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Evidence for spin-polarized bound states in semiconductor–superconductor–ferromagnetic-insulator islands

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We report Coulomb blockade transport studies of semiconducting InAs nanowires grown with epitaxial superconducting Al and ferromagnetic insulator EuS on overlapping facets. Comparing experiment to a theoretical model, we associate cotunneling features in even-odd bias spectra with spin-polarized Andreev levels. Results are consistent with zero-field spin splitting exceeding the induced superconducting gap. Energies of subgap states are tunable on either side of zero via electrostatic gates.

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In hybrid quantum devices with both ferromagnetic and superconducting components, competition to align electron spins or pair them into singlets can result in complex ground states and corresponding electrical properties [1–9]. Recently, the coexistence of proximity-induced superconductivity and ferromagnetism has been demonstrated in hybrid semiconducting nanowires [10]. Coulomb-blockade spectroscopy of superconducting quantum dots provides a window into subgap states and corresponding electrical properties [1–9]. Recently, spins or pair them into singlets can result in complex ground states and corresponding electrical properties [1–9]. Recent investigations of CoFeB and ferromagnetic insulator EuS on overlapping facets. Comparing experiment to a theoretical model, we associate cotunneling features in even-odd bias spectra with spin-polarized Andreev levels. Results are consistent with zero-field spin splitting exceeding the induced superconducting gap. Energies of subgap states are tunable on either side of zero via electrostatic gates.

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processes [37] through a next-to-leading order expansion in the T matrix (see Supplemental Material [35]). Elastic cotunneling gives a bias independent background conductance, while inelastic cotunneling leaves the system in an excited state yielding steps in $G$ when the bias matches excitation energies.

Theoretical values for differential conductance, $g$, of a Coulomb island containing a single spin-split ABS as a function of voltage bias, $v$, and gate-induced charge offset, $n_G$, is shown in Fig. 2(a), where $\epsilon_{\uparrow} > \Delta$ and $\epsilon_{\downarrow} \equiv \epsilon$ are energies of the two spin branches. The main experimental features are captured qualitatively in this simple theoretical model. In particular, the bias value of the conductance step alternates between even (e) and odd (o) island parities. Steps in differential conductance, marked by the red and blue ticks, correspond to transitions between ground and lowest excited states, as illustrated in Figs. 2(b) and 2(c). Red lines marking $\Delta - \epsilon$ for odd valleys and $\Delta + \epsilon$ for even valleys correspond to processes that change the parity of the subgap state, while the blue lines marking $2\Delta$ for both valleys correspond to processes that break Cooper pairs without changing parity. Cotunneling processes involving higher-energy intermediate states with $\pm 1$ charge on the island are shown in Figs. 2(d)–2(f).

Theoretical spectra for spin-degenerate or weakly spin-split ABS show a denser pattern of cotunneling steps associated with excitations to spin-flipped states at fixed charge, as shown in Fig. S3 in the Supplemental Material [35]. A qualitative comparison shows that the experimental data from Fig. 1(c) agree better with the spin-polarized rather than the spin-degenerate theory (see the discussion and Figs. S4 and S6 in the Supplemental Material [35]). Previous measurements of a similar hybrid island, but without the EuS shell, had shown Coulomb spectra that are consistent with the theory for a spin-degenerate bound state; see Fig. S7 in the Supplemental Material [35].

Returning to experiment, transport data for the 400 nm device on wire 2 shown in Fig. 1(c) yield $\Delta + \epsilon = 120 \mu$eV and $\Delta - \epsilon = 60 \mu$eV, giving $\epsilon = 30 \mu$eV and $\Delta = 90 \mu$eV, consistent with the $2\Delta$ feature at 180 $\mu$eV. The data in Fig. 1(c) give a slightly smaller $\Delta = 60 \mu$eV. We note that $\Delta$ can be gate-voltage dependent. In general, the induced gap is

FIG. 1. (a) Schematic of hybrid InAs nanowire showing EuS and Al layers on overlapping facets of a hexagonal InAs nanowire. (b) Scanning electron micrograph of wire 1. Enhanced false coloration highlights the 400 nm island measurement setup. (c) Differential conductance, $G$, as a function of voltage bias, $V$, and upper-gate voltage, $V_l$, for the 400 nm island on wire 1 at zero applied magnetic field. Steps in conductance indicated by arrows alternate between $V = 30$ and 90 $\mu$V.

FIG. 2. (a) Calculated conductance, $g$, as a function of voltage bias, $v$, and charge offset, $n_G$, for a superconducting island with a superconducting gap $\Delta$, charging energy $E_C = 5\Delta$, Zeeman energy $E_Z \gg \Delta$ (shown here for $E_Z = 100\Delta$), and a spin-polarized subgap state at energy $\Delta = \Delta/4$ (see Supplemental Material [35]). Steps in $g$ due to cotunneling are visible at $v = \Delta - \epsilon$ for odd (o) and $v = \Delta + \epsilon$ in even (e) valleys as well as at $v = 2\Delta$ in both valleys. The calculations were done for finite temperature $k_B T = 0.03 \Delta$. (b) and (c) Schematic superconducting density of states for odd (b) and even (c) valleys in (a) with excitations indicated by arrows. (d) In the odd ground state, a quasiparticle in the bound state exits the island, while another quasiparticle leaves the island, while another quasiparticle tunnels into the continuum. (f) In the even ground state, a spin-down quasiparticle from a broken Cooper pair is excited into the empty bound state and the spin-up quasiparticle leaves the island, while another quasiparticle tunnels into the continuum. The total energy cost of the process is $\Delta + \epsilon$. (d) odd $\Delta - \epsilon$, (e) odd/even $2\Delta$, (f) even $\Delta + \epsilon$. (e) $E_Z > \Delta$. (f) $E_Z > \Delta$. 

[35]
on the more positive side. The measured charging energy $E_c$ measurement.

Coulomb valleys. The larger $V$ to even-odd periodicity again at $V = 0.2$ V. The onsets of the lower-energy steps in both valleys align with $\Delta = 90 \mu eV$, indicating that $\epsilon \approx 0$.

We interpret the evolution as reflecting a subgap state that gradually decreases to zero energy as $V_L$ is varied. In other words, the gate-induced electric fields change the electrostatic environment of the hybrid nanowire, thus modifying the parameters of the subgap state and effectively changing its energy. In the present context, the evolving spin-mixing angle at the superconductor-ferromagnetic insulator interface [25] contributes to the gate dependence of $\epsilon$.

Similar measurements at various $V_L$ are shown in Fig. S8 in the Supplemental Material [35]. The even-odd structure in the amplitude of the finite bias conductance steps is expected theoretically and reflects the relative phase difference between electron and hole components of the subgap state (see the discussion and Fig. S9 in the Supplemental Material [35]).

To investigate $\epsilon$ dependence on the electrostatic environment we look over a wider range of gate voltages. The observed even-odd pattern crosses smoothly through 1e periodicity, reflecting the continuous evolution of $\epsilon$ across zero. This is shown in Fig. 4(a) as a function of upper-gate voltage, $V_U$, for the 800 nm island on wire 2. Both the onsets of the high-bias features, at values $c_i$ and the peak spacings, $s_i$, alternate in magnitude. Subscripts $i = 1$ and 2 denote the two different charge occupancies of the island. We define $c_1 = (\Delta + \epsilon)/e$ and $c_2 = (\Delta - \epsilon)/e$, then take the difference between consecutive $c_i$ to extract the subgap-state energy $\epsilon$ as a function of $V_U$ as shown in Fig. 4(b). Within the measured

considerably smaller than the parent Al gap in these wires. The deduced $E_c = 430 \mu eV$ is larger than $\Delta$, consistent with the even-odd periodic Coulomb pattern. The sharp spectral features at the degeneracy points indicate a discrete subgap state.

Decreasing $V_L$ from +0.2 V to 0 modifies the Coulomb blockade peaks from distinctly even-odd to 1e-periodic at zero bias, with consecutive diamonds differing only by the intensity of step features at finite bias, as seen in Fig. 3(b). The onsets of the lower-energy steps in both valleys align with $\Delta = 90 \mu eV$, indicating that $\epsilon \approx 0$.

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![Graph](image_url)

**FIG. 3.** (a) Differential conductance, $G$, as a function of source-drain bias, $V$, and upper-gate voltage, $V_U$, for the 400 nm island on wire 2. A clear even-odd Coulomb diamond pattern is visible with an inelastic onset in $V$ at $\Delta + \epsilon = 120 \mu eV$ for the bigger diamond and $\Delta - \epsilon = 60 \mu eV$ for the smaller one, as well as an additional step at $2\Delta = 180 \mu eV$ in both diamonds, giving $\Delta = 90 \mu e V$ and $\epsilon = 30 \mu e V$. The data were taken at a fixed lower-gate voltage $V_L = 0.2$ V. (b) Similar to (a) measured at $V_L = 0$, giving nearly 1e-periodic Coulomb diamonds with two steps in $G$ for all diamonds at $\Delta = 90 \mu e V$ and $2\Delta = 180 \mu e V$, indicating $\epsilon \approx 0$. The measured charging energy $E_c = 430 \mu e V$.

![Graph](image_url)

**FIG. 4.** (a) Differential conductance, $G$, measured for the 800 nm island on wire 2 as a function of source-drain bias, $V$, over an extended range of the upper-gate voltage, $V_U$. The Coulomb blockade pattern evolves from even-odd at $V_U = -1.47$ V, through 1e around $V_U = -1.43$ V, to even-odd periodicity again at $V_U = -1.35$ V, visible in both inelastic cotunneling onsets $c_i$ and peak spacings $s_i$, where $i = 1$ and 2 denotes Coulomb valleys. The larger $i = 1$ diamonds on the negative side of the measured gate voltage range become smaller than the $i = 2$ diamonds on the more positive side. The measured charging energy $E_c = 320 \mu e V$ is smaller than for the 400 nm island. The data were taken at $V_L = -0.7$ V. (b) Subgap state energy, $\epsilon$, inferred from step heights (black) and peak-spacing differences (red); $\epsilon$ decreases monotonically from roughly $10 \mu e V$ through $0$ to $-10 \mu e V$ as the gate voltage is increased. Black error bars represent standard errors from the $c_1$ measurement at the positive and negative $V$; red error bars were estimated by propagation of error from the Lorentzian peak fitting and lever arm, $\eta$, measurement.
range of $V_1$, $\epsilon$ decreases monotonically from $+10$ $\mu$eV to $-10$ $\mu$eV. Independently, values for $\epsilon$ were extracted from Coulomb peak spacing at zero bias [11,20,38]. For $E_C > \Delta > \epsilon$, peak spacings are given by $s_1 = (E_C + \epsilon)/\eta$ and $s_2 = (E_C - \epsilon)/\eta$, where $\eta$ is a dimensionless lever arm measured from the slopes of the Coulomb diamonds. The subgap-state energy inferred from the Coulomb peak spacing difference agrees well with $\epsilon$ deduced from finite bias steps, as shown in Fig. 4(b). Good qualitative agreement between measured and computed spectra is shown in Fig. S10 in the Supplemental Material [35]. A similar analysis for $\epsilon$ as a function of $V_1$, where the subgap state approaches but does not cross zero energy, is shown in Fig. S11 in the Supplemental Material [35].

The sign of $\epsilon$ depends on the definition of $c_i$ and $s_i$. Assuming strong zero-field spin splitting at zero applied magnetic field leaves it ambiguous whether or not a level has crossed zero energy. We therefore cannot label the even and odd valleys with certainty. We note that while in principle the evolution of $\epsilon$ with applied magnetic field contains information on the spin projection of the bound state and hence the ground-state parity, we are not able to determine if the field predominantly affects the Zeeman splitting or the magnetization of the EuS (see Ref. [10]). Representative magnetic-field data for both islands on wire 2 are shown in Figs. S11 and S12 in the Supplemental Material [35].

Finally, we note that for specific gate configurations there are no inelastic cotunneling steps present in Coulomb diamonds (see Fig. S14 in the Supplemental Material [35]). This can be understood within the model as resulting from the condition $\epsilon > \Delta$, yielding a cotunneling background for all voltage-bias values within odd valleys and nonzero conductance above $2\Delta$ in even valleys.

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