Tunneling Spectroscopy of Quasiparticle Bound States in a Spinful Josephson Junction

W. Chang,[1,2] V. E. Manucharyan,[3] T. S. Jespersen,[2] J. Nygård,[2] and C. M. Marcus[1,2]

[1]Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA
[2]Center for Quantum Devices, Niels Bohr Institute, University of Copenhagen, Universitetsparken 5, 2100 Copenhagen Ø, Denmark
[3]Society of Fellows, Harvard University, Cambridge, Massachusetts 02138, USA

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The spectrum of a segment of InAs nanowire, confined between two superconducting leads, was measured as function of gate voltage and superconducting phase difference using a third normal-metal tunnel probe. Subgap resonances for odd electron occupancy—interpreted as bound states involving a confined electron and a quasiparticle from the superconducting leads, reminiscent of Yu-Shiba-Rusinov states—evolve into Kondo-related resonances at higher magnetic fields. An additional zero-bias peak of unknown origin is observed to coexist with the quasiparticle bound states.

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Spin impurities in superconductors can drastically modify the state of its host, for instance, by suppressing the transition temperature and by inducing subgap states [1]. Using a hybrid superconductor-semiconductor device, one can investigate this process with precise experimental control at the level of a single impurity [2]. Exchange interaction between the single quantum spin impurity and quasiparticles modifies the order parameter locally, thereby creating Yu-Shiba-Rusinov subgap states [3–7]. For weak exchange interaction, a subgap state near the gap edge emerges from singlet correlations between the impurity and the quasiparticles. Increasing exchange interaction lowers the energy of the singlet state and increases a key physical parameter, the normal state Kondo temperature T_K. At $k_B T_K \sim \Delta$ (Kondo regime), where Δ is the superconducting gap, the energy gain from the singlet formation can exceed Δ, resulting in a level-crossing quantum phase transition (QPT) [1,8–10]. The QPT changes the spin and the fermion parity of the superconductor-impurity ground state and is marked by a peak in tunneling conductance at zero bias [11].

A mesoscopic superconductor-quantum dot-superconductor Josephson junction [Figs. 1(a) and 1(b)] is an ideal device to study Yu-Shiba-Rusinov states because it provides a novel control knob that tunes the exchange interaction via the superconducting phase difference across the junction ϕ. A physical picture of the phase tunability of exchange interaction is the following: A spin $1/2$ impurity is created by trapping a single electron in the lowest available orbital of the dot (assuming large level spacing) with a Coulomb barrier [Fig. 1(c)] [12,13]. At the electron-hole (e-h) symmetry point, the spinful state, $|1,0\rangle$, costs less than both the empty, $|0,0\rangle$, and the doubly occupied, $|2,0\rangle$, states by the charging energy U ($U > \Delta$ suppresses charge fluctuations at energies below Δ). Here, $|n_{\mathrm{dot}}, n_{\mathrm{lead}}\rangle$ denotes the electron (quasiparticle) occupancies of the dot (leads), with arrows giving spin orientations when needed. Spin-flip scattering connects the degenerate states $|\uparrow,\downarrow\rangle$ and $|\downarrow,\uparrow\rangle$ via the virtual population of states $|2,0\rangle$ [Fig. 1(d)] or $|0,0\rangle$ [Fig. 1(e)]. These two scattering channels cause an effective (super)exchange interaction between quasiparticles and the spinful dot. Compared to scattering via $|2,0\rangle$, scattering via $|0,0\rangle$ differs by a phase factor $\exp(-i\phi)$ because it is accompanied by a Cooper pair transfer [Fig. 1(e)]. At $\phi = \pi$ these two scattering channels interfere destructively, making the exchange coupling minimal at $\phi = \pi$ and maximal at $\phi = 0$. Consequently, both the singlet excited state $|S\rangle$ and the doublet ground state $|D\rangle$ acquire a phase modulation, albeit only in higher order processes for the latter [14–21].

The ground state of spinful Josephson junctions have been investigated by previous experiments [22–27]. Phase-biased junctions with weak coupling showed negative supercurrent [22,23], consistent with theoretical predictions of the weak phase modulation of $|D\rangle$ [14–16], while for strong coupling, positive supercurrent was observed [24,25]. The latter was interpreted in terms of a QPT associated with the interchange of states $|S\rangle$ and $|D\rangle$ at $k_B T_K \sim \Delta$ [25–27]. Meanwhile, other experiments have performed tunneling spectroscopy on spinful Josephson junctions without phase control [28–31] or with phase control but away from the Kondo regime [32]. This leaves the effect of phase on subgap states in the Kondo regime unaddressed. Tunneling spectroscopy in similar devices has also been used recently to examine signatures of Majorana end states [33–35].

In this Letter, we demonstrate both phase and gate control of subgap states in a Kondo-correlated Josephson junction ($k_B T_K \sim \Delta$) [2]. We also report the first evidence of a singlet to doublet QPT induced by the superconducting phase difference. Our InAs nanowire Josephson junction has an additional normal metal tunnel probe which allows a measurement of the density of states via tunneling in the region between the superconducting contacts (Al). By using normal metal, we avoid the complication of deconvolving the density of states of the probe from the tunneling conductance. At magnetic fields above the critical field of Al, tunneling into the InAs quantum dot with odd electron occupancy showed Kondo resonances [12] with associated Kondo temperatures, $T_K \sim 1$ K. Near zero field, tunneling into the nanowire revealed the superconducting gap of the Al leads, $\Delta \simeq 150\ \mu$eV, and a pair of subgap resonances (SGR) symmetric about zero bias. For certain parameters in gate and phase, the pair of SGRs crosses at zero bias, which we interpret as a level-crossing QPT. However, no such crossing occurred upon suppressing Δ to zero with an applied magnetic field. Instead, the SGRs evolve smoothly into Kondo resonances, and this transition is typically accompanied by the appearance of a separate zero-bias resonance of unknown origin.

Epitaxially grown InAs nanowires approximately 100 nm in diameter were deposited on a degenerately doped Si substrate with a 100 nm thermal oxide. They were then contacted by two ends of a superconducting loop (5/100 nm Ti/Al) of area $\sim$25 μm^2 [Figs. 1(a) and 1(b)]. For this loop area, the flux period, $h/2e$, corresponds to a perpendicular magnetic field period of 72 μT. A third normal metal tunnel probe (5/100 nm Ti/Au) contacted the nanowire at the center of the 0.5 μm long junction. By adjusting ammonium polysulfide etch times, high (low) transparency was achieved for the barrier between Al (tunnel probe) and InAs [36]. The device was measured in a dilution refrigerator with a base temperature of 20 mK, through several stages of low-pass filtering and thermalization.

When superconductivity in the entire device was suppressed by an applied magnetic field B, diamond patterns characteristic of weak Coulomb blockade (CB) were observed in transport between the loop and the normal lead [Fig. 2(a)]. Consecutive diamonds alternate in size, indicating that the orbital level spacing ξ is comparable to the charging energy, $U \simeq 200\ \mu$eV. The smaller (odd occupancy) diamonds contain backgate-independent (V_{BG}) zero-bias ridges that split at higher magnetic fields (see Sec. 1 of the Supplemental Material [37]), typical of the Kondo effect [12,38]. From the temperature dependence of the zero-bias ridges, we estimate T_K to be in the range of 0.5–1 K (Sec. 3 of the Supplemental Material [37]). Poor visibility of the odd diamonds suggests strong coupling to the superconducting leads ($\Gamma_S \gtrsim U$), and the amplitudes of the Kondo ridges indicate an asymmetry between superconducting and normal contacts [39]. While the estimated asymmetry, $\Gamma_N \sim \Gamma_S/10$, will likely broaden the tunneling resonances, it is sufficient to qualitatively treat the Au lead as a weak tunneling probe.

In the superconducting state ($B \sim 0$), gap-related features were observed at tunnel-probe voltages, $V_T \simeq \pm 150\ \mu\mathrm{V} \simeq \pm\Delta/e$, consistent with the gap of Al. SGRs symmetric about zero bias were also observed [Fig. 2(b)]. Comparison of Figs. 2(a) and 2(b) shows that the positioning (in V_{BG}) of SGRs in the superconducting state coincides with CB and Kondo features in the normal state. The SGRs and their symmetric partners converge towards each other and sometimes overlap in an odd CB valley. In contrast, they are pushed towards the gap edge in the even CB valleys. Cuts of the data in Fig. 2 are shown in Sec. 1 of the Supplemental Material [37].

Based on their qualitative dependence on V_{BG} and ϕ, three categories of SGRs in the case of a spinful dot were identified. (i) For small charging energy, $U < (\Delta, \Gamma_S)$, SGRs do not cross the zero-bias axis for any V_{BG} or ϕ [Figs. 3(a), 3(d), and 3(g)]. The SGR energy is maximal at $\phi = 0$ and minimal at $\phi = \pi$ [Figs. 3(d) and 3(g)]—this is the conventional phase dependence of noninteracting

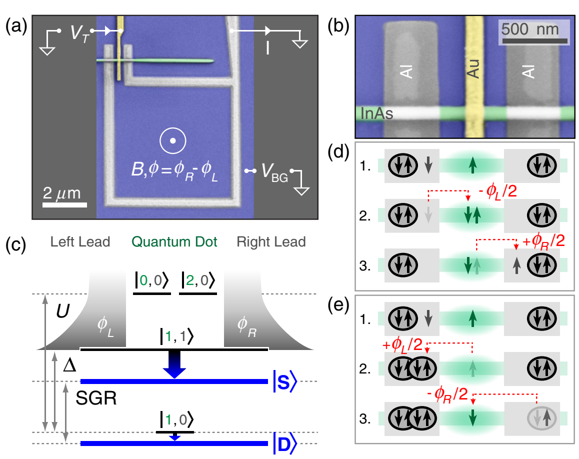

FIG. 1 (color online). (a), (b) Scanning electron micrographs of a lithographically identical device. (c) Lowest energy states of a single-orbital quantum dot at the electron-hole symmetry point for $k_B T_K \ll \Delta$. The states are labeled by their electron or quasiparticle occupation number in the format $|n_{\mathrm{dot}}, n_{\mathrm{lead}}\rangle$. Exchange interaction dresses the states $|1,0\rangle$ and $|1,1\rangle$ as the doublet $|D\rangle$ and the singlet $|S\rangle$ states, respectively. Transition from $|D\rangle$ to $|S\rangle$ produces a subgap resonance (SGR). (d), (e) Phase sensitive spin-flip processes coupling the $|1,1\rangle$ states $|\uparrow,\downarrow\rangle$ and $|\downarrow,\uparrow\rangle$ via virtual occupation of (d) $|2,0\rangle$ and (e) $|0,0\rangle$.

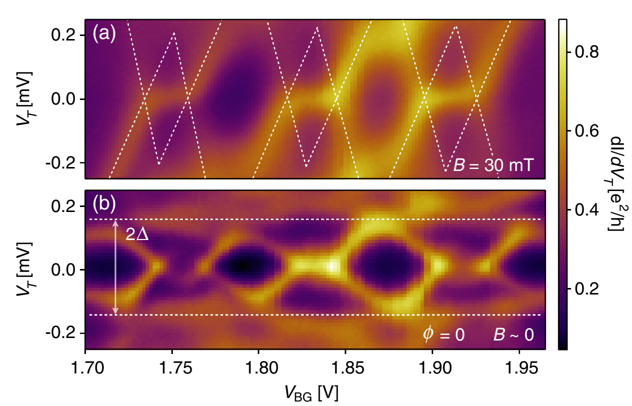

FIG. 2 (color online). Differential conductance as a function of tunnel-probe voltage V_T and backgate voltage V_{BG}. (a) Normal state data, $B = 30$ mT. (b) Superconducting state data, $B \sim 0$ and $\phi = 0$. Coulomb diamonds in (a) and superconducting gap in (b) are highlighted with dotted lines.

Josephson junctions [40]. (ii) For large charging energy, $U > \Delta$, (Sec. 2 of the Supplemental Material [37]) SGRs overlap, crossing zero bias twice as a function of V_{BG} [Fig. 3(c)]. Between zero-bias crossings, the phase dependence of SGR energies is the opposite of the conventional behavior, that is, minimal at $\phi = 0$ and maximal at $\phi = \pi$ [Fig. 3(i)]. We call this a π-shifted phase dependence. Outside the intersections in V_{BG}, the phase dependence of SGR energy is conventional [Fig. 3(f)]. (iii) For moderate charging energy $U \sim \Delta$ [Figs. 3(b), 3(e), and 3(h)], SGRs do not intersect for any V_{BG} at $\phi = 0$ [Fig. 3(b)]. Phase dependence away from the e-h symmetry point is conventional [Fig. 3(e)], but close to the symmetry point, the pair of SGRs intersects twice per phase period of 2π [Fig. 3(h)]. Crossings occur at $\phi = \pi \pm \delta\phi/2$, where $\delta\phi < \pi$ is the phase difference between the two closest crossings [Fig. 3(h)]. With this type of SGR, the phase dependence depends on the phase value itself: it is conventional for $\phi \sim 0$ and π shifted for $\phi \sim \pi$.

In Fig. 4 we examine the magnetic field evolution of three π-shifted SGRs at their e-h symmetry points. The first SGR [Figs. 4(a)–4(c)] is identical to the one shown in Fig. 3(c). Selecting $\phi = 0$ from the full data set (Sec. 9 of the Supplemental Material [37]), the well separated SGRs gradually approach zero bias and merge into a Kondo resonance in the normal state [Fig. 4(b)]. Temperature dependence of the normal-state Kondo peak gives $T_K \simeq 1$ K [12] (Sec. 3 of [37]). Taking $g \sim 13$ from normal-state CB data (Sec. 4 of [37]), the splitting of the Kondo

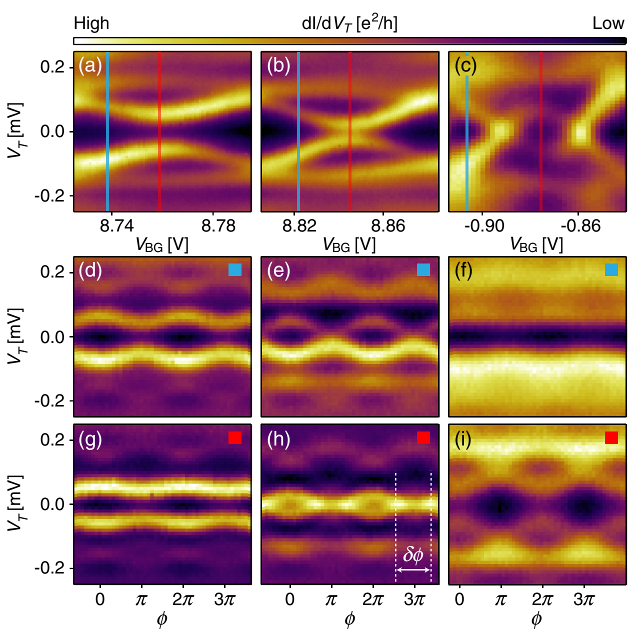

FIG. 3 (color online). Three SGRs arranged in columns of increasing U. (a)–(c) V_{BG} dependence of the SGRs at $\phi = 0$. The lower rows show the corresponding phase dependence off (d)–(f) and on (g)–(i) the electron-hole symmetry point. (d)–(g) Conventional phase dependence, (h) hybrid phase dependence, (i) π-shifted phase dependence.

peak at $\sim$140 mT is consistent with this value of T_K [41] [Fig. 4(b)]. In the other two cases (bottom two rows of Fig. 4), Kondo peaks split at lower fields of $B \sim 50$ mT [Fig. 4(e)] and $B < 20$ mT [Fig. 4(h)], suggesting lower Kondo temperatures.

In the second case [Figs. 4(d)–4(f)], SGRs overlap at zero bias for $\phi = 0$, but are separated for $\phi = \pi$ [Fig. 4(d)]. The overlapping SGRs at zero field evolve continuously into a Kondo resonance as the field is increased into the normal-state regime [Fig. 4(e)]. Phase dependent oscillations of the SGR vanish abruptly at a critical value of field, $B_c = 19.5$ mT [Fig. 4(f)]. The same critical field is observed in Fig. 4(c), and also in higher density regimes of the device (Sec. 5 of the Supplemental Material [37]).

The last case has no phase dependence [Fig. 4(g)], presumably because of poor coupling to one of the superconducting contacts. However, its V_{BG} dependence allows us to establish that this SGR is indeed a π-shifted type (Sec. 8 of [37]). Here, in contrast to the first two cases, the pair of SGRs evolves continuously and directly into a split Kondo peak without ever merging or crossing at zero bias [Figs. 4(h) and 4(i)].

Close inspection of Fig. 4 reveals an unexpected and intriguing feature: a narrow needlelike resonance pinned at zero bias. In Fig. 4(b), this "needle" is absent at $B = 0$ but appears for $B > 10$ mT while the leads are still superconducting. In Fig. 4(d) the needle is hidden by the SGRs at $\phi = 0$, yet it is clearly visible at $\phi = \pi$. In this case, the needle exists at $B = 0$ and merges into the normal-state Kondo resonance at higher field [Fig. 9(f) in the Supplemental Material [37]]. In Fig. 4(h), the needle appears at $B > 10$ mT, similar to the case in Fig. 4(b), despite a large difference in Kondo temperatures. In fact, the strength of the needle appears uncorrelated with T_K of the normal-state Kondo peak (Sec. 10 of [37]). The needle is also distinct from the normal state Kondo resonance as seen in Figs. 4(h) and 4(i), where three separate peaks can be identified: The two peaks flanking the central needle appear to emerge from the SGR at the low-field end and evolve continuously into the split Kondo peaks at the the high-field end. We find that the needle only appears between the two V_{BG} intersection points of π-shifted SGRs, which in turn corresponds to an odd Coulomb diamond (Sec. 6 of [37]). Finally, the needle appears brighter at $\phi = 0$, when the separation between the two SGRs is the smallest [Figs. 4(c) and 4(d)] (Sec. 6 of the Supplemental Material [37]).

We now compare theoretical expectations for SGRs [17] to experimental observations. At the e-h symmetry point of a spinful quantum dot with suppressed charge fluctuations, the phase-tunable exchange interaction detaches a singlet state $|S\rangle$ down from the gap edge [Fig. 1(c)]. Since quantum interference weakens the exchange interaction at $\phi = \pi$ [Figs. 1(d) and 1(e)], a π-shifted SGR is indeed expected (phase modulation of the energy of $|D\rangle$, being a

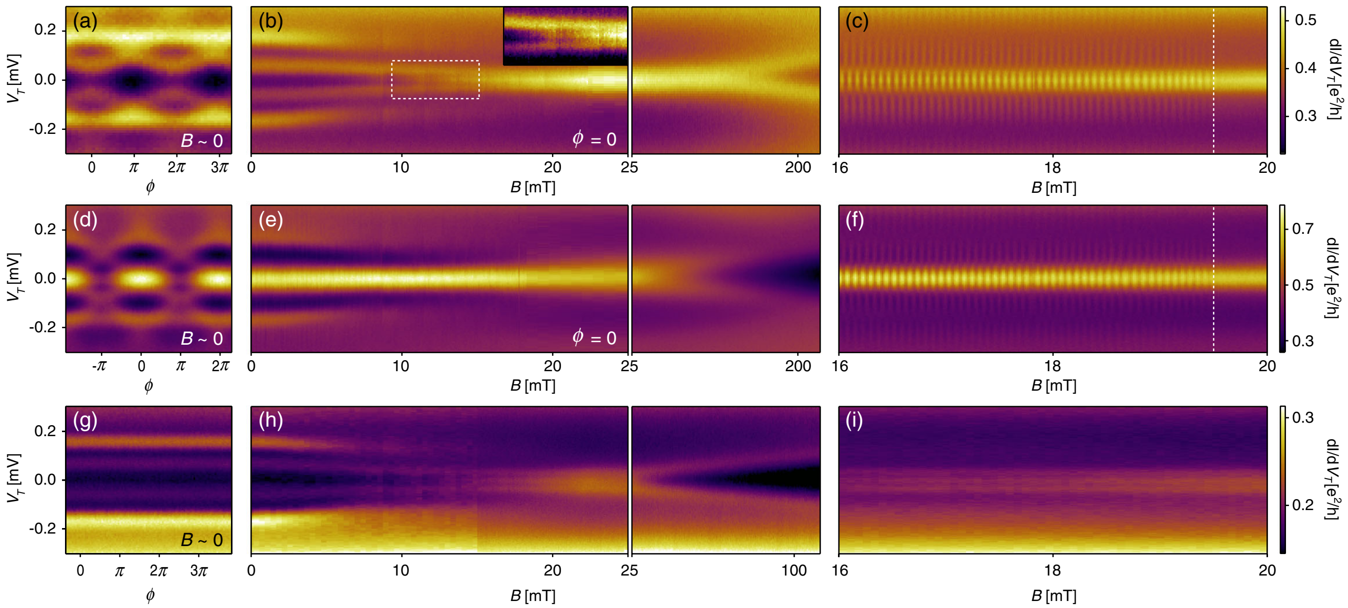

FIG. 4 (color online). Arranged in the order of decreasing T_K, each row shows the evolution of a SGR at the electron-hole symmetry point as a function of phase and magnetic field. The left column shows phase dependence at $B \sim 0$, the center column shows magnetic field dependence at $\phi = 0$, and the right column shows the magnetic field and phase dependence around $B = 18$ mT. To obtain the phase constant panels (b) and (e), we select $\phi = 0$ data points from the full data set. The oscillations of the SGRs disappear abruptly at $B = 19.5$ mT (dotted lines) in both (c) and (f). Inset in (b) is a close-up of the region outlined with dotted lines. A third resonance, pinned at zero bias, is clearly visible in the high contrast color scale.

higher-order effect, is much weaker than that of $|S\rangle$) [18–21]. This is consistent with our experiment, as seen, for example, in Fig. 3(i). Strong coupling to the leads, reflected in the large T_K, should further result in a SGR that is well separated from the gap edge at $\phi = 0$ [9,10]. Detuning V_{BG} towards a neighboring even diamond increases charge fluctuations and mixes either $|0, 0\rangle$ or $|2, 0\rangle$ into $|S\rangle$, thereby lowering its energy. Consequently, one expects a level-crossing QPT to a singlet ground state as V_{BG} approaches an even diamond, in agreement with the zero-bias crossings in Figs. 2(b) and 3(c). This QPT is predominantly governed by the enhanced charge fluctuations away from the e-h symmetry point. Finally, the observed conventional phase dependence in the even state of the dot [Fig. 3(f)] is also expected, because a spinless dot acts effectively as a scatterer in a noninteracting Josephson junction [42].

A more interesting QPT occurs in Fig. 3(h) as a function of phase bias. It corresponds to a situation where the energy gain from the quasiparticle-dot singlet formation makes this state the ground state at $\phi = 0$ but not at $\phi = \pi$. This behavior is known in theory literature as $0'$ junction or π' junction [17,43], and, to our knowledge, has not been reported in previous experiments.

Reducing Δ sufficiently below $k_B T_K$ should result in a level-crossing QPT that is driven entirely by spin fluctuations [1]. Experimentally, we would see a zero-bias crossing of the SGRs at $B < B_c$ as B is increased to suppress Δ. However, this theoretical expectation is not seen in our device as exemplified in Figs. 4(b) and 4(h), perhaps obscured by our current experimental resolution or by the needle feature. The needle may be related to similar features observed in recent experiments [30,31]. An unlikely soft gap in Al may explain such a resonance in terms of conventional Kondo screening. We note, however, that the needle itself does not split with increasing B, as one might expect from a conventional Kondo effect. More intriguingly, the needle appears much stronger at $\phi = 0$ than at $\phi = \pi$, suggesting possible phase dependence and a link to the subgap states (Sec. 11 of the Supplemental Material [37]). While the observed behaviors of subgap states agree at $B \sim 0$ with existing theory on Yu-Shiba-Rusinov states, further theory and experiment are needed to understand the origin of the needle and the magnetic field dependence of the subgap states [44,45].

In summary, tunnel-probe spectroscopy of the density of states of an InAs quantum wire with controlled phase between two superconducting contacts is realized experimentally and investigated in detail. This novel system allows a quantum phase transition between states of different spin and parity to be studied. Crossover between a spinful π junction at low magnetic field and the corresponding Kondo system at higher field shows how these two states connect. An unexplained narrow zero-bias feature at intermediate field with phase dependence is found.

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