.\times\cos\frac{\varphi_n(t) - \varphi_n(t_1)}{2} + \hat{\sigma}_i(t,t_1)\frac{\hbar}{2}\frac{\partial\varphi_n}{\partial t_1}\cos\frac{\varphi_n(t) + \varphi_n(t_1)}{2}\right], \tag{3}$$

where generalized conductivities are given by

$$\hat{\sigma}(t,t_1) = \sigma_{N\perp}\left\langle\frac{\epsilon}{\xi}\,e^{-\tilde{\nu}(t-t_1)}\right\rangle_\epsilon - \hat{\sigma}_i(t,t_1), \qquad \hat{\sigma}_i(t,t_1) = \sigma_{N\perp}\left\langle\frac{\Delta^2}{2\epsilon\xi}\,e^{-\tilde{\nu}(t-t_1)}\right\rangle_\epsilon, \tag{4}$$

$$\hat{\sigma}_b(t,t_1,t_2) = \sigma_{N\perp}\left\langle\frac{\xi}{\epsilon}\,e^{-\tilde{\nu}(t-t_1)}e^{-\nu_b(t_1-t_2)}\right\rangle_\epsilon. \tag{5}$$

Averaging over the angles and quasiparticle energies in (4) and (5) is performed according to

$$\langle \cdots \rangle_\epsilon = -\int_{-\infty}^{\infty} d\epsilon \langle \theta(|\epsilon| - |\Delta(\phi)|) \frac{dn_F}{d\epsilon} (\cdots) \rangle .$$

The first term in (3) describes the Josephson current, $j_c = \hbar c^2/(4\pi e \lambda_\perp^2 d)$ being the critical current, $\lambda_\perp$ is the penetration length for a superconducting current perpendicular to the layers. The quasiparticle contribution is given by the last terms containing retardation effects related to the momentum and branch imbalance relaxation. These effects correspond to factors $(\omega + i\nu)$ in (2). In the limit of $\Delta \to 0$ these terms reduce to the Ohmic current $\sigma_{N\perp} E/(1 + i\omega\tau)$, where $\sigma_{N\perp}$ is the static conductivity in the normal state. In the low frequency limit the quasiparticle contribution may be interpreted as the Ohmic and interference current (the last term), the Ohmic current consisting of two contributions, one of which is related to the branch imbalance and to its relaxation. When φ is a slowly varying function of time, $\omega \ll \tilde{\nu}$, the retardation effects can be neglected, and the terms depending on the phase difference acquire a simple form typical for the Josephson tunnel junctions:

$$j_\perp = j_c \sin\varphi_n + \frac{\hbar}{2ed}\left(\sigma \frac{\partial\varphi_n}{\partial t} + \sigma_i \frac{\partial \varphi_n}{\partial t} \cos\varphi_n\right), \tag{6}$$

where conductivities are given by (4) and (5) with the exponents integrated over time t_1, which results in factors $1/\tilde{\nu}$. Near T_c one gets $\sigma = \sigma_{N\perp}$. At low temperatures, $T \ll \Delta_0$, the quasiparticle conductivity is of the order of the normal state conductivity, because the decrease of the normal carrier density upon cooling is compensated by the decrease of the effective scattering rate of quasiparticles by the same factor $\propto T/\Delta_0$. For the simplest angular dependence of the gap parameter with the d-wave symmetry, $\Delta = \Delta_0 \cos 2\phi$, we find $\sigma = 3\sigma_{N\perp}/4$. For the interference term we get $\sigma_i = (\Delta/T)\sigma_{N\perp}$ at $T \gg \Delta$, and $\sigma_i \sim \sigma_{N\perp}/4$ at lower temperatures.

The value of μ in the current density (3) must be determined from the separate equation describing the branch imbalance dynamics. Such an equation is given by the Poisson's equation with the charge density calculated from the integral of $\mathrm{Tr} g_{nn}^K$ over energies. In the Fourier representation we get for the charge density ρ_n in the nth layer

$$-i\omega\rho_n = -i\omega\gamma \frac{\kappa^2}{4\pi e}\mu_n - \nabla(\sigma_{2\|}\nabla\mu_n/e + i\omega\sigma_{1\|}\mathbf{P}_n) + \frac{1}{ed^2}\sum_{\omega_1}(J_n - J_{n-1}), \tag{7}$$

where $J_n = \sum_{\omega_2} \sigma_{2\perp}(n;\omega,\omega_1,\omega_2)[\mu_n(\omega_2) - \mu_{n-1}(\omega_2)] - \sigma_{1\perp}(n;\omega_1,\omega) 2i\hbar\omega_1\varphi_n(\omega_1)$ describes the flow of quasiparticles between layers n and $n-1$ generating the branch imbalance. Note that $\sigma(n)$ in the equations for charge density depend on the branch imbalance scattering rate, they are different from the conductivities for current densities.

$$\gamma = 1 - \left\langle \frac{\omega a}{(\omega + i\nu_b)} \right\rangle_\epsilon, \qquad \sigma_{k\|} = \sigma_{N\|}\left\langle \frac{i a^{1-2k}\omega^k}{\tau(\omega + i\tilde{\nu})(\omega + i\nu_b)^k} \right\rangle_\epsilon, \tag{8}$$

$$\sigma_{1\perp}(n) = \sigma_{N\perp}\left\langle \frac{i a^{-1}\omega(c_{\omega_1}c_{\omega-\omega_1} - s_{\omega_1}s_{\omega-\omega_1})}{\tau(\omega_1 + i\tilde{\nu})(\omega + i\nu_b)} \right\rangle_\epsilon, \tag{9}$$

$$\sigma_{2\perp}(n) = \sigma_{N\perp}\left\langle \frac{i\omega\omega_2(a^{-3}c_{\omega_1-\omega_2}c_{\omega-\omega_1} - a^{-1}s_{\omega_1-\omega_2}s_{\omega-\omega_1})}{\tau(\omega_1 + i\tilde{\nu})(\omega + i\nu_b)(\omega_2 + i\nu_b)} \right\rangle_\epsilon . \tag{10}$$

From Eq. (7) one can see that the branch imbalance $\mu_n \neq 0$ is generated by a nonuniform quasiparticle current flow.

In the limit of small phase differences between the layers Eqs. (3)–(5) and (7)–(10) describe the linear response characterized by different conductivities for the response to the solenoidal and to the potential electric fields [16].

Using the definition of μ_n we get

$$eV_n - \frac{\hbar}{2}\frac{\partial\varphi_n}{\partial t} = \mu_n - \mu_{n-1}, \tag{11}$$

where $V_n \equiv \Phi_n - \Phi_{n-1}$ is the difference of electric potentials per a layer. Thus, the rhs of (11) describes violation of the Josephson relation.

We consider, first, the uniform case. In the case of spatially uniform current distribution both $\nabla \mathbf{P_n} = 0$ and $J_n = J_{n-1}$, and using in (7) $\rho = 0$ we find $\mu_n = 0$. Then, according to (11), the Josephson relation between the frequency and the electric potential difference per a layer, V_n, is satisfied. In the limit of small frequencies the current-phase relation (6) has the form similar to Josephson tunnel junctions, and the effects typical for Josephson junctions must be observed. However, the analogy is limited by the region of voltages and frequencies smaller than the effective momentum scattering rate of the quasiparticles, $\nu_{\mathrm{qp}} \approx \tilde{\nu}(\epsilon = T)$. At higher voltages the finite scattering time effects, which were neglected in (6), destroy the analogy.

In principle, for typical parameters of a superconductor both current biased and voltage biased regimes in the resistive state are possible. For simplicity we shall concentrate on the case of the voltage bias, which can be realized, for example, when a capacity is connected in parallel with the superconductor. Then we can easily find the time dependence of the phase difference using the Josephson relation, and calculate the current density from Eq. (3).

$$j_\perp = \sigma_{N\perp}\left\langle \frac{V}{2ad\tau(4\omega^2 + \tilde{\nu}^2)}\left[\tilde{\nu}(1 + a^2) + \frac{\Delta^2}{2\xi^2}\sqrt{4\omega^2 + \tilde{\nu}^2}\cos\omega t\right]\right\rangle_\varepsilon, \tag{12}$$

where $\hbar\omega = 2eV$ and we omitted the layer index in the uniform state. Equation (12) resembles the expression for the current density across a tunnel junction. It contains dc and ac components, the characteristic frequency and voltage of such a junction, $\hbar\omega_c \equiv 2eV_c$, are determined by the momentum scattering time: $V_c \sim (\Delta_0/T)^2(\hbar/e\tau)$ at $T \gg \Delta_0$, and $V_c \sim (T/\Delta_0)(\hbar/e\tau)$ at $T \ll \Delta_0$. At $eV \gg \hbar\nu_{\rm qp}$ the dc current decreases with voltage increasing. Note that at lower temperatures $eV_c \sim \hbar\nu_{\rm qp}$; thus, the I-V curve starts to decrease with voltage already at $V \sim V_c$. Though numerically the result depends on the details of the angle dependence of the order parameter, qualitatively it can be illustrated by the explicit calculations with $\Delta = \Delta_0\cos 2\phi$ at temperatures $T \ll \Delta_0$.

$$j_\perp = \frac{\sigma_{N\perp}}{8d}\begin{cases} 3V & \text{at } V \ll V_c\,, \\ (\pi\hbar T/e\Delta_0\tau)^2V^{-1} & \text{at } V \gg V_c\,. \end{cases} \tag{13}$$

The origin of the negative differential conductivity at high voltages can be interpreted as the decrease of the dissipation at frequencies higher than the quasiparticle scattering rate, $\omega \gg \nu_{\rm qp}$, or, equivalently, using the analogy to the negative differential conductivity in semiconductor superlattices [17] at voltages per period higher than both the width of the miniband and the momentum scattering rate. In the latter case the chemical potential in the adjacent layers is shifted by the value of the voltage exceeding the width of the band of the allowed electronic states; therefore, the electron's energy in one layer corresponds to the forbidden states in the neighboring layer. So, the effect must be present in the normal state of layered conductors as well (see also [18]).

The negative differential conductivity indicates to an instability of the uniform resistive state at voltages $eV > \hbar\nu_{\rm qp}$. The instability must result in a nonuniform current distribution in which, according to (7), $\mu \neq 0$ and the Josephson relation is violated. Nonuniform current distribution may be created also at lower voltages due to many other reasons, e.g., due to the Meisner effect, contacts, or nonuniformities of the material. Thus, the correction to the Josephson relation in (11) depends on experimental conditions. For illustration we estimate such a correction for an artificial but easily treatable model of the nonuniformity created by a layer dependent impurity scattering time, τ_n. We consider the case when variations of τ_n with n are small, and the nonequilibrium potential μ can be calculated perturbatively. Then using condition $\rho_n = 0$ we get for the case of low frequencies $\omega \ll \nu_{\rm qp}$

$$\mu_n = \left\langle \frac{t_\perp^2}{2\hbar a}\left(\frac{\dot{\varphi}_n}{\nu_{bn}(\tilde{\nu}_n + \tilde{\nu}_{n-1})} - \frac{\dot{\varphi}_{n+1}}{\nu_{bn}(\tilde{\nu}_n + \tilde{\nu}_{n+1})}\right)\right\rangle_\epsilon. \tag{14}$$

Estimating the integrals in (14) for $T \ll \Delta_0$ we get from (11) $[V_n - (\hbar/2e)\dot{\varphi}_n]/V_n \sim \hbar^{-2}t_\perp^2\tau_n(\tau_{n+1} - \tau_{n-1})(\Delta_0/T)\ln\Delta_0\tau$. One can see that the significant violation of the Josephson relation may be created even by small variations of τ in different layers.

Thus, generally speaking, the Josephson relation in layered superconductors with Josephson interlayer coupling and anisotropic pairing is satisfied under the special conditions of the uniform current distribution only, which is difficult to satisfy. Even in the ideally uniform samples the uniform state is expected to be unstable at higher voltages because of the negative differential conductivity. This may be the origin of the irregular character of the I-V curves observed usually in the measurements of the intrinsic Josephson effect in high-T_c materials.

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