

All-electrical generation and control of odd-frequency s -wave Cooper pairs in double quantum dots

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We propose an all-electrical experimental setup to detect and manipulate the amplitude of odd-frequency pairing in a double quantum dot. The odd-frequency pair amplitude is induced from the breakdown of orbital symmetry when Cooper pairs are injected in the double dot with electrons in different dots. When the dot levels are aligned with the Fermi energy, i.e., on resonance, nonlocal Andreev processes are directly connected to the presence of odd-frequency pairing. Therefore, their amplitude can be manipulated by tuning the level positions. The detection of nonlocal Andreev processes by conductance measurements contributes a direct proof of the existence of the odd-frequency pair amplitude and is available using current experimental techniques.

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Introduction. The symmetry analysis of Cooper pairs is a key element in the study of superconductivity. For example, Cooper pairs at conventional BCS superconductors form a spin-singlet even-parity state, where the electrons have opposite spins and are coupled in momentum space by the isotropic s -wave channel. A current trend in the study of superconductivity is to *engineer* unconventional superconductors by breaking down symmetries of a BCS superconductor. Consequently, a new type of pairing emerges which is odd in frequency, i.e., odd under an exchange of the time coordinates [1–4]. Plenty of theoretical studies suggest the ubiquitous presence of odd-frequency superconductivity in inhomogeneous superconducting systems [5–17]. Unfortunately, experimental evidence for the odd-frequency pair amplitude is very limited. Odd-frequency spin-triplet s -wave superconductivity can explain the long-range proximity effect [18,19], the intrinsic paramagnetic Meissner effect [20–23], and the subgap structure [24] observed in ferromagnet-superconductor hybrids. However, in those experiments, odd-frequency pairs are mixed with conventional ones and their amplitude is not tunable but accidentally determined by the configuration of magnetic moments realized at the junction. To unambiguously establish the presence of odd-frequency pairing, proposals that filter odd-frequency pairs and allow one to control their amplitude are required.

In this Rapid Communication, based on recent experimental progress on double quantum dot (DQD) Cooper pair splitter devices [25,26], we propose a setup that allows for the detection and manipulation of odd-frequency pairing without using any magnetic elements. Such pair splitters consist of a DQD independently connected to two normal leads and one superconducting electrode, as sketched in Fig. 1(a). We show that the symmetry of the induced pairing from the superconducting lead is broken due to the DQD orbital degree of freedom, thus becoming a superposition of symmetric and antisymmetric orbital states. For a spin-singlet superconductor, induced Cooper pairs that are antisymmetric (symmetric) in DQD space must be odd (even) in frequency, according to Fermi-Dirac statistics, since parity and spin rotation symmetries are not broken [27]. Cooper pairs transmitted to the same lead are in an even-frequency state [see Fig. 1(b)]. On the other hand, when the levels are on resonance, only an odd-frequency pair amplitude is responsible for the splitting of Cooper pairs into

different leads [see Fig. 1(c)]. Antisymmetric odd-frequency singlet pairing is greatly enhanced if each dot is coupled differently to the leads, resulting in a measurable contribution to the conductance. Such a connection between symmetry and microscopic transport processes is a unique feature of our proposal. Additionally, the amplitude of odd-frequency Cooper pairs can be controlled by tuning the DQD level positions on or off resonance. Alternatively, using a spin-triplet superconductor, the same geometry can be used for the study of Majorana edge states [28]. In such a case, odd-frequency triplet pairs are now transmitted to the same electrode. Our proposal opens a different direction in the study of exotic Cooper pairing owing to the unique connection between the symmetry of the Cooper pair and tunneling processes and due to the tunability of the pair amplitude.

Model. DQD-based three-terminal devices [29–32] are an ideal platform for exploring the symmetry of induced pairing. Recent experiments are very well modeled by two-level systems and show an exquisite degree of tunability [25,26,33–35]. Moreover, strong evidence for the splitting of Cooper pairs [36–39], which we shall link to the presence of odd-frequency spin-singlet s -wave pairing, has been presented. Here, we consider a system with two quantum levels at positions $\epsilon_{L,R}$. In the limit of large level separation at the quantum dots, it describes very well a DQD close to the crossing point of the dot resonances [40]. In the absence of external magnetic fields and spin-orbit coupling terms, we describe the system in the combined Nambu-dot space using spinor fields $\Psi = (d_{L\uparrow}, d_{R\uparrow}, d_{L\downarrow}^\dagger, d_{R\downarrow}^\dagger)^T$, where $d_{\mu\sigma}$ ($d_{\mu\sigma}^\dagger$) annihilates (creates) an electron with spin $\sigma = \uparrow, \downarrow$ at dot $\mu = L, R$. In the following, $\hat{\sigma}_\nu$ ($\hat{\tau}_\nu$), with $\nu = 0, 1, 2, 3$, are Pauli matrices in dot (Nambu) space, with identity matrix $\hat{\sigma}_0$ ($\hat{\tau}_0$). The Hamiltonian of the isolated DQD is given by $\hat{H}_d = (\epsilon_L \hat{\sigma}_+ + \epsilon_R \hat{\sigma}_- + \Gamma_{LR} \hat{\sigma}_1) \hat{\tau}_3$, with $\hat{\sigma}_\pm = (\hat{\sigma}_0 \pm \hat{\sigma}_3)/2$ and the interdot tunneling rate $\Gamma_{LR} > 0$. Transport properties are characterized by the Green's function

$$\check{g}(\omega) = [\omega \hat{\sigma}_0 \hat{\tau}_0 - \hat{H}_d - \check{\Sigma}_N(\omega) - \check{\Sigma}_S(\omega)]^{-1}, \quad (1)$$

where ω denotes $\omega \pm i0^+$ or $i\omega_n$ for retarded/advanced or Matsubara Green's function, respectively, with $\omega_n = \pi(2n+1)k_B T$ for temperature T , Boltzmann constant k_B , and integer n .

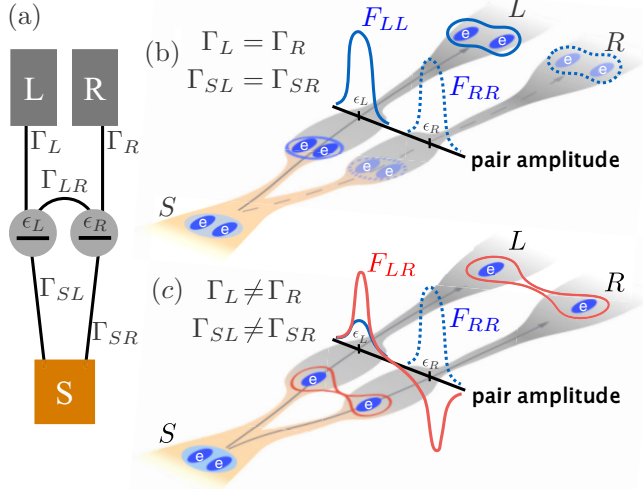


FIG. 1. Proximity-induced superconductivity in a DQD three-terminal device. (a) Schematics of a DQD with level positions ϵ_L and ϵ_R contacted by a superconducting lead S and two normal leads L and R. (b) In a local Andreev process, the electrons of a Cooper pair tunnel through one dot into the normal lead. The amplitude for these processes, $F_{LL,RR}$ (blue dashed lines), is enhanced for symmetric devices, i.e., those where each dot is similarly coupled to the leads. (c) In a nonlocal Andreev process, each electron of the Cooper pair tunnels to a different lead. The nonlocal amplitude, F_{LR} (red solid line), is enhanced in asymmetric devices. On resonance ($\epsilon_L = \epsilon_R = 0$), F_{LR} is odd in frequency if S is a BCS superconductor.

Following the geometry described in Fig. 1(a), we include the normal and superconducting leads as self-energies,

$$\check{\Sigma}_N(\omega) = is(\omega)(\Gamma_L \hat{\sigma}_+ + \Gamma_R \hat{\sigma}_-) \hat{\tau}_0, \quad (2)$$

$$\check{\Sigma}_S(\omega) = i(\Gamma_{SL} \hat{\sigma}_+ + \Gamma_{SR} \hat{\sigma}_-) [g(\omega) \hat{\tau}_0 - f(\omega) \hat{\tau}_1], \quad (3)$$

with $s(\omega \pm i0^+) = \mp 1$, $s(i\omega_n) = \text{sgn}(\omega_n)$, and $\Gamma_\mu, \Gamma_{S\mu} > 0$ the tunneling rates between dots and leads. We consider the regime where Kondo and exchange interactions between dots can be neglected. The effect of Coulomb repulsion on each dot is to renormalize the level positions ϵ_μ and tunneling rates $\Gamma_{S\mu}$ [40–42]. We assume that the superconducting region is well described by a constant pair potential Δ and neglect its spatial dependence at the surface of the superconductor. The dimensionless Green's functions at the edge of the superconducting lead, for a BCS superconductor, are $f(\omega) = -(\Delta/\omega)g(\omega) = -\Delta/\sqrt{\omega^2 - \Delta^2}$.

Symmetry of induced pair amplitude. The uncoupled superconducting lead in Eq. (3) represents an even-frequency spin-singlet s -wave superconductor which satisfies $f(\omega_n) = f(-\omega_n)$, for Matsubara frequency. We analyze the symmetry of the proximity-induced pair amplitude in the DQD system from the anomalous part of the Green's function of Eq. (1), $F_{\mu\nu} = (\check{g})_{\mu\nu}^{eh} \sim \langle d_{\mu\uparrow} d_{\nu\downarrow} \rangle$, with indexes in dot space $\mu, \nu = L, R$. Induced superconductivity in the DQD system thus acquires an extra orbital quantum number. Owing to this symmetry, the elements of $F_{\mu\nu}$ are divided into even- and odd-orbital terms. Defining $F_{s,a} = (F_{LR} \pm F_{RL})/2$, F_a is the only element with odd parity in the dot orbital degree of

TABLE I. Symmetry classification of Cooper pairs according to frequency/spin/momentum/dot. From left to right, pairs can be even (E, +) or odd (O, -) under time-reversal (frequency), spin ($\sigma, \sigma' = \uparrow, \downarrow$), momentum (\mathbf{k}), and dot index (L, R), with S for spin singlet (top rows) and T for triplet (bottom rows). The last column shows the corresponding element of the anomalous Green's function: $F_{\mu\mu}$, with $\mu = L, R$, for local elements and $F_{s,a}$ for the nonlocal ones.

Class	$\omega_n \rightarrow -\omega_n$	$\sigma \leftrightarrow \sigma'$	$\mathbf{k} \rightarrow -\mathbf{k}$	$L \leftrightarrow R$	Element
ESEE	+	-	+	+	$F_{\mu\mu}, F_s$
OSEO	-	-	+	-	F_a
ETEO	+	+	+	-	F_a
OTEE	-	+	+	+	$F_{\mu\mu}, F_s$

freedom. To be consistent with Fermi-Dirac statistics, F_a must be odd in frequency. Explicitly, we find [43]

$$F_s(\omega_n) = -i \frac{f(\omega_n)}{D(\omega_n)} \Gamma_{LR} (\Gamma_{SR} \epsilon_L + \Gamma_{SL} \epsilon_R), \quad (4)$$

$$F_a(\omega_n) = \text{sgn}(\omega_n) \frac{f(\omega_n)}{D(\omega_n)} \Gamma_{LR} (\Gamma_{SR} \omega_L - \Gamma_{SL} \omega_R), \quad (5)$$

with $\omega_\mu = |\omega_n| - \Gamma_\mu$ and $D(\omega_n) = \det[\check{g}^{-1}(\omega_n)] = D(-\omega_n)$. If $f(\omega_n) = f(-\omega_n)$ is satisfied, we find that $F_s(\omega_n) = F_s(-\omega_n)$ and $F_a(\omega_n) = -F_a(-\omega_n)$. A complete description of the allowed symmetries in the DQD system is given in Table I, for both spin-singlet and triplet superconductors.

It is possible to enhance odd-frequency over even-frequency pairing on one of the dots, as sketched in Fig. 1(c). To study this effect, we define the local ratios [44]

$$R_{L,R}(\omega) = \frac{|F_a(\omega)|}{\sqrt{|F_{LL,RR}(\omega)|^2 + |F_s(\omega)|^2}}. \quad (6)$$

From Eqs. (4) and (5), we see that nonlocal pair amplitudes are proportional to the interdot coupling Γ_{LR} , which is an essential element of our model [45]. Moreover, F_s is zero when the dot levels are on resonance ($\epsilon_L = \epsilon_R = 0$) [46]. Therefore, dominant odd-frequency pairing on one of the dots requires left-right asymmetry, which can be achieved by setting $\Gamma_L \neq \Gamma_R$ or $\Gamma_{SL} \neq \Gamma_{SR}$. Odd-frequency pairing is suppressed when the DQD levels are out of resonance ($\epsilon_L \neq 0$ and/or $\epsilon_R \neq 0$).

Detection and manipulation of odd-frequency pairing.

We consider two different transport measurements. First, a voltage bias V is applied symmetrically to both normal leads [Fig. 2(a)]. This configuration is known as a *Cooper pair splitter* setup and has been used in recent experiments [26,33,37,38]. At zero temperature, conductance at lead L is given by

$$G_L(V) = 2G_0 [T_L^{qp}(eV) + T_{LL}^{eh}(eV) + T_{LR}^{eh}(eV)], \quad (7)$$

with $G_0 = 2e^2/h$ and quasiparticle tunneling transmission T_L^{qp} . T_{LL}^{eh} and T_{LR}^{eh} are the contributions from local and nonlocal Andreev processes, respectively. For subgap voltages ($|eV| < \Delta$), conductance is mainly given by Andreev processes while the quasiparticle contribution is almost negligible.

Alternatively, a current can flow through lead L if a voltage is applied to lead R [Fig. 3(a)]. This is the basis for a *nonlocal conductance measurement* [25,36,47] which, at zero

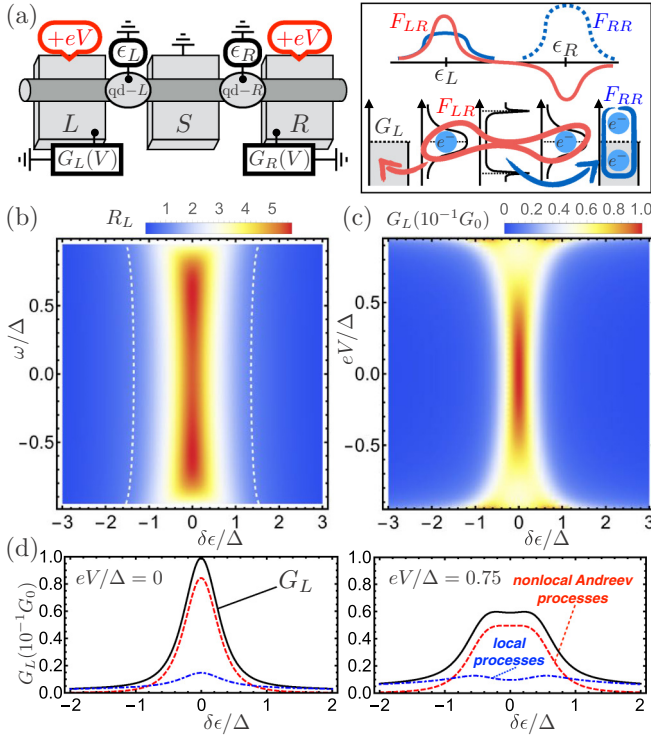


FIG. 2. Cooper pair splitter configuration. (a) A voltage V is applied symmetrically to both normal electrodes which allows us to measure the conductances $G_L(V)$ and $G_R(V)$ (left). Right: In an asymmetric DQD system, odd-frequency nonlocal F_{LR} (red line) can be enhanced on dot L and G_L is mainly due to nonlocal Andreev processes (red arrow). (b), (c) Map of the (b) ratio R_L and (c) conductance G_L as a function of applied voltage and level position $\delta\epsilon = \epsilon_L + 3\epsilon_R$. $R_L > 1$ inside the white dashed line. (d) Conductance (black solid lines), nonlocal Andreev (red dashed lines), and local processes (blue dotted-dashed lines) for $eV = 0$ (left) and 0.75Δ (right). For all plots, $T = 0$, $\Gamma_L/\Delta = 5$, $\Gamma_R = \Delta = 1$, $\Gamma_{SL}/\Delta = 0.1$, $\Gamma_{SR}/\Delta = 0.9$, and $\Gamma_{LR}/\Delta = 0.5$.

temperature, is given by

$$G_{LR}(V) = G_0 [T_{LR}^{eh}(eV) - T_{LR}^{ee}(eV)], \quad (8)$$

where T_{LR}^{ee} represents an electron tunneling process. At zero temperature, transmission probabilities for each process are calculated from the retarded Green's function as $T_{\mu\nu}^{\alpha\beta}(\omega) = 4\Gamma_\mu\Gamma_\nu [|(\check{g}^r)_{\mu\nu}^{\alpha\beta}(\omega + i0^+)|^2 + |(\check{g}^r)_{\mu\nu}^{\alpha\beta}(-\omega + i0^+)|^2]$, with $\alpha, \beta = e, h$ and $\mu, \nu = L, R$ [43]. Specifically, the transmission probability for Andreev processes reduces to

$$T_{\mu\nu}^{eh}(\omega) = 4\Gamma_\mu\Gamma_\nu [|F_{\mu\nu}(\omega)|^2 + |F_{\mu\nu}(-\omega)|^2]. \quad (9)$$

Consequently, we can connect each microscopic process to a symmetry class. Indeed, local even-frequency pair amplitudes $F_{\mu\mu}$ provide the probability amplitude for the transmission of the two electrons of a Cooper pair into the same lead, i.e., a local Andreev process sketched in Fig. 1(b). On the other hand, nonlocal components $F_{LR,RL}$ account for the probability amplitude of a process where the electrons of a Cooper pair split into different leads: a nonlocal Andreev process [Fig. 1(c)]. If both dot levels are aligned to the chemical potential, i.e., when the DQD is *on resonance* with

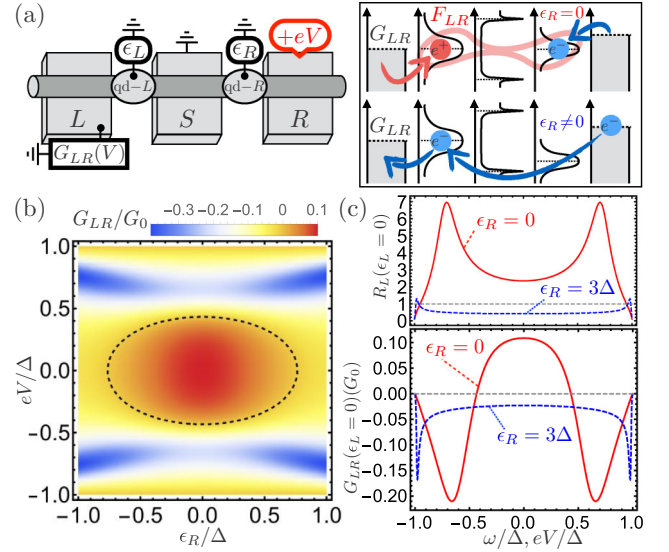


FIG. 3. Nonlocal conductance measurement. (a) A voltage is applied to lead R, allowing one to measure the conductance G_{LR} on lead L (left). Right: On resonance, odd-frequency nonlocal Cooper pairs provide a dominant contribution to G_{LR} (red arrow), while electron tunneling dominates out of resonance (blue arrow). (b) Map of G_{LR} as a function of the applied voltage and the position of dot R, ϵ_R , for $\epsilon_L = 0$. $G_{LR} > 0$ inside the black dashed line. (c) Ratio on dot L (top) and nonlocal conductance (bottom) for $\epsilon_L = \epsilon_R$ (red solid line) and $\epsilon_L \neq \epsilon_R$ (blue dashed line). For all plots, $\Gamma_L/\Delta = 5$, $\Gamma_R/\Delta = 0.2$, $\Gamma_{SL} = \Gamma_{SR} = \Gamma_{LR} = \Delta = 1$, and $T = 0$.

$\epsilon_L = \epsilon_R = 0$, nonlocal pair amplitudes $F_{LR,RL}$ are odd in frequency. The presence of odd-frequency pairing in the DQD and its connection to a specific microscopic process that has been successfully observed in recent experiments is one of the main conclusions of this work.

In an ideal setup, we can choose to uncouple one of the dots from the superconductor setting $\Gamma_{SL} = 0$. By local transmission through that dot, F_{LL} is suppressed and Eq. (6) reduces to $R_L = 1/R_R = \sqrt{\omega^2 + \Gamma_L^2}/\Gamma_{LR}$. As a result, for subgap energies $|\omega| < \Delta$, odd-frequency pairing becomes dominant at dot R (L) if $\Gamma_L < \Gamma_{LR}$ ($\Gamma_L > \Gamma_{LR}$) is satisfied. Consequently, in the Cooper pair splitter configuration, the conductance at one of the leads, Eq. (7), can be completely dominated by nonlocal Andreev processes, which is a signature of the presence of odd-frequency superconductivity [43]. Decoupling one of the dots requires careful patterning of the DQD, similarly to recent experiments in graphene [47]. In many other experiments, however, DQDs are constructed by electrical confinement from electrodes on quasi-one-dimensional materials [25,26], as sketched in Fig. 2(a). It is thus challenging to decouple one of the dots from the superconductor. Therefore, we consider $\Gamma_{SL} \neq 0$ in the following. We start with strong left-right asymmetry by setting $\Gamma_L \neq \Gamma_R$ and $\Gamma_{SL} \neq \Gamma_{SR}$ at the same time. To exclude double occupancy on the dots, we work in the regime with $\Gamma_{L,R} > \Gamma_{SL,SR}$ where a single-particle description of transport at the DQD system is allowed [29]. In Fig. 2(b) we show the ratio on dot L, R_L , as a function of ω and $\delta\epsilon = \epsilon_L + \alpha\epsilon_R$, with α a constant. In agreement with our previous analysis, odd-frequency pairing is dominant on dot L for subgap energies as long as the dot levels are

close to the chemical potential, i.e., for $|\omega|, |\delta\epsilon| \lesssim \Delta$. At zero temperature, the applied voltage corresponds to the frequency ω . The conductance at lead L is enhanced for the same bias voltage regime, as shown in Fig. 2(c). A detailed analysis shows that the conductance is mainly given by nonlocal Andreev processes which stem from induced odd-frequency pairing [red lines in Fig. 2(d)].

A small degree of asymmetry is experimentally inevitable. However, by setting $\Gamma_{SL} \sim \Gamma_{SR}$ in the previous results, the contribution from local Andreev processes is enhanced and becomes comparable to that of nonlocal processes, making it more difficult to establish a connection between conductance and odd-frequency pairing. For weakly asymmetric setups, with $\Gamma_{SL} \sim \Gamma_{SR}$, it is better to perform a *nonlocal conductance measurement* where odd-frequency-induced nonlocal Andreev processes only compete with electron tunneling processes [see Fig. 3(a)]. In principle, the two contributions should cancel each other [30,48]. In a DQD three-terminal device, however, the relative position of the dot levels becomes very important to favor Andreev processes, since they mainly take place when the levels are aligned on resonance. For this condition, the pair amplitude F_{LR} is odd in frequency. Therefore, a positive nonlocal conductance proves the presence of the odd-frequency pair amplitude [11]. Setting $\epsilon_L = 0$, we show in Fig. 3(b) a map of G_{LR} as a function of eV and ϵ_R . Within the black dashed line, conductance is positive, i.e., dominated by nonlocal Andreev processes. As $|eV| \sim \Delta$, however, electron tunneling processes become more important and the conductance changes sign. The connection between positive Andreev-dominated conductance and odd-frequency symmetry is explicitly shown in Fig. 3(c). When the dot R is on resonance (red solid lines), the nonlocal conductance is positive for subgap energies (bottom) while the odd-frequency pair amplitude is dominant on dot L (top). If the dot R is taken out of resonance, the conductance becomes negative and the odd-frequency pair amplitude is suppressed.

Spin-triplet superconducting lead. When the central superconducting lead is a one-dimensional spin-triplet p -wave superconductor, as in the case of a metallic nanowire on Sr_2RuO_4 [49], the induced pairing amplitude at its edges is odd-frequency triplet s wave represented by $f(\omega_n) = \Delta/\omega_n$, which displays Majorana edge states [28,50,51]. On the DQD,

F_{LL} , F_{RR} , and F_s are now odd-frequency functions, while F_a is even-frequency pairing (see Table I). Consequently, conductance measured in the Cooper pair splitter configuration is a very useful tool to study the symmetry of edge states at spin-triplet superconductors. For the perfectly symmetric case, where both $\Gamma_L = \Gamma_R \equiv \Gamma_N$ and $\Gamma_{SL} = \Gamma_{SR} \equiv \Gamma_S$ are satisfied, the even-frequency term F_a vanishes and Cooper pairs injected on the same lead maintain the odd-frequency symmetry of the superconducting lead. F_a increases proportionally to the difference between the tunneling rates for left and right dots. For example, if the asymmetry originates from the coupling to the normal leads (superconducting lead), the even-frequency component follows $F_a \propto \Gamma_S(\Gamma_L - \Gamma_R)$ [$F_a \propto (\Gamma_{SL} - \Gamma_{SR})(|\omega_n| - \Gamma_N)$].

Conclusions. We propose a way to generate odd-frequency spin-singlet s -wave Cooper pairs on DQD-based three-terminal devices. Due to the orbital degree of freedom in the DQD, the symmetry of induced Cooper pairs can be broken, featuring a superposition of even- and odd-frequency terms. Each symmetry type, however, is responsible for a different transport process, a unique feature of this setup. For spin-singlet superconductors, nonlocal Andreev processes on resonance are uniquely caused by odd-frequency pairing. Therefore, odd-frequency pairs can be detected from standard conductance measurements in asymmetric devices where the contribution of nonlocal Andreev processes is greatly enhanced. Additionally, it is possible to manipulate the amplitude of odd-frequency Cooper pairs by tuning the position of the dot levels. The situation is reversed if the central electrode is a spin-triplet p -wave superconductor. Odd-frequency triplet s -wave pairing is now associated with local Andreev processes which are the dominant contribution to the conductance if the dots are symmetrically coupled to the leads.

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