Charge localization and reentrant superconductivity in a quasi-ballistic InAs nanowire coupled to superconductors

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A semiconductor nanowire with strong spin-orbit coupling in proximity to a superconductor is predicted to display Majorana edge states emerging under a properly oriented magnetic field. The experimental investigation of these exotic states requires assessing the one-dimensional (1D) character of the nanowire and understanding the superconducting proximity effect in the presence of a magnetic field. Here, we explore the quasi-ballistic 1D transport regime of an InAs nanowire with Ta contacts. Fine-tuned by means of local gates, the observed plateaus of approximately quantized conductance hide the presence of a localized electron, giving rise to a lurking Coulomb blockade effect and Kondo physics. When Ta becomes superconducting, this local charge causes an unusual, reentrant magnetic field dependence of the supercurrent, which we ascribe to a 0 - π transition. Our results underline the relevant role of unintentional charge localization in the few-channel regime where helical subbands and Majorana quasi-particles are expected to arise.

INTRODUCTION

Nanowires (NWs) with strong spin-orbit coupling and induced superconductivity have the potential to realize topological superconductivity (1, 2). Majorana modes emerge if the one-dimensional (1D) character of the NW is preserved over micrometer-scale lengths and the chemical potential is positioned within the helical gap opened by a properly oriented magnetic field \( B \) (3). The Rashba spin-orbit energy in InAs NWs is, at most, of the order of 100 \( \mu \text{eV} \), as deduced from measurements of weak antilocalization (4). Given the modest size of the spin-orbit energy, the second condition implies that the 1D conduction mode supporting Majoranas should be only slightly filled. For this reason, it is important to explore the properties of semiconductor NWs at low subband filling in the presence of the superconducting proximity effect and a magnetic field. To this aim, we investigate InAs NWs coupled to tantalum-based superconducting contacts with a high in-plane critical field, \( B_c \sim 1.8 \ T \).

Conductance quantization is the experimental paradigm of ballistic 1D transport (5). In semiconductor NWs, this phenomenon is better observed at large magnetic field (6), where backscattering is reduced and spin degeneracy is lifted, leading to conductance steps of \( e^2/\hbar \), where \( e \) is the electron charge and \( \hbar \) is the Planck constant. Recently, conductance quantization was also observed at zero magnetic field, with steps of \( 2e^2/\hbar \) due to twofold spin degeneracy (7–10). In this work, we make use of two independently tunable bottom gates to tailor the potential landscape in the NW channel (10). Proper tuning of the applied gate voltages results in the creation of a local point contact exhibiting approximately quantized conductance plateaus in the few-subband regime. We find that unintentional charge localization, while seemingly suppressed at high magnetic field, becomes apparent at low \( B \) in both normal and superconducting regimes. In the normal state, the spin of the localized charge is screened by Kondo correlations, which shape the linear and nonlinear conductance at the onset of the first plateau.

Owing to the large electron \( g \) factor in InAs and the relatively large \( B_c \), we are also able to investigate the superconducting proximity effect coexisting with a strong spin polarization. We observe a nonmonotonic behavior of the critical current as a function of \( B \) that can be understood as a Zeeman-driven quantum phase transition from a spin singlet ground state, with 0-phase-shift Josephson coupling, to a spin-1/2 ground state, with \( \pi \)-phase-shift Josephson coupling. Upon increasing \( B \), the supercurrent first vanishes at the 0 - \( \pi \) transition and then recovers once the Zeeman energy is large enough to stabilize the spin-1/2 ground state. This interpretation is confirmed by theoretical calculations based on an Anderson-type model coupled to superconducting leads with strong and gate-dependent tunnel couplings. A reentrant supercurrent due to the same mechanism is also observed in a second device exhibiting a clear quantum dot behavior and Kondo effect.

RESULTS

We performed standard two-terminal low-temperature conductance measurements on an InAs NW with Ta superconducting contacts (device 1), under an external magnetic field aligned parallel to the long axis of the wire, shown in Fig. 1A (data for different angles can be found in the Supplementary Materials). The profile of the conductance band was locally tuned by two gates underneath the wire (see inset of Fig. 1D).

To look for conductance quantization, the device was first characterized in the normal state at \( B = 2.9 \ T \), which is well above \( B_c \); the linear conductance, \( G \), was measured as a function of voltages \( V_{\text{G2}} \) and \( V_{\text{G3}} \) on gates 2 and 3, respectively (Fig. 1B). All data are corrected for the series resistance of the measurement circuitry (51.37 kilohms), unless explicitly stated. Two conductance plateaus, around \( 0.9e^2/\hbar \) and \( 1.8e^2/\hbar \), can be identified, which are close to the ideal values for one and two 1D
similar capacitive coupling to gates G2 and G3, hence producing the predominantly diagonal conductance ridges observed in Fig. 1B. The amplitude of these additional features varies over the (V_{G2}, V_{G3}) plane and can vanish at certain regions.

Figure 1C shows three G(V_{G3}) traces taken at different V_{G2}. The green trace exhibits a clearly visible broad peak structure, causing an overshoot of the conductance at the onset of the first plateau. This structure is no longer present in the blue trace, resulting in an essentially flat conductance plateau. Further increasing V_{G2} results in a global suppression of the conductance step (red trace).

From now on, we focus on the intermediate value of V_{G2}, where the first conductance step shows no spurious resonances, thereby resembling the one expected for the onset of the first 1D conduction mode in a ballistic point contact. From a comparison with the other traces, we know that a resonance is lurking in this seemingly ideal plateau. This underlines the importance of double gate control in revealing the nature of the observed transport features. Furthermore, charge localization is apparent in the second plateau where conductance oscillations remain visible (Fig. 1C). The second plateau extends on a much larger V_{G3} range, suggesting that conductance is limited by the barrier induced by V_{G3}.

The first conductance plateau preserves its flat, featureless character over a large B range. Upon reducing B to 1.4 T (Fig. 1D), the plateau shrinks with B due to the decreasing Zeeman energy, E_Z = g gamma mu_B, where mu_B is the Bohr magneton and g is the electron g factor in the point contact, while the conductance remains quantized at 0.9 e^2/h. The full-range B dependence is shown in Fig. 2A. At low B, the superconducting proximity effect causes the divergence of the conductance, simultaneously washing out the 0.9 e^2/h plateau. This behavior can be seen from G(V_{G3}) traces below 1.4 T (Supplementary Materials) but hardly in Fig. 2A. Instead, Fig. 2A shows that, at large B, the 0.9 e^2/h plateau widens linearly with B, as highlighted by two dashed lines. The two lines do not coalesce at B = 0 as expected, if the width of the plateau was simply proportional to E_Z. This zero-field splitting is again due to a localized charge state, most likely the same already identified at B = 2.9 T. The residual splitting is indicative of a sizable charging energy, U, associated with the localized state. We find U \sim 1.3 meV and |g| = 11 (Supplementary Materials). We convert V_{G3} scale into energy with the help of dI/dV measurements at finite source-drain bias voltage, V. This standard procedure (Supplementary Materials) yields a conversion factor of \alpha = 0.0082 meV/mV.

Localized states are often observed in semiconductor NWs. They can form due to a plethora of confining mechanisms: crystal defects or impurities in the NW, tunnel barriers at the contacts, and surface charges. In a gate-defined point contact, where charge density is substantially lowered and electric field screening is consequently reduced, localization is enhanced and Coulomb interaction emerges. Strongly localized states leading to a few rather sharp Coulomb resonances can be observed in the studied gate-induced constriction near full charge depletion. They lie outside the (V_{G2}, V_{G3}) field explored in Fig. 1B (Supplementary Materials). The localized state at the onset of the first conductance plateau has a more subtle nature, and as we have seen, its presence may go unperceived without a proper control of the electrostatic landscape.

Before discussing the superconducting regime, it is instructive to examine the normal type behavior at T \sim T_c. Figure 2B shows G(V_{G3}) at 4.2 K. The onset of conductance through the first spin-degenerate subband is preceded by a shoulder at \sim e^2/h. A shoulder can also be consistently found in a measurement of dI/dV at 15 mK and |eV| \gg \Delta.
Here, $d_\sigma$ and $c_{\sigma\alpha}$ are impurity and lead electron operators, respectively, where $\sigma \in \{\uparrow, \downarrow\}$ and $k$ is the crystal momentum, $n = \sum_\sigma d_\sigma^\dagger d_\sigma$ is the localized level occupancy operator, $\delta$ is its energy position (later, we shall scale $\delta$ to $V_{G3}$ for a direct comparison with the experimental data), and $S_\sigma = (d_\sigma^\dagger d_\sigma - d_\sigma d_\sigma^\dagger)/2$ is the spin operator. The hybridization matrix elements for transitions between zero and one electrons, $V^{(1)}$, and one and two electrons, $V^{(2)}$, can be different ($14$). We parametrize this asymmetry through $x \equiv (V^{(1)} - V^{(2)})/V^{(1)}$, which is adjusted in our calculations to find the best agreement with the data. The coupling between the level and the leads results in a broadening $\Gamma = \pi |V^{(1)}|^2 \rho$, where $\rho = \sum_{\sigma} \delta(w_0 - e_V)$ is the density of states in the leads. Because the localized-state wavefunction depends on the (spin-dependent) trapping potential, we allow corrections to the hopping $V^{(1)}$, which are linear in $\delta$ (and hence $V_{G3}$) and $B$ so that $\Gamma = \Gamma_0 + \Gamma_1(\delta U + \gamma E_2 U)$. Physically, the $B$ dependence can be expected from the influence of the magnetic field on the orbital motion and confinement of electrons. We introduced this broadening term to explicitly demonstrate, by comparing with the data, that the conductance through a correlated quantum dot can mimic plateaus of nearly quantized conductance. Our theory supports our physical interpretation. The parametrization for transport in the normal state qualitatively reproduces the experimental phenomenology in the superconducting state, as shown below. The last term in Eq. 1 accounts for superconducting pairing. The model was solved using the numerical renormalization group (NRG) method ($16$, $17$). Above $\Gamma/U = 0.4$, charge quantization due to Coulomb blockade is lost, as determined from the charge susceptibility (Supplementary Materials). In the experiment, the first conductance plateau and the corresponding supercurrent data described below always occur for $\Gamma/U < 0.4$.

In the normal regime ($\Delta = 0$), the parameters are severely constrained even if only qualitative features of the conductance are to be reproduced for different $T$ and $B$. At $B = 0$, Kondo correlations at finite $T$ enhance the conductance to a value below the unitary limit producing a conductance shoulder at $\delta \sim 0$, as experimentally observed at $T = 4.2$ K (Fig. 2B). At finite $B$, the shoulder evolves into a plateau at $0.9e^2/h$. The results of the NRG calculations reproduce remarkably well the experimental trend, as shown in Fig. 1D. In particular, the calculated conductance at the spin-resolved plateau remains constant despite the large variation of $E_d/U$. The $B$-dependent term (proportional to $c_2$), even if small against the gate-dependent term (proportional to $c_1$), is essential. Without it, the theory would evolve into a local minimum, as we actually find experimentally when $B$ is applied perpendicularly to the NW under the same gate configuration (Supplementary Materials).

We now address the superconducting proximity effect. Figure 3 (A to D) shows supercurrent measurements as a function of $V_{G3}$ at different values of $B$. Except for Fig. 3A, showing switching and retrapping currents directly measured at $B = 0$, the other panels display critical current, $j_c(V_{G3})$, traces obtained from fitting the measured $dI/dV(V)$ curves to the so-called resistively and capacitively shunted junction model (details on the measurement and fitting methodology are given in the Supplementary Materials). Fitting is necessary because increasing $B$ makes the junction overdamped with a Josephson energy comparable to the thermal energy and no zero-resistance branch. While the normal conductance increases monotonically with $V_{G3}$ [see the superimposed $G(V_{G3})$ trace in Fig. 3A], $j_c$ does not, in contrast to the Ambegaokar-Baratoff relation, for which $j_c \sim G_0$. At $B = 0$, the switching currents (closely related to $V_{G3}$) are slightly peaked in correspondence with the Kondo regime. Upon increasing $B$, $j_c(V_{G3})$ develops a minimum around $V_{G3} = -1.25$ $V$ (Fig. 3B) and gets fully suppressed for $B = 0.75$ $T$ (Fig. 3C) before remerging at higher $B$ (Fig. 3D).
DISCUSSION

The behavior shown here can be explained using our model. The phase diagrams in Fig. 3 (E to H) show an open region where \( j_c > 0 \) (corresponding to a spin-singlet ground state) and a closed region where \( j_c < 0 \) (corresponding to a spin-1/2 ground state). The sign reversal reflects a phase shift in the current-phase Josephson relation.

Because of correlated hopping \((x ≠ 0)\), the phase boundary has the shape of a skewed arc. At \( B = 0 \) (Fig. 3E), for odd charge \((-0.5 ≤ δ/U ≤ 0.5)\), strong (weak) coupling tends to stabilize a singlet (doublet) ground state \((18–25)\). The singlet has a predominantly Bardeen-Cooper-Schrieffer character for \( Δ ≫ Γ_S ≫ U \) and a predominantly Kondo character for \( Γ_S > Δ \). The Zeeman effect is antagonistic to both of these many-body phenomena, thereby reducing the singlet binding energy and making the spin-1/2 domain grow with \( B \) (Fig. 3F to H) \((26)\), in agreement with the experiment (see additional \( j_c \) data in the Supplementary Materials).

These phase diagrams can account for the unusual, nonmonotonic \( B \) dependence of \( j_c \) observed experimentally. The white lines in Fig. 3

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Fig. 3. Experimental supercurrent and corresponding quantum dot modeling. (A to D) Measured \( V_{G3} \) dependence of the switching currents \( I_s \) and \( I_r \) at \( B = 0 \) (A) and of the fitted critical current \( j_c \) at different \( B \), hence \( E_z/U \) (data from device 1). The normal-state \( G \) measured at 4.2 K and \( B = 0 \) is overlaid in (A). (E to H) NRG calculations of \( j_c(δ/U, Γ/U) \) for values of \( E_z/U \) corresponding to the left panels. Each phase diagram has a closed region corresponding to a spin-1/2 ground state, surrounded by an open region where the ground state is a singlet. Crossing the boundary between these regions at constant superconducting phase difference results in a reversal of the supercurrent. To underline this effect, in the spin-1/2 region, we conventionally give a negative sign to \( j_c \). The white lines represent the \( Γ'(δ, E_z) \) dependence obtained from the normal-state fit parameters. a.u., arbitrary units.
(E to H) denote the \( \Gamma(\delta, E_2) \) trajectory followed in the experimental sweeps, as deduced from normal-state fit parameters. As the doublet region of the phase diagram grows with \( E_2 \), its phase boundary approaches the \( \Gamma(\delta, E_2) \) trajectory, leading to a suppression of \( j_c \) in the region of closest proximity. At \( E_2/U = 0.37 \), the phase boundary reaches the \( \Gamma(\delta, E_2) \) trajectory, and \( j_c \) is correspondingly suppressed due to a competition between 0- and \( \pi \)-junction behavior. For larger \( E_2 \), the \( \Gamma(\delta, E_2) \) trajectory crosses the spin-1/2 region, where the system acquires a clear \( \pi \)-junction behavior characterized by negative \( j_c \).

Qualitatively, the observed gate dependence of \( I_c \) may also result from the opening of a helical gap due to Zeeman and spin-orbit interaction, as a gate voltage could potentially tune the chemical potential in and out of the helical gap in a hypothetical picture of noninteracting subbands (27). However, this interpretation is incompatible with our other observations in the same device.

In device 1, the charge localization responsible for the observed reentrant supercurrent was hidden by the seemingly ballistic transport. To further support the proposed physical picture, we performed a similar experimental study on a second device (device 2) displaying a quantum dot transport regime with clear Coulomb blockade oscillations. Device 2, shown in Fig. 4A, consists of an InAs NW contacted by superconducting Ta electrodes. Figure 4B shows a map of \( G \) as a function of back-gate voltage, \( V_{bg} \), and side-gate voltage \( V_{sg} \). We observe a Coulomb blockade pattern characteristic of single-dot transport with alternating large/small peak spacing, corresponding to even/odd occupation, respectively. We focus on the odd charge state indicated by a black arrow. The Coulomb peaks of this shell are renormalized by \( V_{sg} \), a sign of \( \Gamma \) tuning.

Before describing the supercurrent behavior, we briefly characterize the normal-state regime by raising \( T \) above \( T_c \approx 0.7 \) K. Figure 4C shows a plot of the charge stability diagram taken at \( V_{sg} = 1 \) V, which exhibits the typical Coulomb diamonds of a high-impedance quantum dot. At odd filling (spin-1/2 ground state), we observe a zero-bias conductance ridge due to the Kondo effect. At finite magnetic field, the Kondo resonance splits due to Zeeman effect, as shown in Fig. 4D.

After corroborating the presence of the Kondo effect, we explored the \( B \) evolution of \( I_c \) at different \( V_{sg} \). In Fig. 4E, we show fitted \( I_c(V_{bg}) \) curves obtained from one of these series of measurements at \( V_{sg} = -1 \) V. The curves display maxima in correspondence to the normal-state Coulomb peaks. As \( B \) is increased, these split apart due to Zeeman effect. Also, and more remarkably, a nonmonotonic behavior is observed in the Kondo valley, where \( I_c \) goes from 0.2 nA at \( B = 0.2 \) T to nearly zero at \( B^* = 0.3 \) T and then reenters at \( B = 0.5 \) T with a value of \( \approx 0.1 \) nA. In our model, this is interpreted as a Zeeman-induced 0 \( \to \pi \) transition from the destabilization of the Kondo singlet due to \( B \)-induced spin polarization. In Fig. 4F, we show the dependence of the crossover field at half-filling, \( B^* \), on \( V_{sg} \), as extracted from the \( B \) evolution of \( I_c \) at other \( V_{sg} \) values. As also predicted by our theory, lowering \( \Gamma \) (by increasing \( V_{sg} \)) reduces \( B^* \) all the way down to zero (\( B^* = 0 \) means that the junction is already at the \( \pi \) phase at \( B = 0 \)).

We conclude that, even when seemingly absent, charge localization may play a crucial role in the transport properties of semiconductor NWs. In the superconducting regime, charge localization gives rise to a strong nonmonotonic behavior of the Josephson current as a function of \( B \) due to a Zeeman-induced 0 \( \to \pi \) phase transition. Our findings are relevant to experiments aiming at detecting Majorana modes in Josephson junction geometries based on depleted NWs under strong Zeeman fields (28–30). The anomalous \( B \)-field dependence of the critical current, owing to the presence of Majoranas in the

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**Fig. 4. Reproduction of reentrant supercurrent data in a closed quantum dot.** (A) Schematics of device 2. (B) Color plot of \( G(V_{bg}, V_{sg}) \) for device 2. The data include the contribution of the series resistance \( (R_s = 51.37 \text{ kilohms}) \) in the measurement circuit. White color corresponds to a nondissipative superconducting regime where \( G = 1/R_s \). (C) Stability diagram, \( dI/dV(V_{sg}, V) \), measured at the position of the red line in (B) at \( T = 0.75 \) K, i.e., just above \( T_c \). A Kondo ridge at odd filling is indicated by an arrow. (D) Zeeman splitting of the Kondo resonance in the normal state taken in the middle of this Kondo ridge at 0.1-T increments. The traces are vertically shifted by \( -0.025e^2/h \) for clarity. (E) \( B \) evolution of fitted \( I_c(V_{bg}) \) at the blue line in (B) and for different \( B \) (no offset). In the Kondo valley, \( I_c \) shows a reentrant behavior, vanishing at a crossover field \( B' \approx 0.3 \) T and reemerging for \( B > B' \), heralding a ground-state transition from a Kondo singlet to a spin-split doublet. (F) Crossover field, \( B' \), versus \( V_{sg} \) as extracted from the \( B \) evolution of \( I_c \) at the corresponding linecuts in (B). The error bars are determined by the 0.1-T increment in \( B \), as in (E).
sectional (31–33), may be masked by the localization effects and the Kondo physics discussed here.

MATERIALS AND METHODS
Device 1, shown in Fig. 1A, was fabricated from a single 65-nm-diameter InAs NW grown by chemical beam epitaxy (34). The NW was deposited on a bed of narrow gate electrodes covered by 12 nm of HfO2. Successively, Ta (60 nm)/Al (15 nm) source and drain contacts with a spacing of 280 nm were defined by electron-beam (e-beam) lithography and subsequent e-beam evaporation. The latter was preceded by a gentle in situ Ar etching to remove the native oxide of the NW. The Ta/Al contacts were measured to be superconducting below a critical temperature, TC ~ 0.8 K, which is consistent with values reported for Ta in the crystalline phase (TC = 0.67 to 0.9 K (35)).

Device 2, shown in Fig. 4A, was fabricated from the same batch of InAs NWs as device 1. A 70-nm-diameter InAs NW was deposited on a single local back gate covered by 10 nm of HfO2. Although the contacting procedure of the NW was similar to device 1, a Nb (5 nm)/Ta (80 nm) bilayer was used instead. The reasoning behind the inclusion of a thin Nb underlayer was the promotion of the high-TC α phase; however, the convoluted superconducting gap was found to be as reduced as without Nb and TC = 0.7 K. A side gate separated by 300 nm of vacuum was defined at the same moment as the contacts. B was applied at an 11° angle with respect to the long axis of the NW.

SUPPLEMENTARY MATERIALS
Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/5/7/eaav1235/DC1

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