

Non-Equilibrium Josephson and Andreev Current through Interacting Quantum Dots

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We present a theory of transport through interacting quantum dots coupled to normal and superconducting leads in the limit of weak tunnel coupling. A Josephson current between two superconducting leads, carried by first-order tunnel processes, can be established by non-equilibrium proximity effect. Both Andreev and Josephson current is suppressed for bias voltages below a threshold set by the Coulomb charging energy. A π -transition of the supercurrent can be driven by tuning gate or bias voltages.

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Non-equilibrium transport through superconducting systems attracted much interest since the demonstration of a Superconductor-Normal-Superconductor (SNS) transistor [1]. In such a device, supercurrent suppression and its sign reversal (π -transition) are achieved by driving the quasi-particle distribution out of equilibrium by means of applied voltages [2, 3, 4, 5]. Another interesting issue in mesoscopic physics is transport through quantum dots attached to superconducting leads. For DC transport through quantum dots coupled to a normal and a superconducting lead, subgap transport is due to Andreev reflection [6, 7, 8, 9, 10, 11]. Also transport between two superconductors through a quantum dot has been studied extensively. The limit a non-interacting dot has been investigated in Ref. 12. Several authors considered the regime of weak tunnel coupling where the electrons forming a Cooper pair tunnel one by one via virtual states [13, 14, 15, 16]. Multiple Andreev reflection through localized levels was investigated in Refs. 17, 18. Numerical approaches based on the non-crossing approximation [19], the numerical renormalization group [20] and Monte Carlo [21] have also been used. The authors of Ref. 22 compare different approximation schemes, such as mean field and second-order perturbation in the Coulomb interaction. Experimentally, the supercurrent through a quantum dot has been measured through dots realized in carbon nanotubes [23] and in indium arsenide nanowires [24].

In this Letter we study the transport properties of a system composed of an interacting single-level quantum dot between two equilibrium superconductors where a third, normal lead is used to drive the dot out of equilibrium. We relate the subgap current flowing into the superconductors to the pair amplitude of the dot $\langle d_{\downarrow} d_{\uparrow} \rangle$, induced by proximity effect. The latter is calculated by means of a kinetic equation derived from a real-time diagrammatic technique. In particular, we identify nonequilibrium situations in which a Josephson current carried by first-order tunnel processes is established.

Model.—We consider a single-level quantum dot connected to two superconducting and one normal lead

via tunnel junctions, see Fig. 1. The total Hamilto-

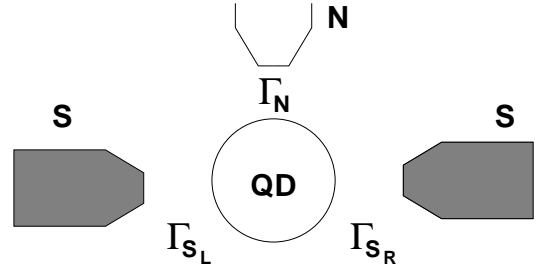


FIG. 1: Setup: a single-level quantum dot is connected by tunnel junctions to one normal and two superconducting leads with tunneling rates Γ_N and $\Gamma_{S,L,R}$, respectively.

nian is given by $H = H_D + \sum_{\eta=N,S_L,S_R} (H_{\eta} + H_{\text{tunn},\eta})$. The quantum dot is described by the Anderson model $H_D = \sum_{\sigma} \epsilon d_{\sigma}^{\dagger} d_{\sigma} + U n_{\uparrow} n_{\downarrow}$, where $n_{\sigma} = d_{\sigma}^{\dagger} d_{\sigma}$ is the number operator for spin $\sigma = \uparrow, \downarrow$, ϵ is the energy level, and U is the charging energy for double occupation. The leads, labeled by $\eta = N, S_L, S_R$, are modeled by $H_{\eta} = \sum_{k\sigma} \epsilon_k c_{\eta k\sigma}^{\dagger} c_{\eta k\sigma} - \sum_k \left(\Delta_{\eta} c_{\eta k\uparrow}^{\dagger} c_{\eta -k\downarrow}^{\dagger} + \text{H.c.} \right)$, where Δ_{η} is the superconducting order parameter ($\Delta_N = 0$). The tunneling Hamiltonians are $H_{\text{tunn},\eta} = V_{\eta} \sum_{k\sigma} \left(c_{\eta k\sigma}^{\dagger} d_{\sigma} + \text{H.c.} \right)$. Here, V_{η} are the spin- and wavevector-independent tunnel matrix elements, and $c_{\eta k\sigma} (c_{\eta k\sigma}^{\dagger})$ and $d_{\sigma} (d_{\sigma}^{\dagger})$ represent the annihilation (creation) operators for the leads and dot, respectively. The tunnel-coupling strengths are characterized by $\Gamma_{\eta} = 2\pi |V_{\eta}|^2 \sum_k \delta(\omega - \epsilon_k)$.

Current formula.— We start with deriving a general formula for the charge current in lead η by generalizing to superconducting leads the approach of Ref. [25]. For this, it is convenient to use the operators $\psi_{\eta k} = (c_{\eta k\uparrow}, c_{\eta -k\downarrow}^{\dagger})^T$ and $\phi = (d_{\uparrow}, d_{\downarrow}^{\dagger})^T$ in Nambu formalism. The current from lead η is expressed as $J_{\eta} = e \langle dN_{\eta}/dt \rangle = i(e/\hbar) \langle [H, N_{\eta}] \rangle = i(e/\hbar) \langle [H_{\text{tunn},\eta}, N_{\eta}] \rangle$ [26], with $N_{\eta} = \sum_k \psi_{\eta k}^{\dagger} \tau_3 \psi_{\eta k}$, where τ_1, τ_2, τ_3 indicate the Pauli matrices

in Nambu space. Evaluating the commutator leads to

$$J_\eta = -\frac{2e}{\hbar} \sum_k \int \frac{d\omega}{2\pi} \text{Re} \left\{ \text{Tr} \left[\tau_3 \mathbf{V}_\eta \mathbf{G}^{<}_{\text{D},\eta k}(\omega) \right] \right\}, \quad (1)$$

with $\mathbf{V}_\eta = \text{Diag}(V_\eta, -V_\eta^*)$ and the lead-dot lesser Green's functions $(\mathbf{G}^{<}_{\text{D},\eta k}(\omega))_{m,n}$ that are the Fourier transforms of $i\langle \psi_{\eta kn}^\dagger(0)\phi_m(t) \rangle$. In the following, we assume the tunneling matrix elements V_η to be real (any phase of V_η can be gauged away by substituting $\Delta_\eta \rightarrow \Delta_\eta \exp(-2i \arg V_\eta)$). The Green's function $\mathbf{G}^{<}_{\text{D},\eta k}$ is related to the full dot Green's functions and the lead Green's functions by a Dyson equation in Keldysh formalism [27]. Using this relation and assuming energy-independent tunnel rates Γ_η , we obtain for the current $J_\eta = J_{1\eta} + J_{2\eta}$ with

$$J_{1\eta} = \frac{e}{\hbar} \int \frac{d\omega}{2\pi} \Gamma_\eta D_\eta(\omega) \text{Im} \left\{ \text{Tr} \left[\tau_3 \left[\mathbf{1} - \frac{\Delta_\eta}{\omega - \mu_\eta} \right] (2\mathbf{G}^{\text{R}}(\omega) f_\eta(\omega) + \mathbf{G}^{<}(\omega)) \right] \right\}, \quad (2)$$

$$J_{2\eta} = \frac{e}{\hbar} \int \frac{d\omega}{2\pi} \Gamma_\eta S_\eta(\omega) \text{Re} \left\{ \text{Tr} \left[\tau_3 \frac{\Delta_\eta}{|\Delta_\eta|} \mathbf{G}^{<}(\omega) \right] \right\}, \quad (3)$$

where $\Delta_\eta = \begin{pmatrix} 0 & \Delta_\eta \\ \Delta_\eta^* & 0 \end{pmatrix}$, and $f_\eta(\omega) = [1 + \exp(\omega - \mu_\eta)/(k_B T)]^{-1}$ is the Fermi function, with T being the temperature and k_B the Boltzmann constant. The dot Green's functions $(\mathbf{G}^{<}_{\text{D}}(\omega))_{m,n}$ and $(\mathbf{G}^{\text{R}}_{\text{D}}(\omega))_{m,n}$ are defined as the Fourier transforms of $i\langle \phi_m^\dagger(0)\phi_n(t) \rangle$ and $-i\theta(t)\langle \{\phi_m(t), \phi_n^\dagger(0)\} \rangle$, respectively. The two weighting functions $D_\eta(\omega)$ and $S_\eta(\omega)$ are given by

$$D_\eta(\omega) = \frac{|\omega - \mu_\eta|}{\sqrt{(\omega - \mu_\eta)^2 - |\Delta_\eta|^2}} \theta(|\omega - \mu_\eta| - |\Delta_\eta|)$$

$$S_\eta(\omega) = \frac{|\Delta_\eta|}{\sqrt{|\Delta_\eta|^2 - (\omega - \mu_\eta)^2}} \theta(|\Delta_\eta| - |\omega - \mu_\eta|).$$

The term $J_{1\eta}$ involves energies above the gap. For $\eta = \text{N}$, this is the only contribution, and the current reduces to the result presented in Ref. 25. For superconducting leads, $J_{1\eta}$ describes quasiparticle transport, but also includes anomalous components of the Green's functions. In the limit of large superconducting gap, only sub-gap contributions to the current, $J_{2\eta}$, describing Josephson as well as Andreev tunneling remain. In the following we consider the large-superconducting-gap limit ($|\Delta_\eta| \rightarrow \infty$), where the current simplifies to

$$J_\eta = \frac{2e}{\hbar} \Gamma_\eta |\langle d_\downarrow d_\uparrow \rangle| \sin(\Psi - \Phi_\eta), \quad (4)$$

with Φ_η being the phase of Δ_η and $\langle d_\downarrow d_\uparrow \rangle = |\langle d_\downarrow d_\uparrow \rangle| \exp(i\Psi)$ the pair amplitude of the dot that has to be determined in the presence of Coulomb interaction, coupling to all (normal and superconducting) leads and in non-equilibrium due to finite bias voltage.

We now focus on a symmetric three-terminal setup with $\Gamma_{\text{S}_\text{L}} = \Gamma_{\text{S}_\text{R}} = \Gamma_{\text{S}}$, $\Delta_{\text{S}_\text{L}} = |\Delta| \exp(i\Phi/2)$ and $\Delta_{\text{S}_\text{R}} = |\Delta| \exp(-i\Phi/2)$, and $\mu_{\text{S}_\text{L}} = \mu_{\text{S}_\text{R}} = 0$. The quantities of interest are the the current that flows between the two superconductors (Josephson current) $J_{\text{Jos}} = (J_{\text{S}_\text{L}} - J_{\text{S}_\text{R}})/2$ and the current in the normal lead (Andreev current) $J_{\text{and}} = J_{\text{N}} = -(J_{\text{S}_\text{L}} + J_{\text{S}_\text{R}})$.

To proximize the quantum dot, the dot states for empty and double occupation should be energetically almost degenerate. Furthermore, in *equilibrium* situations, temperature must be low enough, $k_B T < \Gamma_{\text{S}}$, to resolve the influence of the superconductors on the quantum-dot spectrum [28]. In this paper, however, we focus on the opposite limit, $k_B T > \Gamma_{\text{S}}$, in which proximity is due to *non-equilibrium* occupation of the dot only.

Kinetic equations for quantum-dot degrees of freedom.—The Hilbert space of the dot is four dimensional: the dot can be empty, singly occupied with spin up or down, or doubly occupied, denoted by $|\chi\rangle \in \{|0\rangle, |\uparrow\rangle, |\downarrow\rangle, |D\rangle \equiv d_\downarrow^\dagger d_\uparrow^\dagger |0\rangle\}$, with energies $E_0, E_\uparrow = E_\downarrow, E_D$. For convenience we define the detuning as $\delta = E_D - E_0 = 2\epsilon + U$. The dot dynamics is fully described by its reduced density matrix ρ_{D} , with matrix elements $P_{\chi_2 \chi_1}^{\chi_1} \equiv (\rho_{\text{D}})_{\chi_2 \chi_1}$. The dot pair amplitude $\langle d_\downarrow d_\uparrow \rangle$ is given by the off-diagonal matrix element P_{D}^0 . The time evolution of the reduced density matrix is described by the kinetic equations

$$\frac{d}{dt} P_{\chi_2}^{\chi_1}(t) + \frac{i}{\hbar} (E_{\chi_1} - E_{\chi_2}) P_{\chi_2}^{\chi_1}(t) = \sum_{\chi'_1, \chi'_2} \int_{t_0}^t dt' W_{\chi_2 \chi'_2}^{\chi_1 \chi'_1}(t, t') P_{\chi'_2}^{\chi'_1}(t'). \quad (5)$$

We define the generalized transition rates by $W_{\chi_2 \chi'_2}^{\chi_1 \chi'_1} \equiv \int_{-\infty}^t dt' W_{\chi_2 \chi'_2}^{\chi_1 \chi'_1}(t, t')$, which are the only quantities to be evaluated in the stationary limit. Together with the normalization condition $\sum_\chi P_\chi = 1$, Eq. (5) determines the matrix elements of ρ_{D} . Furthermore, in Eq. (5) we retain only linear terms in the tunnel strengths Γ_η and the detuning δ . Hence, we calculate the rates $W_{\chi_2 \chi'_2}^{\chi_1 \chi'_1}$ to the lowest (first) order in Γ_η for $\delta = 0$. This is justified in the transport regime $\Gamma_{\text{S}}, \Gamma_{\text{N}}, \delta < k_B T$.

The rates are evaluated by means of a real-time diagrammatic technique [29], that we generalize to include superconducting leads. We find for the (first-order) diagonal rates $W_{\chi_1 \chi_2}^{\chi_1 \chi_2} \equiv W_{\chi_1 \chi_2}^{\chi_1 \chi_2}$ the expressions $W_{\sigma 0} = \Gamma_{\text{N}} f_{\text{N}}(-U/2)$; $W_{0\sigma} = \Gamma_{\text{N}} [1 - f_{\text{N}}(-U/2)]$; $W_{D\sigma} = \Gamma_{\text{N}} f_{\text{N}}(U/2)$; $W_{\sigma D} = \Gamma_{\text{N}} [1 - f_{\text{N}}(U/2)]$. The N lead also contributes to the rates $W_{00}^{DD} = -\Gamma_{\text{N}} [1 + f_{\text{N}}(-U/2) - f_{\text{N}}(U/2) + iB]$ and $W_{DD}^{00} = -\Gamma_{\text{N}} [1 - f_{\text{N}}(-U/2) + f_{\text{N}}(U/2) - iB]$ where $B = \frac{1}{\pi} \text{Re} \left[\psi \left(\frac{1}{2} + i \frac{U/2 - \mu_{\text{N}}}{2\pi T} \right) - \psi \left(\frac{1}{2} + i \frac{-U/2 - \mu_{\text{N}}}{2\pi T} \right) \right]$, with μ_{N} being the chemical potential of the normal lead and $\psi(z)$ the Digamma function. Notice that B vanishes when $\mu_{\text{N}} = 0$ or $U = 0$. The superconducting leads do not

enter here due to the gap in the quasi-particle density of states. These leads, though, contribute to the off-diagonal rates $W_{0D}^{00} = W_{0D}^{DD} = \alpha(\Phi)$; $W_{00}^{0D} = W_{DD}^{0D} = \alpha^*(\Phi)$; $W_{00}^{D0} = W_{DD}^{D0} = -\alpha(\Phi)$; $W_{D0}^{00} = W_{D0}^{DD} = -\alpha^*(\Phi)$, where $\alpha(\Phi) = 2i\Gamma_S \cos(\Phi/2)$.

For an intuitive representation of the system dynamics we define, in analogy to Ref. [30], a dot isospin by

$$I_x = \frac{P_0^D + P_D^0}{2}; \quad I_y = i\frac{P_0^D - P_D^0}{2}; \quad I_z = \frac{P_D - P_0}{2}. \quad (6)$$

From Eq. (5), we find that in the stationary limit the isospin dynamics can be separated into three parts, $0 = d\mathbf{I}/dt = (d\mathbf{I}/dt)_{\text{acc}} + (d\mathbf{I}/dt)_{\text{rel}} + (d\mathbf{I}/dt)_{\text{rot}}$, with

$$\hbar \left(\frac{d\mathbf{I}}{dt} \right)_{\text{acc}} = -\frac{\Gamma_N}{2} [1 - f_N(-U/2) - f_N(U/2)] \hat{\mathbf{e}}_z \quad (7)$$

$$\hbar \left(\frac{d\mathbf{I}}{dt} \right)_{\text{rel}} = -\Gamma_N [1 + f_N(-U/2) - f_N(U/2)] \mathbf{I} \quad (8)$$

$$\hbar \left(\frac{d\mathbf{I}}{dt} \right)_{\text{rot}} = \mathbf{I} \times \mathbf{B}_{\text{eff}} \quad (9)$$

where $\hat{\mathbf{e}}_z$ is the z -direction and $\mathbf{B}_{\text{eff}} = \{2\Gamma_S \cos(\Phi/2), 0, -\Gamma_N B - 2\epsilon - U\}$ is an effective magnetic field in the isospin space. The accumulation term Eq. (7) builds up a finite isospin, while the relaxation term Eq. (8) decreases it. Finally, Eq. (9) describes a rotation of the isospin direction.

Non-equilibrium Josephson current.— In the isospin language the current in the superconducting leads is

$$J_{S,L,R} = \frac{2e}{\hbar} \Gamma_S [I_y \cos(\Phi/2) \pm I_x \sin(\Phi/2)], \quad (10)$$

where the upper(lower) sign refers to the left(right) superconducting lead. The I_y component contributes to the Andreev current, while I_x is responsible for the Josephson current. By inspection of Eqs. (7)-(9), it is apparent that for a finite Josephson current J_{jos} , we need that the z -component, $-\Gamma_N B - 2\epsilon - U$, of the effective magnetic field acting on the isospin is non zero. The Josephson current and the Andreev current read

$$J_{\text{jos}} = -\frac{e\Gamma_S}{\hbar} \frac{[2\epsilon + U + \Gamma_N B]\Gamma_S \sin(\Phi)}{|\mathbf{B}_{\text{eff}}|^2 + \Gamma_N^2 [1 + f_N(-U/2) - f_N(U/2)]^2} \times \frac{1 - f_N(-U/2) - f_N(U/2)}{1 + f_N(-U/2) - f_N(U/2)} \quad (11)$$

$$J_{\text{and}} = -\frac{e\Gamma_S}{\hbar} \frac{2\Gamma_N \Gamma_S [1 + \cos(\Phi)]}{|\mathbf{B}_{\text{eff}}|^2 + \Gamma_N^2 [1 + f_N(-U/2) - f_N(U/2)]^2} \times [1 - f_N(-U/2) - f_N(U/2)]. \quad (12)$$

These results take into account only first-order tunnel processes, i.e. the rates $W_{\chi_1 \chi_2}^{\chi_1 \chi_1}$ are computed to first order in Γ_η . The factor $[1 - f_N(-U/2) - f_N(U/2)]$ ensures that no finite dot-pair amplitude can be established if the chemical potential of the normal lead, μ_N , is inside the interval $[-U/2, U/2]$ by at least $k_B T$. In this situation both

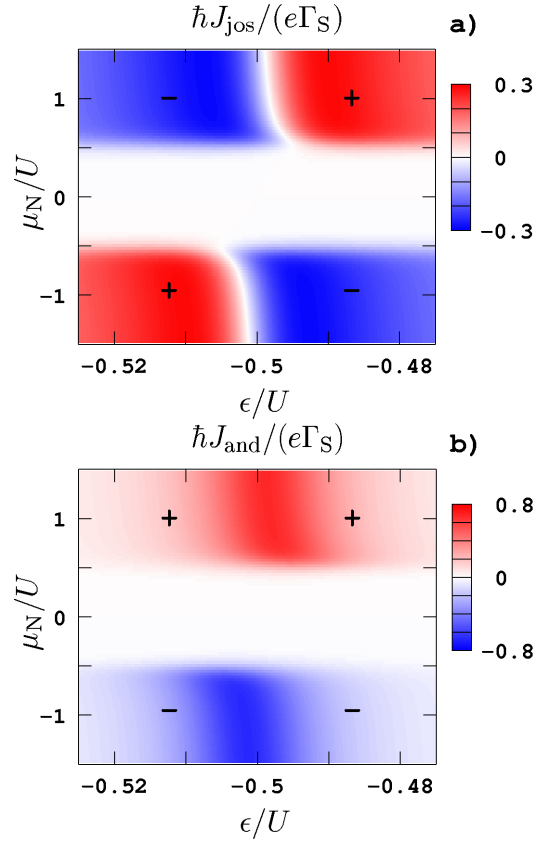


FIG. 2: (color online) Density plot of the a) Josephson and b) Andreev current, for fixed superconducting-phase difference $\Phi = \pi/2$, as a function of the dot-level position ϵ and of the chemical potential of the normal lead μ_N . The symbols \pm refer to the sign of the current. The other parameters are $\Gamma_S = \Gamma_N = 0.01U$, and $k_B T = 0.05U$.

the Josephson and the Andreev currents vanish. On the other hand, this factor takes the value -1 if $\mu_N > U/2$ and the value $+1$ if $\mu_N < -U/2$. Hence, the sign of the Josephson current can be reversed by the applied voltage (voltage driven π -transition). The considerations above establish the importance of the non-equilibrium voltage to induce and control proximity effect in the interacting quantum dot. In Fig. 2 we show in a density plot of (a) J_{jos} and (b) J_{and} for $\Phi = \pi/2$ as a function of the voltage μ_N and the level position ϵ . Both the control of proximity effect by the chemical potential μ_N and the voltage driven π -transition are clearly visible. If the detuning is too large, $|\delta + \Gamma_N B| > \sqrt{\Gamma_N^2 + 4\Gamma_S^2 \cos^2(\Phi/2)}$, it becomes difficult to build a superposition of the states $|0\rangle$ and $|D\rangle$, which is necessary to establish proximity. As a consequence, the Josephson and the Andreev current are algebraically suppressed by δ^{-1} and δ^{-2} , respectively. Fig. 3 shows the Josephson current as a function of $\delta = 2\epsilon + U$. The fact that the Josephson current is non zero for $\delta = 0$ is due to the term $\Gamma_N B$, i.e. of the interaction induced contribution to the z -component of

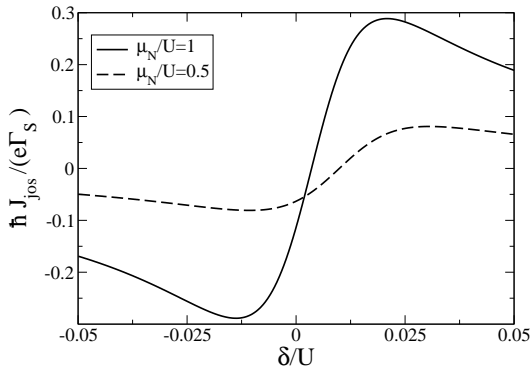


FIG. 3: Josephson current, for fixed superconducting-phase difference $\Phi = \pi/2$, as a function of the detuning $\delta = E_D - E_0 = 2\epsilon + U$ for different values of the chemical potential. The other parameters are $\Gamma_S = \Gamma_N = 0.01U$ and $k_B T = 0.05U$.

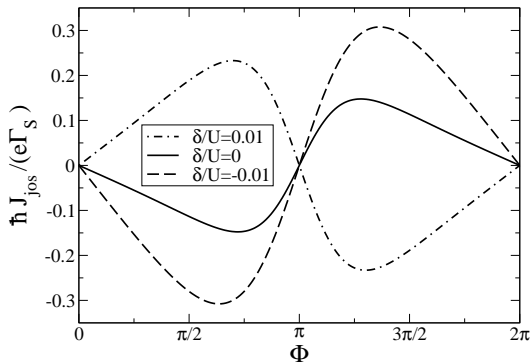


FIG. 4: Josephson current as a function of the superconducting-phase difference Φ for different values of the detuning. The other parameters are $\Gamma_S = \Gamma_N = 0.01U$, $\mu_N = U$, and $k_B T = 0.05U$.

the effective field \mathbf{B}_{eff} acting on the isospin. The term $|B|$ has a maximum at $\mu_N = U/2$, which causes this effect to be more pronounced at the onset of transport. The fact that the value of the Josephson current varies on a scale smaller than temperature indicates its nonequilibrium nature.

A π -transition of the Josephson current can also be achieved by changing the sign of $\delta + \Gamma_N B$, as shown in Fig. 4 where J_{jos} is plotted as a function of the phase difference Φ for different values of the level position. Notice that the current for $\delta = 0$ ($\epsilon = -U/2$) is different from zero only due to the presence of the term $\Gamma_N B$ acting on the isospin.

Conclusion.— We have studied non-equilibrium proximity effect in an interacting single-level quantum dot weakly coupled to two superconducting and one normal leads. By applying a bias voltage between normal and superconducting leads, a Josephson current carried by first-order tunneling processes, is established. A π -transition can be driven either by bias or gate voltage. The charging energy of the quantum dot defines a threshold bias

voltage below which proximity is suppressed.

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- [28] This can, e.g., be seen in the exactly-solvable limit of $U = 0$ together with $\Gamma_N = 0$, where the Josephson current is $J_{\text{jos}} = -(e/2\hbar) \Gamma_S^2 \sin(\Phi) [f(-\epsilon_A(\Phi)) - f(\epsilon_A(\Phi))] / \epsilon_A(\Phi)$ with $\epsilon_A(\Phi) = \sqrt{\epsilon^2 + \Gamma_S^2 \cos^2(\Phi/2)}$.
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