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Excitations in a superconducting Coulombic energy gap

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Cooper pairing and Coulomb repulsion are antagonists, producing distinct energy gaps in superconductors and Mott insulators. When a superconductor exchanges unpaired electrons with a quantum dot, its gap is populated by a pair of electron–hole symmetric Yu-Shiba-Rusinov excitations between doublet and singlet many-body states. The fate of these excitations in the presence of a strong Coulomb repulsion in the superconductor is unknown, but of importance in applications such as topological superconducting qubits and multi-channel impurity models. Here we couple a quantum dot to a superconducting island with a tunable Coulomb repulsion. We show that a strong Coulomb repulsion changes the singlet many-body state into a two-body state. It also breaks the electron–hole energy symmetry of the excitations, which thereby lose their Yu-Shiba-Rusinov character.
n a large superconductor, an adsorbed spin impurity binds to a quasiparticle screening cloud to form a state known as the Yu-Shiba-Rusinov (YSR) singlet, whose excitation energy with respect to the unbound doublet is below the superconducting energy gap, $\Delta$. The miniaturization of the superconductor into an island reduces charge screening and introduces an energy gap for the addition of electrons, the Coulomb repulsion, $E_c$ (see Fig. 1a)\textsuperscript{2,3}, with yet unexplored consequences on the ground state and the subgap spectrum. Such exploration is of relevance in the study of magnetic impurities adsorbed to superconducting droplets\textsuperscript{4,5}, in quantum-dot (QD) readout of Majorana qubits based on superconducting islands\textsuperscript{6-8}, and in realizations of superconducting variants of the multichannel Kondo model\textsuperscript{9-11}. In the absence of a spin impurity, the charging of a superconducting island (SI) depends on the ratio $E_c/\Delta$, with $E_c/\Delta < 1$ leading to Cooper pair $(2e)$ charging and $E_c/\Delta > 1$ to $1e$ charging\textsuperscript{2,3}. In the latter case, even numbers of electrons condense as Cooper pairs, while a possible odd numbered extra electron must exist as an unpaired quasiparticle\textsuperscript{3}. Here we provide the first spectral evidence of the many-body excitations in a superconducting Coulomb gap. The spin impurity resides in a gate-defined QD in an InAs nanowire, and the SI is an Al crystal grown on the nanowire with gate-tunable Coulomb repulsion. Both QD-SI and SI-QD-SI devices are investigated in this work. We demonstrate that a strong Coulomb repulsion forces exactly one quasiparticle in the SI to bind with the spin of the QD in the singlet ground state (GS). The Coulomb repulsion also enforces a positive-negative bias asymmetry in the position of the excitation peaks which is uncharacteristic of YSR excitations.

**Results**

**Excitations in a quantum dot coupled to a superconducting island.** Figure 1b–e summarize the energy dispersions which can arise when a QD is coupled to a superconductor. In Fig. 1b, the usual YSR case ($E_c = 0$) with the QD gate-induced charge tuned to $\nu = 1$ is depicted. The doublet GS and singlet excited state energies are independent of the gate-induced charge in the superconductor, $n_\text{QD}$, and excitations between these two states are electron-hole symmetric\textsuperscript{12}. As shown in Fig. 1c, introducing $E_c > 0$ in the superconductor produces a parabolic dispersion distorted by the hybridization ($\Gamma$) between the QD and the SI, which couples states of the same total charge. For odd $n_\text{QD}$, the energy of the doublet state is increased by $\nu E_c$ (green dot), while for even $n_\text{QD}$ it is the energy of the singlet state which is penalized by this amount. For odd $n_\text{QD}$ and $E_c > \Delta$, the GS is a singlet even if $\Gamma \rightarrow 0$. For $E_c < \Delta$, the singlet can be the GS if the YSR binding energy $E_B$ is large enough so that $E_c > \Delta - E_B$, which is achieved by increasing $\Gamma$\textsuperscript{13}.

Due to $U > 0$, the dispersion against $\nu$ is approximately parabolic in both the $E_c = 0$ (up to a constant) and $E_c > 0$ cases, as shown in Fig. 1d, e. For $E_c = 0$ (Fig. 1d), the electron and hole excitations are symmetric due to the degeneracy of the even-parity parabolas. This ceases to be the case for $E_c > 0$ (Fig. 1e). The asymmetry is maximal in the absence of additional QD levels. For $\nu > 1$ ($\nu < 1$), an extra electron (hole) must be stored in the SI with excitation energy $\Delta + E_c$, but an extra hole (electron) can be added to either the QD or the SI, leading to a superposition of states with excitation energies $- (\epsilon_\text{QD} + U)$ and $-(\Delta + E_c)$, where $\epsilon_\text{QD}$ is the energy level of the QD (details on Supplementary Fig. 1). The extra electron or hole either forms a quasiparticle or a Cooper pair, depending on the parity of the SI occupation of the initial state. Superconducting Coulombic excitations (SCE) are only symmetric at the special gate points where the excited parabolas cross each other. Our QD-SI device (Fig. 2a, b) is modeled as in the scheme shown in Fig. 2c\textsuperscript{13}. The SI is conceived as several hundreds of electronic levels. Its charge is tuned by $n_\text{QD}$, equivalent to top gate voltage $V_\text{G}$ in the device. The corresponding Hamiltonian includes pairing between time-reversed states to produce the superconducting gap, $\Delta$, and coupling to the QD, $\Gamma$, which is tuned by top gate voltage $V_\text{G}$ in the device. We consider constant Coulomb interactions $U$ for the QD, $E_c$ for the SI, and $V$ for the interdot charging due to the QD-SI inter-capacitance, $C_\text{m}$ (as in usual double QDs\textsuperscript{14}). The QD is itself modeled as an Anderson impurity, whose charge is tuned by $\nu$, equivalent to top gate voltage $V_\text{G}$. Other top gates ($V_1$, $V_2$) control the couplings of the QD and SI to the source and drain, not included in the model. The output of the model is the energy spectrum of the system, consisting of a few low-lying many-body states and the edge of the continuum. These states are sketched in Fig. 2d between the source and drain. Table 1 shows device and model parameters.

To record the spectrum of excitations between the low-lying many-body states, the device is biased by a source-drain bias...
trivial bias symmetry. A peak at one polarity thus demonstrates polarity conventions. Symmetric barriers would instead result in a
argument is necessary to account for the voltage drops and
dshown.
parabolas in e.g. Fig.1e, and the continuum (cyan bands), are coupled by GS (dashed line) and ±1 excited states (solid lines) corresponding to the excitation energies at eV.

The capacitances and voltages of top gates 1, 3 and 5 are not shown. In a asymmetry of the SCE. While we cannot account for the

The zero-bias G signal exhibits a strong dependence on Vc and VcN, as shown in the diagram of Fig. 3a. Singlet→doublet GS transitions are observed in the experiment when conductance lines are crossed, as at Vc, these lines appear when the nGS and nGS + 1 (or nGS − 1) states in Fig. 2d are degenerate at zero energy. The repetition of the central hexagonal charge domain in the Vc direction indicates filling of the SI. As a guide of the filling of the QD and the SI, we approximate their charge expectation values as integers nGS, nS in each of the charge domains. This is an approximation as only nGS is integer with nGS = nN + nS (see Supplementary Fig. 1). Small but resolvable 1,1 singlet domains (an example is enclosed in a dotted line) are seen between the 1,2 and 1,0 doublet domains. In contrast, the lines to the sides of the central hexagonal domains, which separate the 0,0 and 0,2 domains and the 2,0 and 2,2 domains, show no splitting at this resolution. The difference stems from finite Γ and V, which stabilize the 1,1 but not the 0,1 and 2,1 domains. The presence of the 1,1 Coulomb-aided YSR singlet is the key difference from a trivial double QD stability diagram and from the Δ = 0 case. For instance, a raise of the interdot coupling in a double QD introduces molecular orbitals which show as avoided crossings at triple points (TPs), whereas in the QD-SI system the YSR singlet is a many-body state for these parameters. Finite Γ and V are also responsible for increasing the distance between the points of multiple degeneracy, for the acute angle between vertical and horizontal conductance lines and, in the case of Γ, for curving the conductance lines.

Our model of the system produces a diagram of GS transitions of the SCE that matches the gate position of the conductance lines, as shown in the comparison of the calculation to the experimental data in Fig. 3a. The quality of the match for model parameters approximately similar to the experimentally measured values (with Δ as the only fit parameter) constitutes a first proof of the presence of SCE in our device.

We corroborate the spin (S) assignment done in Fig. 3a (right panel) at B = 0 from the variation of GS domain sizes with B = 0.3 T (in inset). Doublet domains are stabilized by B more than singlet domains, while triplet domains are stabilized further than doublet and singlet domains. The model fits the data using the g-factors as free parameters, and taking into account the GS transition from singlet to triplet in the 1,1 charge sector (charge parabolas are shown in Supplementary Fig. 3). The g-factors in the Hamiltonian are significantly larger than the measured effective g-factors (see Table 2). These bare g factors produce Zeeman splittings E0,QQ = g0μBμB and E0,SI = g0μNμB in the QD and the SI, where μB is the Bohr magneton. The effective and bare g factors would be equal if the expectation values of the QD and the SI charges increased in steps of exactly 1e across the GS transition lines. For non-zero Γ, (non-integer) charge distributions occur between the QD and the SI on either side of the GS

| Γ (meV) | U (meV) | E0,QQ (meV) | Δ (meV) | V (meV) |
|---------|---------|-------------|---------|---------|
| 0.05    | 0.8 -1.0| 0.19        | ≤0.27   | 0.13    |
| 0.04    | 0.8     | 0.18        | 0.2     | 0.16    |
transition line (see Supplementary Fig. 1), hence the effective $g$-factors are some non-trivial function of the true (bare) $g$-factors which appear in the Hamiltonian.13

Following this comprehensive mapping, we show in Fig. 3b the $G$ spectrum at finite $V_{sd}$ versus $V_S$ for fixed $V_N$, at which the SI contains only Cooper pairs in the GS up to a good approximation. The SCE have a double-$S$ shape, spanning $V_{sd} = -0.37 \rightarrow 0.37$ mV. They are approximately inversion symmetric in position and in $G$ intensity with respect to the electron-hole gate-symmetric filling point of the QD, which corresponds to the center of the 1,0 sector (indicated by a cross), from where removing/adding an electron from/to the QD are equally energetically unfavorable. $G$ jumps in intensity when the SCE cross zero bias, as highlighted by the insert traces at gate points before (gray) and after (black) one of such changes. While the SCE are expected to appear as a pair at asymmetric positive and negative bias positions for a given gate voltage, in practice only one SCE is observed. A GS change brings discontinuously up to the continuum the other state, as charge is suddenly redistributed between the QD and the SI.13

Our model reproduces the position of the subgap resonances, as evidenced in the overlay of the calculated spectrum on the experimental data in Fig. 3b (see also Supplementary Figs. 4–8 and Supplementary Note 2). Differences between the SCE spectrum and the spectrum in the Coulombic ($\Delta = 0$) and Yu-Shiba-Rusinov ($E_S = 0$) limits are shown in Supplementary Fig. 9. The Coulombic spectrum bears resemblance to that of an impurity in the paramagnetic Mott insulator described by the Hubbard model,16,17 despite the differences in the Hamiltonian (local Hubbard interaction versus constant Coulomb repulsion in our model). In both cases the charge transfer from the impurity site to the bath costs energy corresponding to the total charge gap of the system in the absence of the impurity ($\approx U/2$ in the Hubbard model at half-filling, $E_S + \Delta$ in our device), and in both cases there is a (quasi)continuum of fermionic states extending above this gap (doublons/holons in a Mott insulator, and Coulomb quasiparticles with a mixed character of Bogoliubov quasiparticles due to $\Delta$ in our device), leading to the same phenomenology.
Dependence of the singlet domain size on the Coulomb repulsion in the superconductor. To map these limits, we vary continuously the Coulomb repulsion in the superconductor in a second device. We first explore the role of the Coulomb repulsion on the stability of the YSR singlet as the GS. To this aim, we define two quantities, $x = 1 - \Delta/E_c$ and $y$, the YSR singlet GS size in units of $e$. In the $\Gamma/U < 1$ regime, $y = (E_c - \Delta + E_0)/E_c$. Fig. 4a, b explain how $x$ and $y$ are experimentally extracted. In the limits when $E_c \to 0$ and $E_c \to \infty$, $x \to \infty$ and $x \to 1$, respectively. When $E_c = \Delta$, then $x = 0$. Figure 4c shows a measurement of $y$ versus $x$ in a device consisting of a QD coupled to two SIs with hybridization $\Gamma_L$ and $\Gamma_R$ (top inset in Fig. 4c). The SIs have charging energies $E_{cL}$ and $E_{cR}$ and superconducting gaps $\Delta_L$ and $\Delta_R$, and their occupations are tuned with top gate voltages $V_{cL}$ and $V_{cR}$. The advantage of this three-component device over the two-component one is that the presence of only one QD between the two SIs can be verified from stability diagrams similar to that in Fig. 3a against the pairs of gate voltages $(V_{cL}, V_{cSL})$ and $(V_{cL}, V_{cSR})$. In Fig. 4c, $y$ characterizes the GS stability of the YSR state formed by the binding of the spin to the quasiparticle cloud in the right SI, and $x = 1 - \Delta_R/E_{cR}$. To employ the device as this two-component system, the left SI is kept either as a cotunnelling probe for the doublet domain has a trivial linear dependence with a slope of 1 and with endpoints at $(0,0)$ and $(1,1)$, connected by a fitted solid line in the graph. In this regime, $x$ only stabilizes the YSR singlet as the GS. At the other extreme, at the largest $\Gamma_R/U$, $x$ stabilizes the doublet more strongly than the YSR singlet, reducing $y$. In between these two extremes, at $\Gamma_R/U = 0.3$, the behavior is intermediate. When $x \to 1$, $y$ converges to 1e independently of $\Gamma_R/U$, as $x$ stabilizes equally well the doublet and singlet states for even and odd gate-induced charges in the right SI. In the other limit, when $x \to \infty$, $y$ depends exclusively on $\Gamma_R/U$, as in the usual $E_{cR} = 0$ YSR regime.

Dependence of the shape of the excitations on the Coulomb repulsion in the superconductor. Next, we describe how the Coulomb repulsion in the superconductor affects the dispersion of the excitations and how this is related to changes in the stability diagram. In Fig. 5, we show the evolution of the excitations produced by one QD shell on the right SI over a wide range of $V_{cR}$, corresponding to a charge variation of $\pm 960$ electrons. In this range, $E_{cR}/\Delta_R$ goes from 0 to 1.71, as measured from Coulomb-diamond spectroscopy. The increase in $E_{cR}/\Delta_R$ is reflected on the stability diagram. In the usual $E_{cR} = 0$ YSR regime (Fig. 5a), the diagram shows two vertical dispersionless lines, and the spectrum consistently displays a YSR loop (for measurement details, see Methods). When the right SI enters into Coulomb blockade (Fig. 5b, $E_{cR}/\Delta_R = 0.36$), the lines in the stability diagram wiggle as interdot charging and tunneling effects enter into consideration. Consequently, the YSR loop in the spectrum gets skewed rightwards and increases its bias size as the energy gap includes now a Coulombic component. At $E_{cR}/\Delta_R = 0.75$ (Fig. 5c), the entrance of the 1,1 YSR singlet GS breaks the stability diagram into several domains, and the excitations adopt a double-S shape. At this setting, the 1,0 doublet domain has a $V_{cSR}$ size $(E_{cR} + \Delta_R + E_{cBR})/E_{cR} = 2$, where $E_{cBR}$ is the YSR binding energy of the spin in the QD to the quasiparticle cloud in the right SI. This results in a maximum of the bias size of the double-S shape excitation. From then on, an increase in $E_{cR}/\Delta_R$ in Fig. 5d–f reduces the energy of the doublet $\rightarrow$ singlet excitation and the double-S shaped feature shrinks in bias size, concomitantly with the stronger stabilization of the YSR 1,1 singlet GS in the stability diagram.

Discussion

Throughout this article, we provided compelling evidence for the existence of superconducting Coulombic subgap excitations arising from states bound to a semiconductor-superconductor interface, and we showed how these are related to the usual electron-hole symmetric Yu-Shiba-Rusinov excitations. On one
hand, we showed that a small Coulomb repulsion in the superconductor is enough to turn the excitations asymmetric in the polarity of the bias voltage. On the other hand, a strong Coulomb repulsion ($E_\text{c} \to \infty$) converts the YSR singlet many-body state into a two-body state formed by a spin in the QD and a single quasiparticle in the superconductor.

Though our model is successful at matching excitation energies, an extension which includes transport is needed to account for the magnitude of the conductance features and for their bias positions in devices with more symmetric source-drain barriers. The observation of current blockade in a regime of weaker $\Gamma$ hints at elastic cotunnelling as the transport mechanism in our QD-SI device (see Supplementary Note 2). The absence of zero-bias $G$ in the $\Delta > E_\text{c}$ regime (e.g. Supplementary Figs. 10, 11) indicates that Andreev reflection ($2e$ charge transfer) is not a transport mechanism in our devices in this regime. Due to charge transfer between the SI and the QD, the model indicates that an upwards reconsideration of bare $g$-factor values extracted from experimentally-determined $g$-factors of subgap excitations is needed to match the experimental results. Based on its success, the model can also inform on future developments, e.g. qubit and multi-channel devices which utilize the SI-QD-SI device, as outlined in ref. 19.

Given their tunability by gating and by design, our devices can be extended to realize general spin effects, with superconductivity providing an energy gap for resolving the associated excitations. Regular arrays of the demonstrated singlet dimer can be used to provide an energy gap for resolving the associated excitations. The system sketched in Fig. 1a may also be realized with magnetic adatoms when these chains are deposited on the surface, e.g. Pb on an InAs substrate, and probed with scanning tunneling microscopy. Several open questions could be answered with this technique: What is the spatial extension of the excitations in a superconducting Coulombic energy gap? Is there orbital structure in the excitations? How do the excitations behave in chains of magnetic adatoms when these chains are deposited on the surface?
top of an E_c > 0 SIs25 Do chains of magnetic adatoms deposited on finite E_c SIs support Majorana excitations26? Methods Devices fabrication and layout. QD-SI device (Fig. 2a). A 110-nm wide InAs nanowire with a micromanipulator was contacted by 5/200 nm Ti/Au (in yellow) source and drain leads. The 350-nm long 7-nm thick epitaxial Al SI covering three facets of the nanowire was defined by chemically etching the upper and lower sections of the nanowire before contacting. After insulating the nanowire and the leads with a 6-nm thick film of HfO2, five Ti/Au top gates were deposited along the nanowire. The QD was defined in the bare nanowire next to the SI by setting top gates 1 and 3 to negative voltage. A Si/300 nm-thick Al SI substrate backgate was kept at zero voltage throughout the experiment. SI-QD-SI device (Supplementary Fig. 10). The SI-QD-SI device was fabricated using a nanowire from the same growth batch. Two nominally identical 7-nm thick, ≈300-nm long, epitaxially-grown Al SIs were defined by chemical etching. The nanowire was contacted by 5/200 nm Ti/Au leads, and then insulating by a 5-nm-thick layer of HfO2 from seven Ti/Au top gates deposited after the QD was defined between the two SIs by setting top gate 3 and 5 to negative values. The substrate backgate was used to aid the top gates in depleting the device. E_f/E_d was tuned by using an auxiliary QD (QD_aux), defined between the right SI and the source lead. When QD_aux was put near resonance by sweeping V_g, E_f − ΔfH could be tuned to negative values, and when QD_aux was tuned to cotunneling, E_f − ΔfH could be tuned to positive values. Similarly, E_fL − ΔfL was tuned using an auxiliary QD defined between the left SI and the drain lead. The critical B of the superconducting Al film was measured to be B_c = 2.1 T in nanowire devices made from the same batch of nanowires used in the fabrication of the present device27,28, which left ample room for B-resolved measurements in the superconducting state. In the QD-SI and SI-QD-SI devices, the presence of superconductivity at large B was determined from size differences of adjacent charge domains with odd and even occupation of the SL, observed up to B = 1.2 T and B = 1.5 T, respectively. Larger B was not explored. Differential conductance measurements. A standard lock-in technique was used to measure the differential conductance, G = dI/dV, of the QD-SI device by biasing the source with an AC excitation of 5 μV at a frequency of 223 Hz on top of a DC source-drain bias voltage, V_g, and recording the resulting AC and DC currents on the ground drain lead. In the case of the SI-QD-SI device, G was measured at the ground drain with a 5 μV lock-in excitation applied at the source at 84.29 MHz. The measurements were performed in an Oxford Triton dilution refrigerator at 30 mK for the QD-SI device and 35 mK for the SI-QD-SI device, such that η_G ≈ η_fH, where η_0 is the Boltzmann constant and T is the refrigerator temperature. Calculation of subgap and continuum excitations. The calculations were done using the density-matrix renormalization group approach (details in Supplementary Note 3). The quantum numbers are the total number of electrons in the system, n, the z-component of the total spin, S_z (see Supplementary Fig. 3), and the index for states in a given (n, S_z) sector. l = 0, ±1, ±2,..., ±L, where L is the length of the wire. The superconducting Coulomb excitations are given by E = E_f ± ω_n ≈ ± ω_n = S/S_l ± 1/2 ± L, where the edges of the continuum excitations are given by E_{Edge} = E_f ± ω_n = S/S_l ± 1/2 ± L = 1. The finite size of the SI, the continuum in is truth only a quasicontinuum of states. The nature of these states and the excitation energies depend on the values of A and E_f. For A = 0, the quasiparticles are free-electron states. For A ≠ 0, these are Bogoliubov quasiparticles with pronounced interlevel pairing correlations (c_i^+ c_j^+ c_j c_i). If E_f = 0, the excitation spectrum is not affected by the number of preexisting particles in the superconductor (up to finite-size effects). If E_f ≠ 0, the particle-addition and particle-removal energies are affected by the charge repulsion (parabolas). The calculations do not provide direct results for the differential conductance of the system, only information about the energies of the GS and the low-lying excitations. Spectral measurements in Fig. 5. To obtain sharp spectral features visible over the continuum background, we tuned the left SI into a superconducting probe (E_fL = 0, η_fL = 0). The strong hybridization of the left SI with the drain needed to achieve E_fL = 0 resulted in an unintended soft gap in this probe, which produced faint replica of the main excitations. For example, in Fig. 5a, black dotted lines correspond to the YSR replica coming from the QD right SI being probed by the coherence peaks of the probe. The loop is thus followed by negative differential conductance (NDC) and appears at ±E_f = ±E_fL + E_f ± ΔE_f (n = n_fL ± S_fL ± S/S_l ± L/2 ± L), reaching ΔA at GS transitions. The gray dotted lines highlight a YSR replica probing the soft gap of the probe, thus an order of magnitude weaker in conductance and without associated NDC. This replica appears at ±E_c ± 1/2 ± L ± E_fL ± ΔfL ± S/S_l ± L/2 ± L ± S/S_l ± 1/2 ± L ± ΔfL ± S/S_l ± L/2 ± L, and therefore crosses zero bias at GS transitions. When E_fL/ΔfL increases, the relationships between the excitations and the bias positions of the conductance features become approximations due to non-ideal physics in the continuum at higher bias outside the scope of this work. Data availability The experimental data generated in this study have been deposited in the ERDA database of the University of Copenhagen at https://doi.org/10.17894/ucph.58ab2554-e476-4749-a241-bf2194d59516c. Received: 13 August 2021; Accepted: 28 March 2022; Published online: 26 April 2022
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Author contributions
J.C.E.S. and A.V. performed the experiments. P.K. and J.N. developed the nanowires. J.C.E.S., A.V., K.G.-R., J.N., L.P. and R.Z. interpreted the experimental data. L.P. and R.Z. did the theoretical analysis. J.C.E.S. wrote the manuscript with input from A.V., K.G.-R., J.N., L.P. and R.Z.

Competing interests
The authors declare no competing interests.

Additional information
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