Yu-Shiba-Rusinov screening of spins in double quantum dots

Grove-Rasmussen, Kasper; Steffensen, Gorm Ole; Jellinggaard, Anders Robert; Madsen, Morten Hannibal; Žitko, R.; Paaske, Jens; Nygård, Jesper

Published in:
Nature Communications

DOI:
10.1038/s41467-018-04683-x

Publication date:
2018

Document version
Publisher's PDF, also known as Version of record

Document license:
CC BY

Citation for published version (APA):
Grove-Rasmussen, K., Steffensen, G. O., Jellinggaard, A. R., Madsen, M. H., Žitko, R., Paaske, J., & Nygård, J. (2018). Yu-Shiba-Rusinov screening of spins in double quantum dots. Nature Communications, 9, [2376]. https://doi.org/10.1038/s41467-018-04683-x
Yu-Shiba-Rusinov screening of spins in double quantum dots

K. Grove-Rasmussen1, G. Steffensen1, A. Jellinggaard1, M.H. Madsen1, R. Žitko2,3, J. Paaske1 & J. Nygård1

A magnetic impurity coupled to a superconductor gives rise to a Yu-Shiba-Rusinov (YSR) state inside the superconducting energy gap. With increasing exchange coupling the excitation energy of this state eventually crosses zero and the system switches to a YSR ground state with bound quasiparticles screening the impurity spin by $\hbar/2$. Here we explore indium arsenide (InAs) nanowire double quantum dots tunnel coupled to a superconductor and demonstrate YSR screening of spin-1/2 and spin-1 states. Gating the double dot through nine different charge states, we show that the honeycomb pattern of zero-bias conductance peaks, archetypal of double dots coupled to normal leads, is replaced by lines of zero-energy YSR states. These enclose regions of YSR-screened dot spins displaying distinctive spectral features, and their characteristic shape and topology change markedly with tunnel coupling strengths. We find excellent agreement with a simple zero-bandwidth approximation, and with numerical renormalization group calculations for the two-orbital Anderson model.
Yu–Shiba–Rusinov (YSR) states\textsuperscript{1–3} can be imaged in a direct manner by scanning-tunneling spectroscopy of magnetic adatoms on the surface of a superconductor\textsuperscript{4}. Using superconducting tips, high-resolution bias spectroscopy of multiple sub-gap peaks reveals an impressive amount of atomic detail like higher angular momentum scattering channels, crystal-field splitting and magnetic anisotropy\textsuperscript{4–11}. In general, however, it can be an arduous task to model the complex pattern of sub-gap states\textsuperscript{6,7,9,19} let alone to calculate their precise influence on the conductance\textsuperscript{10}.

In contrast, the “atomic physics” of Coulomb blockaded quantum dots (QDs) is simple. Changing the gate voltage, subsequent levels are filled one-by-one and the different charge states alternate in spin, or Kramers degeneracies for dots with spin–orbit coupling, between singlet and doublet. With normal metal leads, charge states with spin-1/2 exhibit zero-bias Kondo resonances at temperatures below the Kondo temperature, \( T \ll T_K \), reflecting a Kondo-screened singlet ground state (GS). If the leads are superconducting with a large BCS gap, \( \Delta \gg k_B T_K \), this resonance is quenched and the GS recovers its doublet degeneracy. The system now displays a YSR singlet excitation close to the gap edge, which can be lowered in energy by increasing the \( k_B T_K/\Delta \textsuperscript{12–17} \). Close to \( k_B T_K = 0.3\Delta \) it crosses zero and becomes the YSR-screened singlet GS\textsuperscript{9,11,18–20}, which eventually crosses over to a Kondo singlet at \( k_B T_K \gg \Delta \).

YSR states were first discussed in the context of gapless superconductivity arising in the presence of randomly distributed paramagnetic impurities\textsuperscript{1–3}. However, the ability to assemble spins into dimers, chains, and lattices, has recently prompted the exciting idea of engineering YSR molecular\textsuperscript{20,21}, YSR sub-gap topological superconductors and spiral magnetic states\textsuperscript{4,22–24}. QDs have the advantage of being tunable via electrical gates, and this plays an important role in recent proposals for topological superconductivity in systems of coupled QDs\textsuperscript{25–27}.

Here we utilize this electrical control to manipulate YSR states in a double quantum dot (DQD) formed in an InAs nanowire. Using multiple finger gates to tune the total DQD spin and the interdot coupling, we demonstrate control of the YSR phase diagram, including electrical tuning between YSR singlets, and a novel YSR doublet arising from the screening of an excited spin triplet.

### Results

**Device and model.** A scanning electron micrograph of an actual device (Device A) is shown in Fig. 1a, where bottom gates are used to define a normal (N)-DQD-superconductor (S) structure\textsuperscript{11}. The corresponding schematic is shown in Fig. 1b, where plunger gates labeled \( gN \) and \( gS \) control left (QDN) and right (QDS) quantum dot, respectively, while an auxiliary gate, \( g_d \), tunes the interdot tunneling barrier. The essential physics of this system can be understood in terms of a simple zero-bandwidth (ZBW) model in which the superconductor is modeled by a single quasiparticle coupled directly to an orbital in QDS via \( t_d \) and indirectly to QDN through \( t_N \). A normal metal electrode with weak coupling \( t_N \) and correspondingly low Kondo temperature to the left dot is used to probe the DQD-S system. Figure 1c shows the corresponding energy diagram in the regime of dominating on-site charging energies. In the Supplementary Note 3 we compare this model to numerical renormalization group (NRG) calculations to establish its reliability as a quantitative tool.

In Fig. 2a, b, we reproduce the well-known sub-gap state behavior for a single dot coupled to a superconductor within the ZBW model. The panels show excitation energy as a function of the dimensionless gate voltage \( \bar{\nu} \) (corresponding to the noninteracting average occupation of QD\textsubscript{S}) for weak and strong \( t_d \). As expected the sub-gap excitations cross (do not cross) zero energy for weak (strong) coupling. The GS of the system for odd occupancy is thus a doublet or a YSR singlet (screened spin)\textsuperscript{13,17,18}.

In Fig. 2f–i we extend the ZBW model to a DQD (finite \( t_d \)) and calculate stability diagrams for increasing \( t_d \). For weak coupling the characteristic honeycomb pattern is observed similar to DQD in the normal state\textsuperscript{28–30}. However, as \( t_d \) increases, entirely new types of stability diagrams emerge. In Fig. 2g, the pattern resembles two mirrored arcs, where the lack of zero-energy excitations as a function of \( \bar{\nu} \) for even occupation of QD\textsubscript{N} is due to a doublet to singlet transition in QD\textsubscript{S} (see Fig. 2a, b). Moreover, as the coupling increases even further, the GS in the \( (\bar{\nu}_N, \bar{\nu}_S) = (11) \) region becomes a YSR doublet altering the stability diagram to vertically shaped rectangular regions.

To understand this behavior, we show the states of the system in the (11) region in Fig. 2c. Two electrons in the DQD may form either a singlet \( S_{11} \) or a triplet \( T_{11} \) state with energy splitting \( J_0 \).
Due to the superconductor, a third state may also exist in the gap. In analogy with the QD-S system, where a doublet state may be screened to a YSR singlet, the triplet state may be screened to a YSR doublet, which becomes the ground state (GS) for large enough \( t_d \). The energy of the YSR-singlet GS is the ground state (GS) for odd occupancy, \( \tilde{N}_S \), and the YSR-triplet GS is the ground state for even occupancy, \( \tilde{N}_S \). The relevant GSs and corresponding regimes with two GS transitions in the \( (t_S, t_d) \)-plane of the DQD-S system is shown in Fig. 2f, g. The first occurs when the system transitions from a honeycomb pattern to the case where the spin in QDS is delocalized, \( (t_S, t_d) \). The second transition happens when the \( D_{YSR} \), which becomes the GS for \( \tilde{N}_S \) = 1, changes to the YSR regime, \( \tilde{N}_S \), and the latter eventually becomes the GS at strong coupling.

The relevant GSs and corresponding regimes with two GS transitions in the \( (t_S, t_d) \)-plane of the DQD-S system is shown in Fig. 2f, g. The first occurs when the system transitions from a honeycomb pattern to the case where the spin in QDS is delocalized, \( (t_S, t_d) \). The second transition happens when the \( D_{YSR} \), which becomes the GS for \( \tilde{N}_S \) = 1, changes to the YSR regime, \( \tilde{N}_S \), and the latter eventually becomes the GS at strong coupling.

**Fig. 2** YSR phase diagrams. a, b Two distinct YSR sub-gap spectra vs. occupation \( \tilde{N}_S \) of QDS for \( \tilde{N}_S = 0 \) (single dot case). For weak and strong coupling to the superconductor, the doublet \( D \) (a) and YSR singlet \( S_{YSR} \) (b) is the ground state for odd occupancy, respectively. c ZBW model calculation in the (11) charge state, with singlet \( S_1 \) and triplet \( T_1 \) separated by the interdot exchange energy \( J_d \). The triplet is YSR-screened and gives rise to a sub-gap YSR doublet \( D_{YSR} \), which becomes the ground state (GS) for large enough \( t_d \). d, e Double-dot YSR phase diagram hosting three regimes (e), honeycomb (HC), partly screened (PS) and screened (SC). The ground states in the (01) and (21), and the (11) charge regions for the three regimes are shown in d.

**Measurements.** With the qualitative behavior of this system in place, we explore the different regimes experimentally. The honeycomb regime is presented in the Supplementary Note 2 (Device B), while below we focus on the stronger coupled regimes. Figure 3a shows linear conductance versus plunger gates for a two-orbital DQD shell (i.e., one spin-degenerate level in each dot). A pattern of two arcs is observed, resembling the PS regime. To verify that the conductance resonances originate from sub-gap states, gate traces for different fillings of the two dots are measured. Figure 3c–e trace out the filling of electrons in QD\(_N\) along the red arrows in Fig. 3a, keeping the electron number in QD\(_S\) constant. The sub-gap spectroscopy plots c,e for even filling of QD\(_S\) show similar behavior, differing from d with odd occupation. When fixing (sweeping) the occupation of QD\(_N\) (QD\(_S\)), the qualitative behavior is switched (Fig. 3f–h corresponding to green arrows in Fig. 3a). For even occupancy in QD\(_N\) (1h) no zero-bias crossing is observed, while the opposite is true for odd occupancy (g). In particular Fig. 3d, g are interesting, since they involve the (11) charge state region. In contrast to single dot systems, the singlet GS shows different behavior whether tuning the electrochemical potential of the dot close to the superconductor or the normal lead, i.e., concave and convex excitation behavior versus gate voltage in the (11) state. The experimental data clearly confirm that the resonances in the stability diagram originate from sub-gap excitations. The stability diagram generated by our DQD-S model for realistic parameters reproducing the experimental behavior is shown in Fig. 3b, and corresponding gate traces for fixed occupations are shown in Fig. 3i–n. The qualitative agreement between theory and experiment is striking and even subtleties like the asymmetry of the sub-gap resonance splitting in j (see arrows) are reproduced.
The transition between different YSR states can also be driven by changing the singlet-triplet ($S_{11}$–$T_{11}$) splitting by tuning $t_d$ (cf. Fig. 2c). In Fig. 4c–h we show calculated diagrams for $t_d$ in a parameter range, where the GS in the (11) charge state transitions from $S_{11}$ ($h$) to $D_{YSR}$ ($e$), i.e. from double-arc, to vertical-lines diagrams. The corresponding measured stability diagrams for the two orbitals analyzed in Fig. 3 are shown in Fig. 4a–d, where the gate voltage between the two dots are tuned to more negative values (decreasing $t_d$). The effect of this tuning qualitatively follows the expectation of the model: a transition from $S_{11}$ to $D_{YSR}$ in the screened regime where all spin states are YSR screened.

The gate-dispersion of sub-gap excitations also shows good overall correspondence between ZBW modeling and experiment. We measured the sub-gap spectra in the screened regime along the red and green arrows in c and a. The first case c is almost at the transition, where the singlet and doublet states are degenerate in (11). Figure 4i, j shows sub-gap states versus $V_{gD}$ and $V_{gB}$, respectively, with a zero-bias peak at $V_{gS} = 2.62\, \text{V}$ reflecting a degeneracy at this value of $t_d$. The corresponding ZBW modeling in Fig. 4k, l (for $t_d = 0.25\, \text{meV}$) places the system just barely in the screened regime with a $D_{YSR}$ (11) GS and a nearby $S_{11}$ excitation dispersing very much like in the measurement. A $T_{11}$ triplet state is predicted inside the gap, and should be accessible from the $D_{YSR}$ GS. As demonstrated in the Supplementary Note 3, this is confirmed by more accurate NRG calculations, which however reveal a strong suppression of spectral weight on this state, explaining why it may be difficult to observe in experiment. For even lower $t_d$, case a, Fig. 4m–p again show good overall correspondence between experiment and theory, except for the triplet state, which should be weak, and in this case hardly resolved within the linewidth broadening in the data. Future experiments with hard gap superconductors or improved resolution may eventually lead to capability to detect even such low-weight spectral features. A detailed discussion of nonlinear conductance and broadening of the YSR sub-gap spectra is provided in the Supplementary Note 4. In particular, the electron–hole (e–h) asymmetry of the sub-gap resonance amplitude in, e.g., Fig. 3f is due to relaxation from the sub-gap state to quasiparticles above the gap (i.e. in the case of no relaxation the sub-gap resonance amplitude is expected to be e–h symmetric).

### Methods

**Fabrication.** The devices are made by defining bottom gate Au/Ti (12/5 nm) electrodes (pitch 55 nm) on a silicon substrate capped with 500 nm SiO$_2$ followed by atomic layer deposition of 3 × 8 nm HfO$_2$. InAs nanowires (70 nm in diameter) appropriately aligned on bottom gate structures are contacted by Au/Ti (90/5 nm) normal and Au/Ti (95/5 nm) superconducting electrodes separated by ~350 nm. The superconducting film has a critical field of around 85 mT.

**Measurements techniques.** The samples are mounted in an Oxford Instruments Triton 200 dilution refrigerator with base temperature of around 30 mK and are measured with standard lockin techniques. For the data (Figs. 3 and 4) shown in the PS regime, the voltages on the gates define a double dot potential with values (V) $V_{gD} = 0, V_{gB} = -1.3, V_{gS} = -0.3, V_{g5} = 2.65, V_{g6} = -0.3$, and $V_{g7} = 0.3$. Here the gate numbers correspond to gates from left to right in Fig. 1a. Gates 3, 4, and 5 are thus the left plunger, the tunnel barrier and right plunger gates, which are tuned within some range of the values stated.

**Data availability.** The data presented above can be found at the following [link](https://sid.erda.dk/public/archives/ec32617f4b179926cb9343ce4e650b11/published-archive.html).
Fig. 4 Tuning interdot coupling $t_d$. a–d Stability diagrams for different voltages on the tuning gate $g_d$, while compensating on plunger gates ($T = 30 \text{ mK}$). The plot in d is analyzed in Fig. 3 and the voltage on $g_d$ is decreased in steps of $-20 \text{ mV}$ from d to a (compensating in steps of 10 and $-15 \text{ mV}$ on $g_N$ and $g_s$, respectively). e–h, Stability diagrams generated by the zero-bandwidth model for different $t_d$ (in meV), qualitatively reproducing the experimental behavior in a–d. Orange regions marking ground state transition have singlet-doublet splitting less than 0.015 meV. i, j, m, n Bias spectroscopy of sub-gap states vs. individual plunger gates swept along red, and green arrows in c and a. All plots show clear sub-gap resonances consistent with the ground states (doublet $D$ or singlet $S$) for different gate voltages indicated below. k, l, o, p Zero-bandwidth model calculation of sub-gap excitations for $t_d = 0.2 \text{ meV}$ and $t_d = 0.125 \text{ meV}$ corresponding to experimental plots i, j, m, n. The triplet excitation has very low spectral weight and therefore does not show up in the measured bias spectroscopy. Parameters are fixed to the same values as in Fig. 3.

Received: 9 November 2017 Accepted: 17 May 2018
Published online: 18 June 2018

References
1. Yu, L. Bound state in superconductors with paramagnetic impurities. Acta Phys. Sin. 21, 75–91 (1965).
2. Shiba, H. Classical spins in superconductors. Prog. Theor. Phys. 40, 435–451 (1968).
3. Rusinov, A. I. Superconductivity near a paramagnetic impurity. JETP Lett. 9, 85–87 (1969). [Zh. Eksp. Teor. Fiz. 9, 146 (1968)].
4. Heinrich, B. W., Pascual, J. I. & Franke, K. J. Single magnetic adsorbates on s-wave superconductors. Prog. Surf. Sci. 93, 1–19 (2018).
5. Yazdani, A., Jones, B. A., Lutz, C. P., Crommie, M. F. & Eigler, D. M. Probing the local effects of magnetic impurities on superconductivity. Science 275, 1767–1770 (1997).
6. Ji, S.-H. et al. High-resolution scanning tunneling spectroscopy of magnetic impurity induced bound states in the superconducting gap of Pb thin films. Phys. Rev. Lett. 100, 226801 (2008).
7. Rudy, M., Peng, Y., von Oppen, F., Heinrich, B. W. & Franke, K. J. Orbital picture of Yu–Shiba–Rusinov Multiplets. Phys. Rev. Lett. 117, 186801 (2016).
8. Franke, K. J., Schulze, G. & Pascual, J. I. Competition of superconducting phenomena and Kondo screening at the nanoscale. Science 332, 940–944 (2011).
9. Hatter, N., Heinrich, B. W., Ruby, M., Pascual, J. I. & Franke, K. J. Magnetic anisotropy in Shiba bound states across a quantum phase transition. Nat. Commun. 6, 8988 (2015).
10. Ruby, M. et al. Tunneling processes into localized subgap states in superconductors. Phys. Rev. Lett. 115, 087001 (2015).
11. Žitko, R., Bodensiek, O. & Pruschke, T. Effects of magnetic anisotropy on the subgap excitations induced by quantum impurities in a superconducting host. Phys. Rev. B 90, 205440 (2014).
12. Pillet, J.-D. et al. Andreev bound states in supercurrent-carrying carbon nanotubes revealed. Nat. Phys. 6, 965–969 (2010).
13. Deacon, R. S. et al. Tunneling spectroscopy of andreev energy levels in a quantum dot coupled to a superconductor. Phys. Rev. Lett. 104, 076805 (2010).
14. Grove-Rasmussen, K. et al. Superconductivity-enhanced bias spectroscopy in carbon nanotube quantum dots. Phys. Rev. B 79, 134518 (2009).
15. Lee, E. J. H. et al. Spin-resolved Andreev levels and parity crossings in hybrid superconductor-semiconductor nanostructures. Nat. Nanotechnol. 9, 79–84 (2014).
16. Chang, W., Manucharyan, V. E., Jespersen, T. S., Nygård, J. & Marcus, C. M. Tunneling spectroscopy of quasiparticle bound states in a spinful Josephson junction. Phys. Rev. Lett. 110, 217005 (2013).
17. Jellinggaard, A., Grove-Rasmussen, K., Madsen, M. H. & Nygård, J. Tuning Yu–Shiba–Rusinov states in a quantum dot. Phys. Rev. B 94, 064520 (2016).
18. Satori, K., Shiba, H., Sakai, O. & Shimizu, Y. Numerical renormalization group study of magnetic impurities in superconductors. J. Phys. Soc. Jpn. 61, 3239–3254 (1992).
19. Bauer, J., Oguri, A. & Hewson, A. C. Spectral properties of locally correlated electrons in a Bardeen–Cooper–Schrieffer superconductor. J. Phys.: Cond. Mat. 19, 486211–486230 (2007).
20. Yao, N. Y. et al. Phase diagram and excitations of a Shiba molecule. Phys. Rev. B 80, 241108 (2014).
21. Kezulebieke, S., Dvorak, M., Ojansen, T. & Liljeroth, P. Coupled Yu–Shiba–Rusinov states in molecular dimers on NbSe2. Nano. Lett. 18, 2311–2315 (2018).
22. Nady-Perge, S. et al. Observation of Majorana fermions in ferromagnetic atomic chains on a superconductor. Science 346, 602–607 (2014).
23. Pierik, F., Glazman, L. I. & von Oppen, F. Topological superconducting phase in helical shiba chains. Phys. Rev. B 88, 155420 (2013).
24. Schecter, M., Flensberg, K., Christensen, M. H., Andersen, B. M. & Paaske, J. Self-organized topological superconductivity in a Yu–Shiba–Rusinov chain. Phys. Rev. B 93, 140503 (2016).
25. Fulga, I. C., Haim, A., Akhmerov, A. R. & Oreg, Y. Adaptive tuning of Majorana fermions in a quantum dot chain. New J. Phys. 15, 045020 (2013).
26. Su, Z. et al. Andreev molecules in semiconductor nanowire double quantum dots. Nat. Commun. 8, 585 (2017).
27. Sherman, D. et al. Normal, superconducting and topological regimes of hybrid double quantum dots. Nat. Nano 12, 212–217 (2017).
28. Jeong, H., Chang, A. M. & Melloch, M. R. The Kondo effect in an artificial quantum dot molecule. Science 293, 2221–2223 (2001).
29. van der Wiel, W. G. et al. Electron transport through double quantum dots. Rev. Mod. Phys. 75, 1–22 (2002).
30. Chorley, S. J. et al. Tunable Kondo physics in a carbon nanotube double quantum dot. Phys. Rev. Lett. 109, 156804 (2012).

Acknowledgements
We would like to thank M. C. Hels and C. B. Sørensen. Research was supported by the Center for Quantum Devices, The Danish National Research Foundation, Carlsberg Foundation, The Independent Research Fund Denmark (Natural Sciences), the FP7 FET-Open SE2ND project and the Slovenian Research Agency (ARRS) under Program P1-0044 and J1-7259.

Author contributions
K.G.-R. and A.J. performed the measurements, A.J. fabricated the devices and M.H.M. grew the nanowires. K.G.-R., A.J., and J.N. designed the double quantum dot experiments, G.S., K.G.-R., and J.P. made the ZBW analysis, G.S. and J.P. the conductance asymmetry calculations, and R.Z. provided the NRG analysis. K.G.-R., G.S., J.P., R.Z., and J.N. participated in discussions, analysis and wrote the paper.

Additional information
Supplementary Information accompanies this paper at https://doi.org/10.1038/s41467-018-04683-x.

Competing interests: The authors declare no competing interests.

Reprints and permission information is available online at http://npg.nature.com/reprintsandpermissions/

Publisher’s note: Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.