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Andreev Molecule in Parallel InAs Nanowires

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ABSTRACT: Coupling individual atoms fundamentally changes the state of matter: electrons bound to atomic cores become delocalized turning an insulating state to a metallic one. A chain of atoms could lead to more exotic states if the tunneling takes place via the superconducting vacuum and can induce topologically protected excitations like Majorana or parafermions. Although coupling a single atom to a superconductor is well studied, the hybridization of two sites with individual tunability was not reported yet. The peculiar vacuum of the Bardeen—Cooper—Schrieffer (BCS) condensate opens the way to annihilate or generate two electrons from the bulk resulting in a so-called Andreev molecular state. By employing parallel nanowires with an Al shell, two artificial atoms were created at a minimal distance with an epitaxial superconducting link between. Hybridization via the BCS vacuum was observed and the spectrum of an Andreev molecule as a function of level positions was explored for the first time.

KEYWORDS: Andreev molecule, Yu-Shiba-Rusinov, superconductivity, nanowire, hybridization

INTRODUCTION

On the basis of Bardeen—Cooper—Schrieffer (BCS) mean-field theory, the superconducting vacuum only allows the addition of individual electrons with energy above the superconducting gap, however, it serves as a free source and drain of electron pairs, known as Cooper pairs (see Figure 1a). The interplay between an artificial atom, namely a quantum dot (QD), and the BCS vacuum was studied intensively, contributing to the formation of a subgap excitation, a so-called Yu—Shiba—Rusinov (YSR) state (or called Andreev Bound states in other limits, see Figure 1f). These excitations are shared between the QD and the superconductor (SC). Two of such bound states, which are formed with two spatially separated QDs, could be hybridized by the common SC lead, which we call an Andreev molecule. Coupling two QDs to a joint SC is also a basic building block of a Cooper pair splitter (CPS), where the QDs are attached to two normal leads allowing one to create spatially separated entangled electron pairs via crossed Andreev reflection. While a CPS favors weak SC-QD couplings, an Andreev molecule requires the opposite limit. Several theoretical works investigated how two bound states localized on separated dots effectively couple via the SC for example, as a minimal model for Majorana chain. Hybridization of YSR states was studied recently in a few scanning tunneling microscopy measurements placing different dimers on an SC surface. The target of the present work is to realize an Andreev molecular state with artificial atoms allowing individual tunability of the atomic sites and to explore its spectrum as a function of level position for the first time.

Realization of an Andreev molecule with QDs imposes a set of challenging constraints: the QDs must be strongly coupled to the SC and their distance should be minimized while preventing direct tunneling between them. To fulfill these requirements we construct our artificial atoms in a novel superconducting hybrid nanostructure, where double InAs nanowires are grown in close vicinity and are connected by an epitaxial Al shell (see Figure 1b). Whereas two QDs can be formed in separate wires thereby excluding the direct tunneling between them, the epitaxial Al shell yields a defect-free SC—semiconductor interface ensuring the strong proximity and SC-QD coupling as used in various hybrid quantum devices, like Andreev—quubits, or Majorana devices. Recent works have already reported the Cooper pair splitting signals and nonlocal pair tunnelings in individual nanowires placed parallel close to each other manually with a micromanipulator. Moreover, Andreev bound states were also coupled by direct tunneling between QDs in series. However, none of them has realized the strong
Figure 1. (a) General concept of a molecular state formed by tunneling via a barrier between two atomic sites (top). In our case, the interaction between the QDs is mediated by an SC (bottom), where two electrons can be created from the SC vacuum. (b) High-resolution scanning electron micrograph (SEM) of the as-grown parallel wires. The epitaxial Al connects the two InAs nanowires. (c) False-color SEM and (d) schematic illustration of the device. The epitaxial Al shell (green) is etched in the middle where the QDs are formed in the two InAs wires (brown). The QDs are formed by the epitaxial full-shell Al, illustrated in Figure 1c, and the epitaxial Al connects the two InAs nanowires. (e) Sketch of the setup used for modeling the system with tunnel coupling barriers. While the left electrode with a gap of 250 nm long segment. Two parallel double QD was formed in the two wires. Low-temperature electronic transport measurements were carried out at a base temperature of 40 mK (for details, see Methods). In two-terminal subgap spectroscopy, the differential conductance \( G = I_{SC}/V_{AC} \) was measured with the tuning of the QDs by individual plunger gates, as depicted in Figure 1d (\( V_T \) corresponds to the top, \( V_B \) to the bottom gate voltage). The source terminal biased with \( V_{SD} \) was found to be coupled strongly to the QDs, whereas the other one worked as a SC tunnel probe leading to a SC–QD–I–SC junction, where I stands for insulator. Two different devices are presented in this paper; one did not show strong coupling between nanowires and thus serves as a reference junction (device A), whereas for the other a strong hybridization of artificial atoms via a SC needed for the formation of the Andreev molecular state, the elementary building block of a Majorana chain. In this paper, we report the signature of an Andreev molecule, for the first time in parallel InAs nanowires. We discuss first the case of uncoupled YSR states and then compare it to the strongly interacting system involving the hybridization via SC and Coulomb repulsion, both experimentally and theoretically.

**RESULTS AND DISCUSSION**

**Device Outline.** The specific system studied here is illustrated in Figure 1c–e. A parallel double QD was formed in a pair of InAs nanowires merged by epitaxial full-shell Al, which was etched away on a ~250 nm long segment. Two common superconducting (Ti/Al) electrodes were attached to epitaxial Al on the nanowires forming parallel SC–QD–SC junctions in the two wires. Low-temperature electronic transport measurements were carried out at a base temperature of 40 mK (for details, see Methods). In two-terminal subgap spectroscopy, the differential conductance \( G = I_{AC}/V_{AC} \) was measured with the tuning of the QDs by individual plunger gates, as depicted in Figure 1d (\( V_T \) corresponds to the top, \( V_B \) to the bottom gate voltage). The source terminal biased with \( V_{SD} \) was found to be coupled strongly to the QDs, whereas the other one worked as a SC tunnel probe leading to a SC–QD–I–SC junction, where I stands for insulator. Two different devices are presented in this paper; one did not show strong coupling between nanowires and thus serves as a reference junction (device A), whereas for the other a strong hybridization of the QDs and signatures of the Andreev molecular states was observed (device B).

When a QD is coupled strongly to an SC electrode, subgap states are formed by the hybridization between a QD level and the SC electrode. Let us consider the QD being singly occupied (called doublet ground state due to degeneracy in the spin of freedom, noted by “D”). In this case, the lowest excitation available is either adding an electron costing the charging energy, \( U_1 \), or adding a quasi-particle to the SC band, which costs the energy of the superconducting gap, \( \Delta \). However, the interaction between the QD and the SC induces a lower-lying excitation: a singlet (marked by “S”) formed by an electron on the QD and a quasi-particle in the SC is hybridized with the empty QD and quasi-particle state via a Cooper pair transfer to the superconducting condensate. This results in a so-called YSR subgap state in the \( U > \Delta \) limit (orange state in Figure 1f). In a QD, the level position can be tuned, and the YSR energy develops in an “eye-shaped” curve as a function of it, that is, the plunger gate voltage similarly to the one sketched in inset I. of Figure 2a. For large (or small) enough gate voltage, the ground state of the QD changes to the singlet state bearing double (or zero) occupation (for more details see Supporting Information or, for example, ref 14).

In the following, we review the spectrum of the parallel double QD structure in three steps: (i) two independent, uncoupled YSR states; (ii) adding interdot Coulomb repulsion, and (iii) including the superconducting coupling. We label the QDs and their features as top (T) and bottom (B) ones, marked with red and green as in Figure 1, respectively,
Figure 2. Uncoupled parallel YSR states (device A). (a) Schematic illustration about the stability map of parallel QDs with joint electrodes. Resonances of the top and bottom QDs are depicted with red and green, respectively. The lever arms refer to a finite cross capacitance of each gate to the opposite QDs. Inset 1 depicts the YSR spectrum residing in the top QD ("S" and "D" refer to singlet and doublet ground states, respectively). Δ₁ and Δ₂ are the superconducting gaps of the strongly coupled electrode and the SC probe. The pink circle and the blue diamond indicate the energy of YSR₁ along the cuts taken in the stability map. (b) Expected excitation spectrum of YSR states along the pink line shown in panel a. Whereas YSR₂ (green) evolves along the cut since it is sensitive to its own gate (V_b), the YSR₁ state (red) stays on constant energy as the slice is parallel to the red resonances. (c) Bound state spectrum along the blue line in panel a. The excitation of the YSR₂ state moved to lower energy compared to the one in panel b (the original energy is depicted with pink dashed lines) as the charge degeneracy of the top QD was approached (see the blue diamond in inset I of panel a). (d) Measured conductance as a function of gate voltages for device A in the normal state. (e) Finite-energy originating from the SC tunnel probe, which can be identified as YSR₁ since the on-site energy of the top QD is kept constant due to the parallel slicing in both panels. The excitations do not touch at zero V_SD but stay always at finite energy originating from the SC tunnel probe, which introduces a ± Δ₂ gap in the excitation spectrum. These minima correspond to the ground state transitions of the QD addressed also in the figure. Depending on the position of the slice, the energy of the constant line can vary between Δ₁ and Δ₁ + Δ₂. Obviously, the YSR₂ excitation can occupy the lowest energy Δ₁ when the corresponding (top) QD is close to resonance (Figure 2c, blue line in Figure 2a), while moving deeper in the blockade brings its energy toward the gap edge Δ₁ + Δ₂ regardless of the parity of the ground state. The movement of the signal while approaching a resonance is indicated with red arrows in Figure 2c. For clarity, inset I in Figure 2a depicts YSR₁ as the function of its own plunger gate (V_T), in which the markers assign the actual excitation energies considered in Figure 2b,c.

The measurements of device A follow well our basic predictions outlined above. The stability map in the normal state, which was recorded by applying 250 mT out-of-plane magnetic field, is shown in Figure 2d. Two different bias cuts parallel to the top QD resonance in Figure 2e,f reveal the movement of YSR₁ (red arrow) while the development of YSR₂ remains intact. By a careful inspection of the data, one can identify an additional excitation line at higher energy, which can be attributed to another orbital of the bottom QD or to a higher-lying transition. Recording the spectrum along the other gate direction V_T a similar behavior of the bound states was observed (for details see Supporting Information), where the movements of (multiple) YSR₂ states are also trackable. On the basis of the two dominant lever arms in the stability sweep and the fact that different YSR states were

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supposing only a single YSR state residing in each QD (YSR₁ and YSR₂).

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captured by tuning either $V_T$ or $V_B$ confirmed the model of having YSR states in both QDs.

We note that the Kondo effect\cite{8,66,67} was suppressed in most of the gate settings in both device A and B since the Kondo temperature did not exceed the superconducting gap ($k_B T_K < \Delta$). As the QDs enter a more open regime, Kondo correlations appear competing with the superconductivity, which reduces the visibility of the outlined YSR behaviors (for details see Supporting Information).

**Interdot Coulomb Repulsion.** The question arises of how the spectrum is modified compared to the uncoupled case if there is significant interdot capacitance (see C in Figure 1e). The normal state stability map turns into the so-called honeycomb pattern (see Figure 3a) well-known for double QDs.\cite{65} Hence, slicing parallel to any resonances along a straight line in the gate map gives no longer a constant-energy YSR state but a charge state-dependent one. For example, along the pink dashed line, the top YSR state develops according to the symbols in inset I, where YSRT is depicted as a function of its own plunger gate, $V_T$. For small $V_B$ values, the line cut is off-resonance and YSRT is in the doublet ground state (diamond symbol). By increasing $V_B$, the bottom QD is brought to resonance which leads to an effective gating of the top QD. This shifts the top QD closer to its resonance and lowers the energy of YSRT (circle symbol). Going through another resonance of the bottom QD (by further increasing
As the YSRB and YSRT excitations approach each other, the charge degeneracy points (pink symbols) demonstrate the hybridization of the states, which is expected in a simple capacitive picture. The difference between the predicted spectrum and the measured data is more obvious for cuts along the other gate direction as the comparison of Figure 3c,f shows. Assuming only capacitive coupling between QDs (panel c), YSR is expected to be red shifted, whereas for YSRT observed particularly in Figure 3b. The YSR state is mostly bound to the gap edge and develops rapidly at the ground state transitions matching the green curve in Figure 3b. As a result of using a superconducting tunnel probe, negative differential conductance around the YSR signals also appear in both of the experiments and the simulations.

In general, there are several characteristic features in Figure 3f, which strongly deviate from the capacitive picture, nevertheless, they are qualitatively recovered in the simulation in Figure 3i. First of all, (i) the eye-shaped YSR resonance is completely distorted in the measurement as well as in the numerical data. Moreover, (ii) the expected horizontal YSR signal does not stay flat in the doublet region of YSR (indicated by the blue triangle) as it reaches a peak at $V_T = -0.92$ V and $V_T = -0.87$ V but rather follows the curvature of YSR similarly to the simulation. Though well-pronounced anticrossings are absent, (iv) extra dispersive lines (one example is marked by the green triangle) arise between the YSR and YSR signals, in the numerical results (see white arrows in Figure 3h). As a result of the theory not matching the measurements in a few aspects. On one hand, the discrepancies originate from several simplifications the model takes. The calculations neglect the presence of multiple QD orbitals and exclude the relaxation from excited states, hence allowing arbitrary high-energy virtual states, which are usually not visible in a bias-spectroscopy measurement. On the other hand, further limitation comes from the broad line width of the measured YSR states smearing the neighboring excitation lines. Overall, despite the theory being simplified, many prominent features of the measured data were captured in the simulation qualitatively by assuming hybridization via the SC, which supports our interpretation of an observed Andreev molecule.

The two investigated devices (A and B) showed very different behaviors. Whereas for A no hybridization was observed, sample B exhibited signatures of the Andreev molecule. Careful SEM analysis revealed an important structural difference: for sample B the two InAs nanowires were merged by the epitaxial Al, whereas for device A the wires have separated and became only connected by the ex situ evaporated contacts (blue in Figure 1a, see Supporting Information). These Ti/Al contacts were established in Figure 3g by assuming hybridization between the QDs via the SC. As shown below, our fully interacting two-dot simulation reproduces all of the main features of the unique experimental spectra.

To proceed, we modeled the QDs with single sites tunnel-coupled to the SC. The SC was treated in the zero bandwidth approximation by considering a single quasi-particle level, and the tunnel probe was handled perturbatively. In this subsystem, we calculated the eigenstates and the transport with exact diagonalization of the Hamiltonian (for detailed description of the model see Methods and Supporting Information). All relevant parameters ($U_T$, $U_B$, $C$, $t_1$, $t_B$, $\Delta_1$, $\Delta_2$, shown in Figure 1e) were directly extracted from the experimental data, thus the model has no fitting parameters. The simulated normal state stability map (Figure 3g) and the spectra taken along the pink and blue lines marked in Figure 3a,g are shown in Figure 3h,i.

After a quick comparison of Figure 3b, c with Figure 3h, i, one can see that the hybridization via the SC strongly restructures the spectra. Let us now carefully compare the measurements in Figure 3e,f with the calculated spectra in Figure 3h, i, and show that they qualitatively match well. (i) The anticrossings and the bended of YSRT observed in Figure 3e,i are restored in the numerical results (see white arrows in Figure 3h). As a result of using a superconducting tunnel probe, negative differential conductance around the YSR signals also appear in both of the experiments and the simulations.

In summary, we have found strong interactions between parallel YSR states realized in double InAs nanowires connected by an epitaxial Al shell. The small geometrical distance between the QDs resulted in capacitive coupling, while the shared epitaxial Al source contact enabled hybrid-
ization via the SC vacuum. The latter one allowed the emergence of an Andreev molecular state, whose spectrum was explored as a function of the QD level positions for the first time. The detected spectroscopic features were reproduced by our numerical calculations. Our result is an important milestone toward artificial topological superconducting systems, where Kitaev-like chains are assembled from sites hybridized via SCs. With the strong superconducting coupling demonstrated here, double InAs nanowires can be also promising candidates to host non-Abelian excitations, like parafermions as a key ingredient of topological quantum computation.

Methods

Device Fabrication. In As nanowires were grown by MBE in the wurtzite phase along the (0001) direction catalyzed by Au. The pattern of the predefined Au droplets allowed one to control the geometrical properties of the proposed double nanowires, including the diameter, distance, and the corresponding alignment of the cross sections. The 20 nm thick full-shell Al was evaporated at low temperature in situ, by rotating the substrate, providing epitaxial, oxide-free layers. The evaporation on such a pair of adjacent nanowires resulted in the merging by the Al. Nanowires with ~80 nm diameter and ~4 μm length were deposited on a p-doped Si wafer capped with 290 nm thick SiO2 layer by using an optical transfer microscope with micromanipulators. The Al shell on a ~250 nm long segment was removed by means of wet chemical etching. A MMA/MAA EL-6 double-layer performed as a masking layer, which was locally exposed by EBL, allowing the MP-321 selective developer to access the Al (60 s). The etching was followed by a careful localization of the wires with high-resolution SEM. Both source-drain and side gate electrodes were installed in a common EBL step. The sample was exposed to RF Ar milling in the evaporator chamber to remove the native Al2O3. The process was followed by the metalization of Ti/Al (5/95 nm) without breaking the vacuum.

Experiments. Low-temperature characterization was carried out in a Leiden Cryogenics dry dilution refrigerator with a base temperature of 40 mK. Transport measurements were performed with a standard lock-in technique by applying 10 μV AC signal at 113 Hz on one of the SC electrodes, whereas the differential conductance was recorded via a home-built current amplifier on the other one. DC bias was adjusted by the offset of the amplifier. We note that due to the geometry, the features of both QDs were measured simultaneously in a single measurement, and hence, the sum of two excitation spectra was captured. Out-of-plane magnetic field was realized by an AMI superconducting magnet.

Modeling. In the theory, the QDs are modeled by capacitively interacting single sites (see Figure 1e) coupled to the SC with tunnel amplitudes \( t_f \) and \( t_b \). This restricts the electron number on the QDs between 0 and 2. The left SC in Figure 1e is handled in the zero bandwidth approximation \( 28,69 \) effectively assessing quasi-particle sites with energy \( \Delta_1 \). The right superconducting probe with \( \Delta_2 \) superconducting gap is treated perturbatively with Dynes-like density of states. By allowing only a limited number of quasi-particles to be present in the system, the energy spectra with the eigenstates can be derived with exact diagonalization of the Fock-space Hamiltonians. Transition rates between the states, involving the processes of adding and removing an electron to the Andreev molecule, are expressed by Fermi’s golden rule. The transport (net current) is obtained by solving the classical master equation in the stationary limit, which governs the time evolution of the QD occupations (for more details see Supporting Information). Simulations with only capacitively interacting YSR states were also carried out. In those calculations, the QDs were coupled to two, separate single sites with energies \( \Delta_1 \) effectively turning off the superconducting hybridization between them. The simulated spectra are in good agreement with the sketches of Figure 3b,c (for further details see Supporting Information). In the model, we have used charging energies \( U_f = 1.2 \, \text{meV} \) and \( U_b = 2.2 \, \text{meV} \), off-site repulsion energy (proportional to the interdot capacitance) \( C = 0.1 \, \text{meV} \), and superconducting gaps of \( \Delta_1 = 200 \, \mu\text{eV} \) and \( \Delta_2 = 120 \, \mu\text{eV} \), which were extracted from the measurements. Tunnel amplitudes were estimated as \( t_f = 0.15 \, \text{meV} \) and \( t_b = 0.05 \, \text{meV} \) based on the shape of the YSR states.

Associated Content

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.1c01956.

Detailed explanation of YSR states, further data analysis of both device A and B, and the description of the theoretical model (PDF)

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Author Contributions
O.K. and I.L. fabricated the devices. O.K., Z.S., and G.F. performed the measurements and did the data analysis. Z.S. built the theoretical model and developed the numerical simulations. T.K. and J.N. grew the nanowires. All authors discussed the results and worked on the manuscript. P.M. and S.C. proposed the device concept and guided the project.

Notes
The authors declare no competing financial interest.

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