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From Cooper pair splitting to nonlocal spectroscopy of a Shiba state

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Cooper pair splitting (CPS) is a way to create spatially separated, entangled electron pairs. To this day, CPS is often identified in experiments as a spatial current correlation. However, such correlations can arise even in the absence of CPS, when a quantum dot is strongly coupled to the superconductor, and a subgap Shiba state is formed. Here, we present a detailed experimental characterization of those spatial current correlations, as the tunnel barrier strength between the quantum dot and the neighboring normal electrode is tuned. The correlation of the nonlocal signal and the barrier strength reveals a competition between CPS and the nonlocal probing of the Shiba state. We describe our experiment with a simple transport model and obtain the tunnel couplings of our device by fitting the model’s prediction to the measured conductance correlation curve. Furthermore, we use our theory to extract the contribution of CPS to the nonlocal signal.

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1. INTRODUCTION

Cooper pair splitter devices were extensively studied both experimentally [1–8] and theoretically [9–19] in recent years. They are proposed to serve as the source of spatially separated entangled pair electrons. The standard Cooper pair splitter [see Fig. 1(a)] consists of a central superconducting electrode, which serves as the source of Cooper pairs, two normal leads, to which the split electron pair is transferred, and two quantum dots in between. The gate voltages of the quantum dots serve as the experimental knobs to tune the currents in the two different arms, utilizing the discrete energy spectrum of the dots.

Theoretically, several ways were proposed to test the performance of a Cooper pair splitter device, starting from current or noise correlations [20–22], through spin selective measurements [23–26] to proving the entanglement by demonstrating the violation of the Bell inequality [19,27–33] or using more complex spin-qubit or circuit quantum electrodynamics architectures [18,34,35] or even time domain measurements [36].

The experiments primarily focused on spatial current correlations [1–8]. In general, the currents flowing in the two arms are attributed to two different processes. The first process is the local pair tunneling (LPT), when both electrons of a Cooper pair are transferred to the same normal lead [see the black and green arrow on Fig. 1(a)]. This process involves only one arm of the device, and correspondingly it is independent of control parameters of the other arm. The second process is the Cooper pair splitting (CPS), when the two electrons end up in different normal electrodes [see the black and red arrows on Fig. 1(a)]. As both arms of the device are involved, the CPS process depends on the control parameters of both arms. Hence it results in a nonlocal signal, i.e., the current in one arm of the device is tuned by the control parameters (on-site energy (plunger gate voltage) and tunnel amplitude (barrier gate voltage)) of the other arm via tuning the CPS contribution.

In Ref. [37], we have shown that a nonlocal signal can be also present when the conductance of one of the arms of the Cooper pair splitter is quenched. It is achieved by isolating the quantum dot from its normal lead by a strong tunnel barrier. Since in this situation the CPS is forbidden, there must be another transport process that produces a nonlocal signal. This process emerges if the tunnel coupling between the superconductor and the isolated quantum dot is sufficiently strong, which gives rise to the formation of the Yu-Shiba-Rusinov (Shiba) state [38–41].

The subgap Shiba state is the result of the hybridization of the dot states and quasiparticle states of the...
superconductor and was extensively studied in a variety of hybrid nano devices [42–67]. They inherited their name from the subgap states in an analogous system, namely magnetic adatoms on a superconducting surface, which are widely studied by scanning tunneling microscope experiments [68–84]. We will call the transport mechanism responsible for the nonlocal signal does not create spatially separated electron pairs. The left QD is drawn with a dashed line, because it is not tuned throughout this work, but fixed as it only weakly transmits. [(b), (c)] Cross section and false color SEM image of the device. The InAs nanowire (NW) is placed on an array of bottom gate electrodes and contacted by the two normal and one superconducting electrode. The QD is formed in the NW, and tuned by the voltage on the plunger gate, \( g_p \). The tunnel coupling to the \( N_R \) lead is tuned by the barrier gate, \( g_b \).

FIG. 1. (a) Schematics of a Cooper pair splitter. Cooper pairs are extracted from a central superconducting electrode (SC) to two normal leads (\( N_L \) and \( N_R \), through quantum dots (QDs). The currents are built up of two contributions, the Cooper pair splitting (CPS, black and red arrows) and the local pair tunneling (LPT, black and green arrows). In the presence of a Shiba state at zero energy—if the SC and the QD are strongly coupled—a third process, the Shiba-assisted local pair tunneling (SPT, black and blue arrows) emerges. This produces a CPS-like nonlocal signal but does not create spatially separated electron pairs. The left QD is drawn with a dashed line, because it is not tuned throughout this work, but fixed as it only weakly transmits. [(b), (c)] Cross section and false color SEM image of the device. The InAs nanowire (NW) is placed on an array of bottom gate electrodes and contacted by the two normal and one superconducting electrode. The QD is formed in the NW, and tuned by the voltage on the plunger gate, \( g_p \). The tunnel coupling to the \( N_R \) lead is tuned by the barrier gate, \( g_b \).

strong), a significant contribution of the nonlocal signal comes from the SPT process. Hence, in this parameter range, it is necessary to use our framework for a sound analysis. Since following earlier approaches, e.g., Refs. [1,2,5–7], attributing all of the nonlocal signal to CPS would overestimate the real CPS contribution in our case. We also show that in the case when the quantum dot of the Cooper pair splitter is well coupled to the \( N_R \) lead, the nonlocal signal dominantly comes from the splitting of Cooper pairs, and correspondingly the earlier approaches gives a reasonable result for the CPS contribution in that parameter range.

Our paper is structured as follows. In Sec. II, the device geometry and measurement techniques are introduced. In Sec. III, the experimental results are presented. We demonstrate the presence of a nonlocal signal for a wide range of parameters, we show a correlation between the local and the nonlocal signal, and we relate them to the Shiba state. In Sec. IV, the theoretical model and the transport calculation are introduced, and are used to fit the experimental data. In Sec. V, we discuss our findings, namely, the origin of the nonlocal signal and the correlation between the local and the nonlocal signal, we calculate the contributions of the CPS and SPT processes, and we discuss the validity of earlier splitting efficiency definitions. Finally, in Sec. VI, we conclude our findings.

II. DEVICE GEOMETRY

The Cooper pair splitter device is illustrated in Fig. 1. The schematics of the cross-section and a false-color SEM image of the device are shown in Figs. 1(b) and 1(c), respectively. The basis of the circuit is an InAs nanowire (NW), with a diameter of 70 nm, grown by cold catalyst assisted molecular beam epitaxy [88,89], using a two-step growth method to suppress the stacking faults [90]. First, an array of bottom gate electrodes (made of 4/18 nm Ti/Pt in brown) was defined by e-beam lithography and evaporation. The two outermost electrodes of the 9 are 1.3 \( \mu \)m wide and placed below the normal contacts. The middle electrode is 250 nm wide and placed below the superconductor. The remaining 3+3 electrodes are 30 nm wide with 100-nm period and serve to tune the electron density in the nanowire. The gate electrodes are covered by a 25 nm thick SiN\(_x\) layer (green), grown by plasma-enhanced chemical vapor deposition to electrically isolate the gates from the rest of the device. The SiN\(_x\) was removed by reactive ion etching with CHF\(_3\)/O\(_2\) at the end of the gate electrodes to contact them [91]. The nanowire is contacted by two 4.5/100 nm thick Ti/Au normal electrodes (\( N_L \) and \( N_R \) in yellow) and a 4.5/110/20 nm thick Pd/Pb/In superconducting contact (SC in red) [92]. The nanowire was cut by focused ion beam prior to the deposition of the superconducting contact [see Fig. 1(b)] to suppress the direct tunnel coupling between the two arms of the device [93,94].

The differential conductance of the two arms were measured simultaneously with lock-in technique, using 10 \( \mu \)V AC signal on the superconducting contact at 237 Hz, on the two normal contacts by home-built current-voltage (I/V) converter. The measurements were carried out in a Leiden Cryogenics CF-400 top-loading cryo-free dilution refrigerator at a bath temperature of 35 mK. Prior to cooldown, the sample
Fig. 2. [(a), (b)] Simultaneously measured zero-bias differential conductances of the two arms of the device, as the functions of the $V_B$ (barrier) and $V_P$ (plunger) gate voltages [37]. The quantum dot-like resonances of the right arm [marked with black square and red triangle in panel (b)] also appear in the conductance of the left arm (a). This nonlocal signal persists even when the conductance of the right arm is quenched. (c) Color-coded line cuts from panels (a) and (b) illustrating the correlation between the nonlocal signal in the left arm and the local conductance of the right arm. A gate-dependent background conductance is subtracted from each $G_L$ curve. The conductance maxima values are extracted from each line cut along the resonances, as illustrated on panel (c) by the marks. These value pairs are plotted as a scatter plot in panel (d) to further demonstrate the correlation of the two conductances. The three particular example pairs of panel (c) are highlighted by color-coded circles. The nonlocal signal in the left arm increases with an increasing conductance of the right arm. (e) Finite-bias scatter plot in panel (d) to further demonstrate the correlation of the two conductances. The three particular example pairs of panel (c) are extracted from each line cut along the resonances, as illustrated on panel (c) by the marks. These value pairs are plotted as a scatter plot in panel (d) to further demonstrate the correlation of the two conductances. The three particular example pairs of panel (c) are highlighted by color-coded circles. The nonlocal signal in the left arm increases with an increasing conductance of the right arm. (e) Finite-bias characterization of the quantum dot with open barrier, measured along the white dashed line on panel (b), at $V_B = -0.83 \text{ V}$. The eye-shaped crossing is the signature of a Shiba state, which highlights the strong coupling between the superconductor and the quantum dot. The black dashed lines are used to estimate the charging energy to $U \approx 0.4 \text{ meV}$ [37].

was pumped overnight to remove the absorbed water from the surface of the nanowire.

III. EXPERIMENTAL RESULTS

The experimental results are summarized in Fig. 2. Figures 2(a) and 2(b) show the simultaneously measured zero-bias differential conductance (in units of $G_0 = 2e^2/h$) of the two arms of the Cooper pair splitter device, $G_L$ and $G_R$, as the function of the gate voltages, $V_B$ and $V_P$, which are applied on gate $g_B$ and $g_P$, respectively. Note that both gate electrodes are placed under the same arm of the device [see Fig. 1(b)], and the left arm is not tuned, but fixed as it weakly transmits. Hence, contrary to usual Cooper pair splitters, instead of a quantum dot and a normal lead, the left arm can be considered as a tunnel coupled normal electrode. The barrier gate voltage, $V_B$, dominantly tunes the tunnel coupling to the $N_R$ electrode, while $V_P$, the plunger gate voltage, tunes the level position of the quantum dot. The conductance $G_R$ through the right arm [see Fig. 2(b)] is completely quenched for $V_B < -1.1 \text{ V}$, while for more positive barrier gate voltages it starts to conduct along two parallel resonance lines (marked by a square and a triangle). This conduction pattern indicates a quantum dot-like behavior.

Remarkably, the two $G_R$ resonances of the quantum dot also appear in the conductance $G_L$ of the left arm [see Fig. 2(a)] as a conductance enhancement on top of a smooth background. We will denote this conductance enhancement by $\Delta G_L$, and call it the nonlocal signal. When the conductance of the right arm is finite ($V_B > -1.1 \text{ V}$), the presence of this nonlocal signal is in agreement with previous measurements on Cooper pair splitters [1, 2, 5–7]. That is, when the quantum dot in the right arm is tuned to resonance, an additional transport channel opens, Cooper pairs are not only transferred to the left normal electrode by LPT processes, but they can be split between the two arms, and correspondingly increase the conductance of both arms. This $V_B > -1.1 \text{ V}$ regime of the data is qualitatively consistent with the picture of CPS.

However, the $V_B < -1.2 \text{ V}$ range of the data in Figs. 2(a) and 2(b) excludes the scenario that CPS is the only transport process inducing the nonlocal signal. There, the conductance $G_R$ of the right arm is suppressed, but the nonlocal signal, i.e., the peaks in $G_L$, persists throughout the whole map. When $G_R$ is quenched, the presence of the nonlocal signal cannot be attributed to the CPS anymore, since no electron can flow to the $N_R$ electrode. Both electrons of the Cooper pairs have to leave to the left normal lead.

Furthermore, our data reveal that the amplitude of the nonlocal signal $\Delta G_L$ is controlled via the local conductance $G_R$. In other words, by tuning the rightmost barrier more transparent, the current moving in the left arm is also increased. This is illustrated in Fig. 2(c), by line cuts from 2(a)
and 2(b) along the color coded lines. The smooth background conductance is subtracted from each line cut of $G_L$ and only the nonlocal signal, $\Delta G_L$, is plotted. When $G_R$ is quenched, the nonlocal signal goes up to about $\Delta G_L \approx 0.005 \, G_0$ [see the orange curves on Fig. 2(c), measured at $V_B = -1.281 \, V$]. As the local conductance increases, the amplitude of the nonlocal signal also increases (see the purple and blue curves, measured at $V_B = -1.098$ and $-1.119 \, V$, respectively). Note that $G_R$, and correspondingly the nonlocal signal also, varies in a nonmonotonic fashion as the barrier gate voltage is tuned; we attribute this to disorder in the sample.

The above-mentioned systematic relation of the local conductance $G_R$ and the nonlocal signal $\Delta G_L$ is reinforced by the data shown in Fig. 2(d). Let us exemplify how we derive this data from the previous panels, using the line cuts in Fig. 2(e), focusing on the right maxima of the blue curves (marked by black squares). The maximum value of the nonlocal signal (left panel) is about $\Delta G_L^{\text{max}} \approx 0.017 \, G_0$, while the maximum value of the local conductance (right panel) is $G_R^{\text{max}} = 0.08 \, G_0$. These values give the vertical and horizontal coordinates of the corresponding data point in Fig. 2(d), respectively (see the black square highlighted by the blue circle). The other points of Fig. 2(d) were obtained similarly by reading the maxima of all vertical line cuts on Figs. 2(a) and 2(b). As Fig. 2(d) shows, even when the conductance of the right arm is quenched, i.e., $G_R^{\text{max}} = 0$, the amplitude of the nonlocal signal is finite, residing between 0.002 $G_0$ and 0.01 $G_0$. As $G_R$ is increased by lowering the barrier to the N$_R$ lead, the nonlocal signal also increases in a monotonous, saturating fashion. Only data points with $G_R^{\text{max}} < 0.1 \, G_0$ are shown in the figure. We choose this requirement to ensure the weak coupling to the right lead, which will be assumed in the modeling.

Using finite-bias measurements, we establish that the tunnel coupling between the superconducting lead and the quantum dot is strong, and hence a Shiba state is formed in the device. The finite-bias differential conductance measured along the white dashed line on Fig. 2(b) at $V_B = -0.83 \, V$ is shown in Fig. 2(e). The eye-shaped crossing conductance lines are the usual signature of a Shiba state, formed in a strongly coupled superconductor–quantum dot hybrid (see, e.g., Ref. [54]). Correspondingly, the resonances Figs. 2(a) and 2(b) are the signatures of the zero-energy Shiba states rather than charge degeneracies of a quantum dot. These observations suggest that in our model of the device (see next section) we have to incorporate a strong, coherent hybridization between the superconductor and the quantum dot. Based on the finite-bias measurement shown in Fig. 2(e) the charging energy of the quantum dot can also be estimated, following the dashed lines, yielding $U \approx 0.4 \, meV$.

Our conclusion from this analysis is that when the conductance $G_R$ of the right arm is quenched, the conductance enhancement $\Delta G_L$ of the left arm originates from the (zero-energy) Shiba state, i.e., the SPT mechanism, rather than the splitting of Cooper pairs. Hence, the conductance enhancement can be viewed as the nonlocal probing of the Shiba state. We analyzed this region in detail in Ref. [37].

In the following, we will use a simple model to explain the experimental findings, namely (i) the presence of the nonlocal signal when the right arm is quenched and (ii) the correlation between the conductances when $G_R$ is finite. We will model the Shiba state using the zero bandwidth approximation for the superconductor. This model will enable us to identify and separate contributions of the SPT and CPS processes [see Fig. 1(a)] to the nonlocal signal $\Delta G_L$.

### IV. MODEL

#### A. Zero-bandwidth approximation of the Shiba state

The Shiba state is formed by the hybridization of the quasiparticles of the superconductor and the quantum dot degrees of freedom induced by the strong tunnel coupling between the dot and the superconductor. A simple way to describe the Shiba state is by treating the superconductor using the zero bandwidth approximation (ZBA) [55,85–87]. In this approximation, the quasiparticles of the superconductor are restricted only to a discrete energy level at energy $\Delta$, which can host 0, 1, or 2 quasiparticles. The corresponding Hamiltonian is

$$H_{\text{ZBA}}^{\text{SC}} = \Delta c_\uparrow^\dagger c_\downarrow + \Delta c_\downarrow^\dagger c_\uparrow = \Delta \sum_\sigma \gamma_\sigma \gamma_\bar{\sigma},$$

where $\Delta$ is the superconducting gap, $c_\sigma^\dagger$ is the electron annihilation (creation) operator in the superconductor, and $\gamma_\sigma$ is the annihilation (creation) operator of the quasiparticles, defined by the usual Bogoliubov transformation, $c_\sigma = \frac{1}{\sqrt{2}}(\gamma_\sigma - \sigma \gamma_\bar{\sigma})$.

The quantum dot is described by the single-impurity Anderson Hamiltonian,

$$H_{\text{QD}} = \sum_\sigma \varepsilon n_\sigma + U n_\uparrow n_\downarrow,$$

where $\varepsilon$ is the level position of the dot, $U$ is the charging energy, and $n_\sigma = d_\sigma^\dagger d_\sigma$ is the number of the electrons with spin $\sigma$ on the dot, with $d_\sigma^\dagger$ being the annihilation (creation) operator of electrons with spin $\sigma$ on the dot. Note that compared to the usual Cooper pair splitter geometry, where quantum dots are placed on both side of the superconductor [as illustrated in Fig. 1(a)], in this model we only consider the dot of the right arm. Since in the experiment the left arm of the device was not tuned but fixed in a weakly transmitting region, the left normal electrode and the dot left is modeled as a tunnel coupled normal lead for simplicity [see Fig. 3(a) and Sec. IV B].

The tunnel coupling between the superconductor and the dot is described by

$$H_{\text{TS}} = t_S \sum_\sigma (d_\sigma^\dagger c_\sigma + c_\sigma^\dagger d_\sigma).$$

Using the Bogoliubov transformation above, this translates to

$$H_{\text{TS}} = \frac{1}{\sqrt{2}} \sum_\sigma t_S [d_\sigma^\dagger (\gamma_\sigma - \sigma \gamma_\bar{\sigma}) + (\gamma_\bar{\sigma} - \sigma \gamma_\sigma) d_\sigma].$$

The composite system of the superconductor and the quantum dot is hence modeled by $H_{\text{ZBA}}^{\text{SC}} + H_{\text{QD}} + H_{\text{TS}}$. A natural basis of the corresponding 16-dimensional Fock space is the product basis of the electron Fock space of the QD and the quasiparticle Fock space of the superconductor, that is, the basis $|i, j\rangle = |i\rangle_{\text{QD}} \otimes |j\rangle_{\text{SC}}$. Here, $\{|0\rangle_{\text{QD}}, |1\rangle_{\text{QD}}, |\downarrow\rangle_{\text{QD}}, |\uparrow\rangle_{\text{QD}}\}$ is the canonical basis of the electron Fock space of the QD and
The tunnel coupling $t_S$ between the superconductor (SC) and quantum dot (QD) creates a Shiba state (in blue). The normal leads, $N_L$ and $N_R$, probe the Shiba state with coupling strength $t_L$ and $t_R$, respectively. For simplicity, the SC is treated within the zero bandwidth approximation, in which the quasiparticles are restricted to a discrete quantum dot (QD) creates a Shiba state (in blue). The normal leads, $N_L$ and $N_R$, probe the Shiba state with coupling strength $t_L$ and $t_R$, respectively. In the shaded blue region, the nonlocal signal is not a good measure of the CPS, since the SPT contribution is not negligible.

(c) Illustration of the dynamics induced by the coupling to the normal leads. The tunneling induces transitions between the different ground states of the dot-superconductor system, the singlet and the doublets. The highlighted, colored arrows illustrate the tunnel processes shown on Fig. 1(a).

(d) Fitting the experimental data with the ZBA model. Purple line is the fit with $t_S = 0.145$ meV, $\Gamma_L = 1$ meV, and $\Gamma_R = 0-3.6$ meV.

(e) Contribution of the CPS (red) and the SPT (blue) processes to the nonlocal signal of the left arm and the relative ratio of the CPS processes (black). In the shaded blue region, the nonlocal signal is not a good measure of the CPS, since the SPT contribution is not negligible.

FIG. 3. (a) Schematics (top) and energy diagram (bottom) of our model. The tunnel coupling $t_S$ between the superconductor (SC) and quantum dot (QD) creates a Shiba state (in blue). The normal leads, $N_L$ and $N_R$, probe the Shiba state with coupling strength $t_L$ and $t_R$, respectively. For simplicity, the SC is treated within the zero bandwidth approximation, in which the quasiparticles are restricted to a discrete quantum dot (QD) creates a Shiba state (in blue). The normal leads, $N_L$ and $N_R$, probe the Shiba state with coupling strength $t_L$ and $t_R$, respectively. In the shaded blue region, the nonlocal signal is not a good measure of the CPS, since the SPT contribution is not negligible.

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These transitions are illustrated in Fig. 3(c) and described in the following.

For simplicity, we assume zero temperature, a gate-voltage configuration enabling resonant tunneling, i.e., that \( E_S = E_o \) for the energy eigenvalues of the singlet and double ground states and that a small negative bias voltage is applied on the superconducting lead, to ensure that electrons flow only to the vicinities of the superconducting lead, to ensure that electrons flow only to the normal leads perturbatively, using Fermi’s golden rule.

The tunnel coupling to the normal leads is kept weak in the experiment, we treated the electron transport to the normal leads perturbatively, using Fermi’s golden rule. The tunnel coupling to the normal leads induces parity changing transitions between the different states of the superconductor–quantum dot hybrid as illustrated in Fig. 3(c). For example, the black arrow denotes the tunneling event, which brings the system from the \(|S\rangle \) state to the \(|\downarrow\rangle \) state, while an electron is transferred to the N lead. The time evolution of the occupation probabilities of the \(|S/\sigma\rangle \) states, \( P_{S/\sigma} \) is described by a classical master equation,

\[
\frac{dP_{S}}{dt} = \sum_{\sigma} \left[ \left(W_{S\sigma}^{L} + W_{S\sigma}^{R}\right)P_{\sigma} - \left(W_{S\sigma}^{L} + W_{S\sigma}^{R}\right)P_{S}\right],
\]

\[
\frac{dP_{\sigma}}{dt} = \left(W_{S\sigma}^{L} + W_{S\sigma}^{R}\right)P_{S} - \left(W_{S\sigma}^{L} + W_{S\sigma}^{R}\right)P_{\sigma},
\]

(7)

together with the normalization condition \( P_{S} + \sum_{\sigma} P_{\sigma} = 1 \).

After determining the stationary solution \( \left(dP_{\sigma}/dt = 0 \right) \) of the master equation, the current in lead \( N_{LR} \) is given by

\[
I_{LR} = \varepsilon \sum_{\sigma} \left( W_{S\sigma}^{LR} P_{S} + W_{S\sigma}^{LR} P_{\sigma}\right).
\]

(8)

C. Comparison of the experiment and the theory

To compare the experimentally measured differential conductances to the calculated currents, one should find a relation connecting them. A naive way would be assuming a linear relation and simply using the 10 \( \mu V \) amplitude of the AC bias voltage for the conversion, i.e., \( I_{AC}^{LR} = G_{LR} \times 10 \mu V \). However, while the calculation results in a zero-width conductance peak, in the experiment the peak is broadened, and therefore the naive way strongly underestimates the saturation DC current at high bias. We estimate the saturation current using the zero-bias data in the following way. We estimate the broadening of the peaks by converting the width of the gate-dependent resonance curves measured at zero bias [see, e.g., Fig. 2(c)] using the lever arm, determined from the finite bias measurement on Fig. 2(e). This yields about 50 \( \mu V \) width, barely varying with \( V_0 \). Assuming a Lorentzian shape for the resonance with this width the ratio of the saturation DC current and the zero-bias AC current is

\[
\frac{I_{LR}^{sat}}{I_{LR}^{AC}} = \frac{\int_{0}^{\infty} G_{LR}(V) dV}{G_{LR}(V = 0)V_{AC}} \approx 4.
\]

(9)

In the following, we use this relation to convert the measured zero-bias differential conductances to saturated currents.

We used the framework detailed above to fit our experimental data. The charging energy and the superconducting gap is fixed to the experimental value of \( U = 0.4 \) meV and \( \Delta = 0.25 \) meV, respectively. As \( \varepsilon \) is fixed by the degeneracy condition, only \( t_S \) remains a free parameter in the model of the coupled superconductor and QD. In the following, we will focus first on the degeneracy point near \( \varepsilon = 0 \), i.e., the 0-1 transition of the dot. Our results can be directly translated to the other degeneracy point, using symmetry arguments. Besides \( t_S \), which determines the transition matrix elements, the couplings to the normal leads, \( \Gamma_L \) and \( \Gamma_R \), are also fitting parameters of the model. Note that \( \Gamma_R \) is tuned in the experiment, and therefore the fitting does not give a single value but a range starting from zero. Therefore, the fitting parameter is not \( \Gamma_R \) itself, but its maximal value \( \Gamma_R^{max} \).

The fitted curve is shown in Fig. 3(d) with the purple line, using \( t_S = 0.145 \) meV, \( \Gamma_L = 1 \) meV, and \( \Gamma_R = 0 - 3.6 \) meV. The fit captures well the main features of the experiment. First, the model reproduces the finite current in the left lead even when the conductance of the right arm is quenched. Second, \( G_L \) also increases as \( G_R \) is enhanced by the increasing \( \Gamma_R \). Third, the model gives a saturating tendency for the nonlocal signal as the barrier to the right lead is opened up. Note that the precise values of the fitted parameters may slightly vary when one or the other assumption used during the evaluation is relaxed, but the qualitative behavior remains unchanged.

V. DISCUSSION

Previously we demonstrated that the ZBA model of the Shiba state well reproduces our experimental findings: (i) the presence of the nonlocal signal, even when \( G_R \) is quenched, and (ii) the correlation between the height of the local and the nonlocal conductance peaks. The model allows for the detailed understanding of the experimental features, and furthermore, it can distinguish between the SPT and CPS contribution of the currents.

First, we have to discuss why the Shiba language (i.e., treating the superconductor–quantum dot hybrid as a coherently coupled unit) is necessary to describe our measurements. Often in Cooper pair splitters the electron transition between the superconductor and quantum dots is also described by tunnel rates, derived in a perturbative model, which assumes weak tunnel couplings [1,2,6]. In our measurements, the
presence of the eye-shaped subgap resonances in Fig. 2(e) implies that the tunnel coupling $t_5$ between the superconductor and the quantum dot is comparable to the value of the gap (which we also verified by the fit, $t_5 \approx 0.145$ meV). Therefore, it cannot be treated perturbatively, but the coherent hybridization of the superconductor and the dot has to be taken into account. Considering the usual perturbative description of the Cooper pair splitters, the presence of the nonlocal signal in the region, where $G_R$ is zero, is surprising, since the process, which can generate a nonlocal signal—the CPS—is forbidden. However, in the Shiba description, this nonlocal signal is naturally present, as we will see below.

Second, we observed that the amplitude of the nonlocal signal increases as $G_R$ is increased by enhancing the tunnel coupling to the $N_R$ lead. This correlation is a result of a bottleneck effect. When the right lead is isolated from the rest of the device, all electrons have to leave to the $N_L$ lead. This means that both the singlet→doublet and the doublet→singlet transitions must occur, involving a tunneling to the left lead. If the transition rates are much different, the slower one creates a bottleneck. Indeed, using the model parameters obtained by the fit, one finds that $\hbar W_{S5}^L \approx 0.036$ μeV [the blue and the top left gray arrow on Fig. 3(c)] and $\hbar W_{S5}^R \approx 0.191$ μeV [the black and the bottom left gray arrow on Fig. 3(c)]. The former rate is more than five times smaller than the latter one, i.e., the left current is limited by the doublet→singlet transition. However, opening up the barrier to the right lead opens up another transport channel for the slower process. As the $W_{S5}^R$ rate becomes larger than $W_{S5}^L$, the doublet→singlet process happens more favorably by transferring an electron to the right lead, and the bottleneck starts to be resolved. Although the presence of the new transport channel first means that not all the electrons are transferred to the left lead (which would mean the decrease of the nonlocal signal), but the whole transport sequence speeds up so much that altogether the left current increases.

The presence of nonlocal signals was reported previously in Cooper pair splitters hosting superconducting subgap states [53,57]. The authors used the infinite superconducting gap limit description—frequently referred as Andreev bound states—to model their experimental results. Using this approximation, all transition matrix elements contain the determinants’ matrix elements, and therefore the ratio of the left rates and the right rates are the same, i.e., $W_{S5}^R = W_{S5}^L$. With a simple derivation it can be shown that increasing $\Gamma_R$ cannot resolve the bottleneck effect, since it drops out of the formula of $I_L$. Here, first we introduce how these processes are defined based on single tunneling events, and then we calculate their contribution.

The whole transport consists of a sequence of singlet→doublet and doublet→singlet transitions. Assuming that one starts from the singlet ground state, the transport can be grouped into a random sequence of SPT and CPS processes. Let us illustrate this using Fig. 3(c). The highlighted arrows indicate a CPS and a SPT process. Starting from the singlet ground state, let us assume that first an electron (with up spin) is transferred to the left lead. This is indicated by the black arrow. At the end of the tunneling event, the superconductor–quantum dot hybrid system stays in the $|\downarrow\rangle$ state. To return to the singlet state, there are two possibilities. First, a second electron (with down spin) leaves to the left lead also (indicated by the blue arrow), and second, this electron is transferred to the right lead (red arrow). In the first process, two electrons with opposite spins are transferred to the same lead, and therefore a SPT event occurred, while in the second process the two electrons end up in different leads, corresponding to a CPS process. As the system returned to the singlet state, the transport cycle starts again.

The contribution of the CPS and SPT processes can be calculated within the framework of our model. Substituting the stationary solution of the master equation (7) to the expression of the currents, Eq. (8), one finds that the currents can be partitioned as

$$I_{LR} = I_{SPT,L/R} + I_{CPS}$$

where

$$I_{SPT,L/R} = A \sum_\sigma 2W_{S5}^{LR} W_{\sigma S}^{LR}$$

is the SPT contribution and

$$I_{CPS} = A \sum_\sigma (W_{S5}^{L\sigma S} W_{\sigma S}^{R} + W_{S5}^{R\sigma S} W_{\sigma S}^{L})$$

is the CPS one. In $I_{SPT,L/R}$, the factor of 2 indicates that both electrons are transferred to the same lead, and the normalization factor is $A = 4e(\sum_\sigma(W_{S5}^{L\sigma S} + W_{S5}^{R\sigma S} + 2W_{S5}^{L\sigma S} + 2W_{S5}^{R\sigma S}))^{-1}$. Note that to obtain these formulas, the spin rotational symmetry was used, which implies the spin independence of the transition rates, e.g., $W_{S5}^{L\sigma S} = W_{S5}^{1\sigma S}$. We should remind the reader that the presented model does not account for the LPT process, as it is independent of the Shiba state, consisting only of Cooper pairs tunneling from the superconductor directly to the normal lead.

This separation of $I_L$ using the obtained fit parameters is plotted in Fig. 3(e). The CPS and SPT contributions are plotted in red and blue, respectively [the total current is shown in Fig. 3(c) in purple], and furthermore the CPS efficiency is plotted in black, which depends only on the $\Gamma_R/\Gamma_L$ ratio for fixed $t_5$, not on their values. When $G_R = 0$ the splitting of Cooper pairs is forbidden, since no electrons can be transferred to the right lead. Accordingly, the CPS contribution is zero. The total current in the left lead comes only from the SPT process. This limit corresponds to the nonlocal spectroscopy of the Shiba state [$V_B < -1.2$ V region on Fig. 2(a)]. As the conductance of the right arm is increased, the amplitude of the SPT processes decreases while the CPS one strongly increases. When the barrier to the right lead is transparent, about 95% of the nonlocal signal comes from CPS processes. This is the usual Cooper pair splitter limit [$V_B > -1.1$ V region on Figs. 2(a) and 2(b)].

In between the two limits, there is a region where a significant contribution of the nonlocal signal comes from SPT processes, and this is indicated by the blue shaded region. In this region, the usual splitting efficiencies, which assume that the nonlocal signal comes only from the splitting of Cooper pairs, overestimate the real contribution of CPS processes.

Our results can be generalized to the conventional Cooper pair splitter geometry, i.e., the N–QD–SC–QD–N setup, in a straightforward way. The strongly coupled QD–SC–QD system hosting Andreev or Shiba molecular states was recently
studied in Refs. [95] and [96]. Similarly to the presented model, in these systems the ground state can be either a unique singlet or a twofold degenerate doublet and currents can flow to the normal leads when all of these three states are degenerate. Therefore, the transport processes illustrated in Fig. 3(c) and the partitioning of the currents, presented in Eqs. (10)–(12), also applies to the conventional splitter setup. The transport data measured in such a setup can be evaluated analogously to obtain the microscopic parameters and to partition the currents, and only the transition matrix elements have to derived from the N–QD–SC–QD–N setup’s Hamiltonian.

In conclusion, close to the quenched regime a significant contribution of the nonlocal signal comes from the SPT processes, but in the transparent barrier limit the nonlocal signal provides a good measure of the CPS processes.

VI. CONCLUSIONS

We have investigated the nonlocal signal of a Cooper pair splitter as the tunnel coupling to one of the normal leads was varied. It was shown that the nonlocal signal persists even upon quenching the conductance in the opposite arm of the splitter, contradicting the expectations since in this limit the CPS process is forbidden. We resolved the contradiction by considering the Shiba state, originating from the hybridization of the quantum dot and the superconductor states. In the quenched limit, the nonlocal signal is attributed to the largely extended zero-energy Shiba state, which we studied in detail in Ref. [37].

The unexpected nonlocal signal we observed implied that figures of merit for Cooper pair splitting efficiencies used in previous experimental works could not be applied for our device. Therefore, we constructed a transport model using the ZBA for the superconductor to reproduce our experimental findings and to separate the Shiba related nonlocal signal from the CPS. Despite of its simplicity, the model captures well the main features of the experiments. Utilizing the model and the obtained microscopic parameters we showed that closed to the quenched region, the nonlocal signal mostly consists of the SPT process, and therefore, the previously applied efficiency indicators overestimate the real CPS contribution. However, when the tuned barrier is transparent, the nonlocal signal is dominated by the CPS process, and accordingly, our results are consistent with those based on the standard CPS efficiency indicators.

The data that underlie the plots within this paper and other findings of this study are available in Ref. [97].

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