Scaling of subgap excitations in a superconductor-semiconductor nanowire quantum dot

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A quantum dot coupled to a superconducting contact provides a tunable artificial analog of a magnetic atom in a superconductor, a paradigmatic quantum impurity problem. We realize such a system with an InAs semiconductor nanowire contacted by an Al-based superconducting electrode. We use an additional normal-type contact as a weakly coupled tunnel probe to perform tunneling spectroscopy measurements of the elementary subgap excitations, known as Andreev bound states or Yu-Shiba-Rusinov (YSR) states. Indeed, theoretical proposals suggest that the bound states, known as Andreev levels or Yu-Shiba-Rusinov (YSR) states that appear in this limit [1–3], are precursors of a one-dimensional (1D) topological superconductor with zero-energy Majorana edge modes [4–11]. However, in spite of its importance, quantitative experimental studies of the S-coupled Anderson impurity remain scarce [12–15]. In particular, the scaling of Andreev levels with respect to the relevant physical parameters (e.g., the tunnel coupling $\Gamma_S$ between S and the impurity) has not yet been addressed. Here, we present a joint experimental-theoretical work aimed at filling this void. We exploit the versatility of semiconductor quantum dots (QDs), which effectively behave as quantum impurities, to investigate the scaling of Andreev levels in a direct manner, by tunneling spectroscopy. Our quantitative analysis is further supported by numerical renormalization group (NRG) calculations performed without fitting parameters, showing remarkable agreement with the measured data.

The ground state of the S-coupled Anderson impurity is defined in a competition involving the superconducting proximity effect, Coulomb blockade, and Kondo correlations. There are two possibilities: a magnetic doublet, enforced by strong Coulomb interactions, and a spin singlet, favored by strong coupling to S. Transitions between the ground state and the first excited state of the system, i.e., between a doublet and a singlet state, or vice versa, are manifested as a subgap Andreev level of energy $\zeta$, where the latter is equivalent to the excitation energy. Remarkably, the theory predicts that $\zeta$ scales with $\Gamma_S$, and that a quantum phase transition (QPT) between singlet and doublet ground states takes place when $\zeta$ changes sign ( signaled by the crossing of Andreev levels at zero energy) [16–24]. In this Rapid Communication, we employ tunneling spectroscopy to study the Andreev levels associated with a QD formed in hybrid superconductor-semiconductor nanowire structures. With the aid of a dual-gate device geometry, we are able to continuously tune $\Gamma_S$ while probing the same QD charge state. In this way, we demonstrate full electrical control over Andreev levels as well as over the singlet-doublet QPT. By further studying the evolution of Andreev levels in the parameter space, we obtain an experimental phase diagram of the system, and verify that the tuning of Andreev levels is consistent with the predicted scaling with the dimensionless ratio between the Kondo temperature and the superconducting gap $\Delta_K/\Delta$. We note that while a similar tuning of the QPT has been indirectly studied in the supercurrent behavior of QD-based Josephson junctions [13], here we provide a spectroscopical demonstration, which is also fully supported by numerically exact NRG simulations. Finally, we point out that the herein discussed formation of QDs in hybrid nanowire devices, and the sensitivity of device parameters to the local electrostatic environment, are relevant effects to be considered in experiments aimed at the detection of Majorana modes.

The device geometry adopted in this study is shown in Fig. 1(a), where N represents a normal metal tunnel probe weakly coupled to the QD. InAs/InP core/shell nanowires (NWs) [25] were randomly dispersed onto highly doped Si/SiO$_2$ substrates containing prepatterned local bottom gate arrays. Source and drain contacts were defined by e-beam lithography, followed by metal deposition and lift-off. The finalized devices contained a single local gate, later used as a plunger gate (pg), between the N (2.5 nm Ti/45 nm Au)
The nanowires are contacted by N and S leads comprising Ti/Au and Ti/Al bilayers, respectively. A Ti/Au thin strip covered by HfO₂ dielectrics acts as a local plunger gate (pg), whereas the degenerately doped substrate is employed as a global back gate (bg). (b) False color scanning electron micrograph of a typical device. (c) Qualitative phase diagram of the QD-S system in the wide-gap limit ($\Delta \rightarrow \infty$). The horizontal (vertical) line underscores QPTs between the singlet and doublet states (circles) that occur upon varying the QD level position (QD-S coupling). (d) Schematics of the Andreev level spectroscopy transport cycle. Current is measured across the N-QD-S device when the chemical potential of the tunnel probe ($\mu_{N}$) is aligned with an Andreev level at energies $\pm|\zeta|$. Transport occurs via Andreev reflection, whereby an injected electron (hole) is reflected back to N as a hole (electron), forming (breaking) a Cooper pair in S.

FIG. 1. (a) Schematics of the studied dual-gate N-QD-S devices. The nanowires are contacted by N and S leads comprising Ti/Au and Ti/Al bilayers, respectively. A Ti/Au thin strip covered by HfO₂ dielectrics acts as a local plunger gate (pg), whereas the degenerately doped substrate is employed as a global back gate (bg). (b) False color scanning electron micrograph of a typical device. (c) Qualitative phase diagram of the QD-S system in the wide-gap limit ($\Delta \rightarrow \infty$). The horizontal (vertical) line underscores QPTs between the singlet and doublet states (circles) that occur upon varying the QD level position (QD-S coupling). (d) Schematics of the Andreev level spectroscopy transport cycle. Current is measured across the N-QD-S device when the chemical potential of the tunnel probe ($\mu_{N}$) is aligned with an Andreev level at energies $\pm|\zeta|$. Transport occurs via Andreev reflection, whereby an injected electron (hole) is reflected back to N as a hole (electron), forming (breaking) a Cooper pair in S.
FIG. 2. (a) Fitting of normal-state linear conductance at $V_{bg} = 4.5$ V (black dots) with NRG model (red line). A reliable estimation of device parameters results from the fit (see text and Supplemental Material [34] for details). (b) Effect of $V_{bg}$ on the QD-S tunnel coupling, and (c) coupling asymmetry. The plots demonstrate a continuous back gate-induced tuning of $\Gamma_s/\Gamma_N$. N behaves as a tunnel probe irrespective of $V_{bg}$, even if $\Gamma_s/\Gamma_N$ is also affected by the back gating.

Odd-occupancy states display a much richer subgap structure. Figure 3(a) shows a series of plots corresponding to the same odd charge state but taken at different $V_{bg}$, hence different $\Gamma_s/U$ values. To gain a better understanding of their meaning, we start by discussing the top left panel in greater detail. The most remarkable features are pronounced subgap $dI/dV$ peaks that show a striking $V_{pg}$ dependence. These peaks can be ascribed to Andreev levels appearing at energies $eV = \pm |\zeta|$. Their gate modulation reveals a marked sensitivity of $\zeta$ with respect to $\epsilon_0$. Of particular interest are the two points where the

Andreev levels cross at zero bias. They represent degeneracies between the singlet and doublet states where the QPTs take place. Intuitively, this can be understood by recalling that the $V_{pg}$ range covered in the measurement is qualitatively equivalent to that represented by the horizontal line in the phase diagram in Fig. 1(c). Specifically, as $V_{pg}$ is swept to more positive voltages from the left, the ground state changes twice upon crossing the phase boundaries. Importantly, the observation of two crossings is consistent with a measurement taken at a relatively weak QD-S coupling.

The following panels in Fig. 3(a) reveal a clear trend for increasing $\Gamma_s/U$. This corresponds to an upward shift of the horizontal line in the phase diagram of Fig. 1(c). At first, the zero-bias crossing points move closer together, signaling that the doublet region shrinks. By further increasing $V_{bg}$ to 6 V, the two crossings merge approximately at the center of the Coulomb diamond. For even higher $\Gamma_s/U$, the Andreev levels no longer cross, suggesting that the singlet becomes the ground state throughout the entire $V_{bg}$ range. We notice that an unexpected feature emerges in the strong coupling, singlet regime. It consists of a zero-bias $dI/dV$ peak, which is clearly visible for $V_{bg} = 15$ V and persists at higher $V_{bg}$ where it gets overshadowed by the increasing magnitude of the Andreev resonances (the latter is due to the increasing values of $\Gamma_N$ and $\Gamma_s$, and the color scale has been adjusted accordingly).

A similar zero-bias feature was observed in Ref. [38] and a possible explanation in terms of a Kondo-type anomaly was suggested [39]. This interpretation may hold also in the present case. Since the available data do not allow us to go beyond this speculative level, we shall not discuss this observation any further.

Altogether, the behavior of the subgap levels shown in Fig. 3(a) demonstrates a QPT driven by the electrical tuning of $\Gamma_s/U$, which is corroborated by the $V_{bg}$ dependence of device parameters. To provide further support to our interpretation, we have calculated the NRG density of states (DOS) spectra of

FIG. 3. (a) Series of $dI/dV$ vs $(V, V_{pg})$ plots depicting the impact of back gating on the energy of Andreev levels. $V_{bg}$ increases from −4.5 to 39 V. The horizontal lines in the top left panel highlight the positions of the superconducting gap, $eV = \Delta$, and the Fermi level, $eV = 0$. $|S\rangle$ and $|D\rangle$ refer to the singlet and doublet ground states, respectively. The doublet ground state region is gradually suppressed for increasing $V_{bg}$, suggesting a QPT induced by the electrical tuning of $\Gamma_s$. Overlaid to the plots are the density of states spectra calculated by NRG (dashed lines). The DOS spectra, also shown in (b), were calculated using $U$, $\Gamma_s$, and $\Delta$ extracted from the experimental data.

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the QD-S system with parameters previously extracted from the normal data fitting (see Fig. 2 and the corresponding discussion). The numerical results, presented in Fig. 3(b) and overlaid to the plots in Fig. 3(a) (dashed lines), show remarkably good agreement with the experimental data.

As a subsequent step, we gathered the information contained in Figs. 2 and 3 in the form of an experimental phase diagram (Fig. 4). Two different methodologies were used to estimate the experimental phase boundaries (open circles). The most straightforward method relied on directly tracking the $V_{\text{PG}}$ position of Andreev level crossings in $dI/dV(V, V_{\text{PG}})$ plots taken at fixed $V_{\text{PG}}$, to extract the $e_0/U$ coordinates of the boundaries. These were later associated with the corresponding $\Gamma_S/U$ coordinates obtained from Fig. 2. However, owing to the finite width of the Andreev levels, this task became increasingly difficult as the crossing points moved closer together. To circumvent this issue, the phase boundaries around the particle-hole symmetry point were estimated from the half width of the normal-state Kondo resonances ($\Delta_L = 30$ mT, $g$ factors ~3.5 and ~5.75, as measured in similar devices [33,40]).

Finally, we study the scaling of the Andreev levels with respect to $T_K/\Delta$. For this analysis, we used $T_K$ values estimated from the half width of the normal-state Kondo resonances measured at the center of the Coulomb diamonds ($e_0 = 0$). The Andreev level energy at the same position $\xi(e_0 = 0)$ is plotted against $T_K/\Delta$ in Fig. 5, which includes data from a second device (device 2, presented in more detail in Ref. [34]). Interestingly, both data sets display nearly identical scaling which, for $T_K/\Delta \gtrsim 0.3$, also shows an excellent agreement with the NRG calculations. From the intersection of the data with $\xi = 0$, we estimate that the QPT occurs at $T_K/\Delta \approx 0.6$. This value agrees with those reported in the literature [41] after suitably rescaling $T_K$ to account for different definitions of this quantity. We attribute the discrepancy between the experiment and the theory for low $T_K/\Delta$ to an overestimation of $T_K$ in the weak coupling limit. Indeed, the agreement significantly improves by taking into account the broadening of the Kondo resonances imparted by the Zeeman effect due to the 30 mT magnetic field applied to suppress superconductivity. When the Zeeman splitting is included in the NRG calculations (dashed lines), it results in an upturn of the calculated curves in the weak coupling limit, where the Zeeman energy $E_Z$ is comparable to $T_K$.

Our herein reported findings of electrically tunable Andreev levels, in combination with a previous demonstration of their spin polarization [33], constitute important milestones towards pursuing proposals of engineering topological superconductors from arrays of proximity-coupled QDs.

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[34] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.95.180502 for details of the theoretical model and of the device parameter estimation procedure, as well as additional data of device 2, and a discussion of the width of the Andreev level resonances.
[37] Given the device geometry, the QD couples mostly likely to the portion of the semiconductor nanowire covered by the S contact. In this hypothesis, $\Delta$ corresponds to the induced superconducting gap.