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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 spectra [11] and their spin structure [12].

Multiple Andreev scatterings at superconducting boundaries of a small normal conductor give rise to Andreev bound states (ABSs) [12]. The states can carry supercurrent through the normal region and appear in tunneling spectroscopy as discrete levels below the superconducting gap [13,14]. Coulomb effects modify transport via ABSs [15,16], for instance, resulting in supercurrent reversal [17,18]. When magnetic fields [19,20] or magnetic materials [21] are involved, spin-degenerate ABSs split and become spin selective, as seen in tunneling spectroscopy [22] and circuit quantum electrodynamics measurements [23]. The spin-active interface between a superconductor and, for example, a ferromagnetic insulator [24] can also lead to spin-split ABSs [25] or, in some cases, triplet superconductivity [26].

Recently, a new class of triple-hybrid materials was realized based on semiconducting InAs nanowires with strong spin-orbit coupling and large g factor, coated with epitaxial superconducting Al, and ferromagnetic insulator EuS shells [27,28]. We investigate nanowires with hexagonal cross-sections and partly overlapping two-facet shells, as shown schematically in Fig. 1(a). Tunneling spectroscopy into the ends of long grounded hybrid wires [10] showed signatures consistent with topological superconductivity, as recently investigated theoretically [29–34].

Here we report transport through Coulomb islands, 400 and 800 nm in length, made from the same batch of wires with normal metal leads and several top- and side-gate electrodes that independently control tunnel-barrier conductances and charge occupancy [Fig. 1(b)]. We observe characteristic features in Coulomb blockade that indicate gate-dependent, discrete subgap states whose energy can be tuned to zero. Qualitative comparison of cotunneling spectra to theoretical models suggests that the subgap states are spin polarized at zero magnetic field, as discussed in detail below.

Spectroscopy of four Coulomb island devices fabricated on two wires (denoted wire 1 and wire 2) showed similar results. Measurements were carried out using standard low-noise lock-in techniques in a dilution refrigerator with a base temperature of 20 mK, equipped with a three-axis vector magnet (see Supplemental Material [35]).

Differential conductance, \( G = dI/dV \), of the 400 nm island on wire 1 as a function of source-drain voltage bias, \( V \), and upper-gate voltage, \( V_G \), showed Coulomb diamonds of alternating height [Fig. 1(c)]. Once the tunneling barriers were coarsely tuned, this behavior is typical of all measured devices. Within a Coulomb valley, low-bias \( G \) was suppressed below the experimental noise floor. At higher bias, \( G \) showed a step-like increase at an alternating bias, as seen in Fig. 1(c). The value of \( V \) at which this first-step feature occurs could be tuned using the lower-gate voltage, \( V_L \). A less pronounced second step in \( G \) at higher bias \([around V = 120 \mu V in Fig. 1(c)]\) did not alternate from valley to valley nor varied with \( V_L \) (see Fig. S1 in the Supplemental Material [35]). The charging energy, \( E_C = 300 \mu eV \), measured from the Coulomb diamonds, is larger than the superconducting gap of the parent Al shell, \( \Delta_{Al} = 230 \mu eV \), hence also larger than the induced gap, \( \Delta \), which is reduced by the coupling to EuS [10]. The 800 nm island on wire 1 showed similar even-odd periodic Coulomb blockade with step-like cotunneling features at finite bias (see Fig. S2 in the Supplemental Material [35]).

To understand the conductance features and relate them to ABSs and spin, we model transport through a superconducting Coulomb island, including a single subgap state, spin-split by Zeeman energy, \( E_Z \). Sequential single-electron tunneling through an ABS on the island yields characteristic Coulomb diamonds [11,36]. To account for intermediate strengths of tunnel couplings to both leads we also include cotunneling...
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 $\varepsilon^\uparrow > \Delta$ and $\varepsilon^\downarrow \equiv \varepsilon$ 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 $\Delta - \varepsilon$ for odd valleys and $\Delta + \varepsilon$ 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 $\Delta + \varepsilon = 120 \mu$eV and $\Delta - \varepsilon = 60 \mu$eV, giving $\varepsilon = 30 \mu$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

\begin{align*}
\text{(a)} & \quad \text{(b)} \\
\text{(c)} & \quad \text{(d)} \\
\text{(e)} & \quad \text{(f)}
\end{align*}

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 $\varepsilon = \Delta/4$ (see Supplemental Material [35]). Steps in $g$ due to cotunneling are visible at $v = \Delta - \varepsilon$ in odd (o) and $v = \Delta + \varepsilon$ 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 from a lead tunnels into the continuum. The excitation energy of $\Delta - \varepsilon$ is supplied by the voltage bias. (e) In both valleys, a voltage bias of $2\Delta$ can break a Cooper pair. In that case, one quasiparticle leaves the island, while the other, together with a quasiparticle from a lead, is excited to 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 $\Delta + \varepsilon$.
on the more positive side. The measured charging energy $E_C$ is 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 $\epsilon \approx 0$.

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 $c_1$ 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
range of $V_L$, $\epsilon$ decreases monotonically from $+10 \mu eV$ to $-10 \mu eV$. Independently, values for $\epsilon$ were extracted from Coulomb peak spacing at zero bias \cite{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 \cite{35}. A similar analysis for $\mu$ is a dimensionless lever arm deduced from finite bias steps, $\epsilon$ depends on the definition of $\epsilon$ 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 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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