Letter

Subgap states in superconducting islands

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We study an interacting quantum dot in contact with a superconducting island described by the Richardson model with a Coulomb repulsion term controlling the number of electrons on the island. This Hamiltonian admits a compact matrix-product-operator representation and can be efficiently and accurately solved using the density-matrix renormalization group. We systematically explore the effects of the charging energy E_c. For E_c comparable to the superconducting gap Δ, the subgap states are stabilized by the combination of Kondo exchange coupling and charge redistribution driven by the Coulomb interaction. The subgap states exist for both even and odd superconductor ground-state occupancy, but with very distinctive excitation spectra in each case. The spectral peaks are not symmetric with respect to the chemical potential and may undergo discontinuous changes as a function of gate voltages.

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The study of long-lived excited states inside the bulk spectral gap of superconductors ("subgap states" for short), induced by impurities and interfaces, drives the development of technologically important quantum devices. For example, the Yu-Shiba-Rusinov (YSR) states that result from the exchange interaction which binds a Bogoliubov quasiparticle at the magnetic impurity site [1–4] are instrumental in realizing topological superconductivity with Majorana edge modes [5]. The excellent understanding of YSR states rests on a theoretical description based on the Anderson impurity model with a superconducting (SC) bath described by the Bardeen-Cooper-Schrieffer (BCS) mean-field Hamiltonian [6–9], which can be tackled using modern impurity solvers [10–16].

A recent development involves devices with the SC material epitaxially evaporated on the nanowire hosting the impurity [quantum dot (QD)] [17,18]. The small SC island in these devices has a considerable charging energy E_c and strong even-odd occupancy effects [19–22] that require an appropriate description [23–29]. This is similar to SC metallic grains [30–34] described by the Richardson model, a charge-conserving Hamiltonian with pairing between the time-reversal-invariant pairs of states in the orbital basis, which is the appropriate generalization of the BCS pairing Hamiltonian to a situation with no translation invariance [35,36]. For weak to moderate pairing and a dense set of levels, the Richardson model is fully equivalent to a BCS superconductor and has the same low-energy excitation spectrum, but it is more general: It also describes the transition to a Bose-Einstein condensate for strong pairing, and it remains applicable for a very small number of levels. Without the impurity, the Richardson model can be expressed in terms of hard-core bosons (paired electrons) and exactly solved via Bethe ansatz (Richardson-Gaudin equations) [37–41]. The impurity breaks integrability by splitting the electron pairs through exchange scattering, thereby precluding this approach. The problem also cannot be solved using conventional impurity solvers because the bath is interacting, while the mean-field decoupling of the charging term leads to incorrect results [42]. Furthermore, the charge-counting trick [43,44] is not applicable to a gapped spectrum [42]. A theoretical tool for this family of problems has therefore been sorely lacking, and some key questions remained unanswered, in particular, whether any states remain present in the gap for odd occupancy of the SC and, if so, what is their nature.

Here, we show that Richardson-type Hamiltonians with long-range (all-to-all) interactions coupled to an interacting QD admit a compact representation in terms of matrix product operators (MPOs) with small 9×9 matrices and can be efficiently solved without any approximations in all parameter regimes using the density-matrix renormalization group (DMRG) [45–47]. Possible extensions include the capacitive coupling between the QD and the island [48], the spin-orbit coupling in the SC, the case of a QD in the junction between two islands, and various multiple-QD problems.

In this Research Letter, we systematically investigate the subgap excitations of the simplest situation: a single QD coupled to a single SC island. The qualitative behavior depends on the ratio of E_c over the SC gap Δ. For $E_c \lesssim \Delta$, the Kondo coupling drives the YSR singlet-doublet transition. For $E_c \gtrsim \Delta$, even-odd effects arise from charge quantization: The occupancy of the SC island varies in steps of one electron similar to a QD in the Coulomb blockade (CB) regime. Subgap states are present also for odd occupancy of the SC, but they disperse very differently compared with even occupancy. The crossover $E_c \approx \Delta$ regime shows complex charging patterns and subgap states with unique properties that strongly depend on the parity of the number of electrons in the superconductor. For parameters that are typical of actual devices, the nature of the subgap states is mixed: It is different from the prototypical YSR states (large-U limit, $E_c = 0$) and Andreev bound states ($U = 0$, $E_c = 0$), as well as from the subgap states of QDs coupled to normal-state Coulomb-blockaded reservoirs.

Model. The Hamiltonian we study in this Research Letter is $H = H_{\rm imp} + H_{\rm SC} + H_{\rm hyb}$ with [24,27,34,49,50]

$$H_{\rm imp} = \epsilon \hat{n}_{\rm imp} + U \hat{n}_{{\rm imp},\uparrow}\hat{n}_{{\rm imp},\downarrow} = (U/2)(\hat{n}_{\rm imp} - \nu)^2 + \text{const},$$

$$H_{\rm SC} = \sum_{i,\sigma} \epsilon_i c^\dagger_{i\sigma} c_{i\sigma} - \alpha d \sum_{i,j} c^\dagger_{i\uparrow} c^\dagger_{i\downarrow} c_{j\downarrow} c_{j\uparrow} + E_c(\hat{n}_{\rm sc} - n_0)^2,$$

$$H_{\rm hyb} = (v/\sqrt{N}) \sum_{i\sigma} (c^\dagger_{i\sigma} d_\sigma + \text{H.c.}).$$

Here, d_σ and $c_{i\sigma}$ are the annihilation operators corresponding to impurity and bath, $\sigma = \uparrow, \downarrow$, $\hat{n}_{{\rm imp},\sigma} = d^\dagger_\sigma d_\sigma$, and $\hat{n}_{\rm imp} = \sum_\sigma \hat{n}_{{\rm imp},\sigma}$. ϵ is the impurity level controlled by the gate voltage applied to the QD, U is the electron-electron repulsion, and $\nu = 1/2 - \epsilon/U$ is the impurity level in units of electron number. The SC has N levels spaced by $d = 2D/N$, where $2D$ is the bandwidth, the orbital indexes i and j range between 1 and N, the dimensionless coupling constant for pairing interaction is α, $\hat{n}_{\rm sc} = \sum_{i\sigma} c^\dagger_{i\sigma} c_{i\sigma}$, and n_0 is the gate voltage applied to the SC expressed in units of electron number. The hybridization strength is $\Gamma = \pi \rho v^2$, where $\rho = 1/2D$ is the normal-state bath density of states. A schematic representation of this Hamiltonian is shown in the top panel of Fig. 1. Most calculations in this Research Letter are performed for $N = 800$ and $\alpha = 0.23$ (magnitude appropriate for Al grains [24]), with $D = 1$ as the energy unit. The corresponding gap in the thermodynamic limit is $\Delta \approx 0.026$ D [42]. The interlevel separation is $d = 2D/N = 0.0025$ D $\approx \Delta/10$; thus the finite-size corrections to BCS theory [22–24,42,51,52] are relatively small [42]. Unless specified otherwise, the QD interaction is $U = 0.1 \approx 4\Delta$, which is a typical value for nanowire devices, and $\Gamma = 0.1U$, which corresponds to intermediately strong coupling. In this Research Letter we focus on the situation where the QD-SC system is not strictly isolated, but in contact with weakly coupled tunneling probes. The ground state (GS) with fixed (integer) total number of electrons $n = n_{\rm gs}$ is determined by the gate voltages ν and n_0. We use (0), (+1), and (−1) as shorthand for the GS and the lowest-energy (subgap) excited states with occupancy $n_{\rm gs} \pm 1$, respectively.

Results. The evolution from the YSR regime to the CB regime is clearly visible in the charging diagrams in the (ν, n_0) plane; see Fig. 1. For small E_c there is a $2e$ periodicity along the n_0 axis with only a weak even-odd modulation of the subgap state energies, while for large E_c, the system instead shows a clear $1e$ periodicity. The transition between the two regimes occurs gradually for E_c of order Δ, with the charge stability regions deforming from a pattern of vertical stripes into a well-defined honeycomb diagram. With increasing Γ, the singlet (blue) regions increase in size because the singlet energy decreases with respect to the doublet energy, while the phase boundaries become smoother (less rectangular) and develop a diagonal slant because for large Γ each gate voltage influences occupancy in *both* parts of the system [42].

To better understand this evolution, in Fig. 2 we follow the dependence on the coupling Γ at fixed $\nu = 1$, where the QD hosts a local moment. In the $\Gamma \to 0$ limit the impurity is decoupled, and for $E_c < \Delta$ the SC is always in a conventional BCS state with even $n_{\rm sc} = \langle \hat{n}_{\rm sc} \rangle$. For $E_c \approx 0$, we uncover the conventional singlet-doublet YSR transition at $T_K(\Gamma)/\Delta = 0.3$ [7,12,53] for a value of Γ that does not depend on n_0; here, $T_K(\Gamma)$ is the impurity Kondo temperature at the given value of Γ. With increasing $E_c < \Delta$ the transition point moves to larger values of Γ around even n_0, where the charging term makes the existence of Bogoliubov quasiparticles energetically unfavorable. The opposite holds around odd n_0. As E_c grows beyond Δ we observe a qualitative change. The SC state in the $\Gamma \to 0$ limit now depends on n_0: For n_0 close to an even integer value, it is a BCS state, while for n_0 close to an odd integer value, an additional unpaired electron (Bogoliubov quasiparticle) sits at the bottom of the quasiparticle band [19] and for $\Gamma \neq 0$ interacts with the electron at the impurity site via exchange interaction, forming a singlet GS. The exact location of the phase boundary depends in a nontrivial way on Γ, U, and E_c due to a three-way competition between Kondo

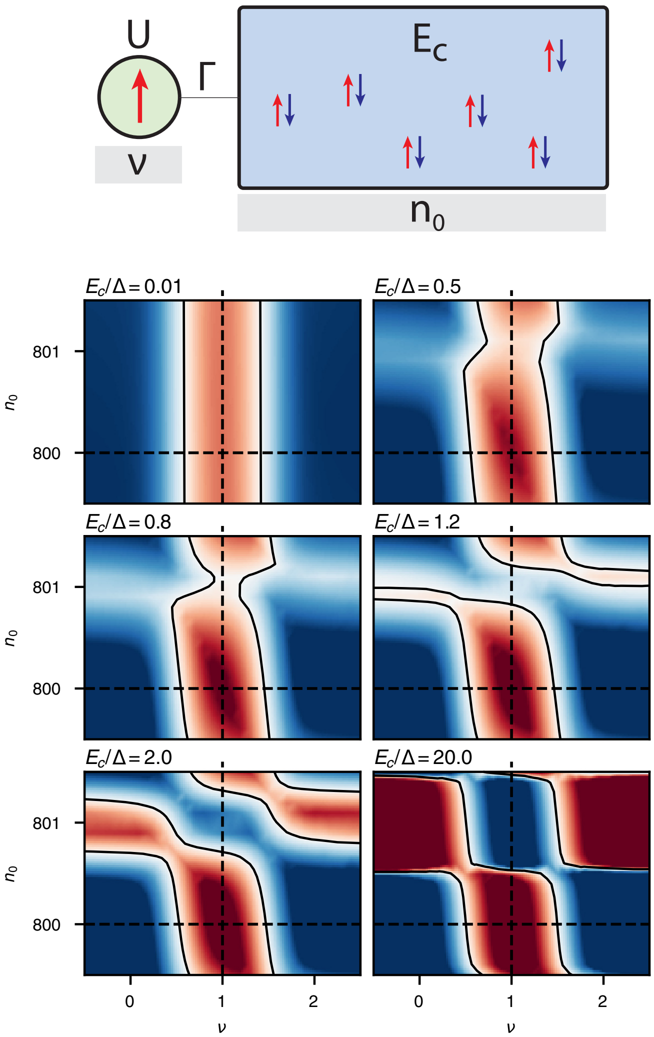

FIG. 1. Top: schematic representation of the system. Bottom: phase diagrams as a function of gate voltages applied to the QD (ν) and to the SC (n_0). Dashed lines correspond to half-filling (particle-hole symmetric) lines of QD and SC at $\nu = 1$ and $n_0 = N = 800$. Red, doublet; blue, singlet. The color indicates the energy difference.

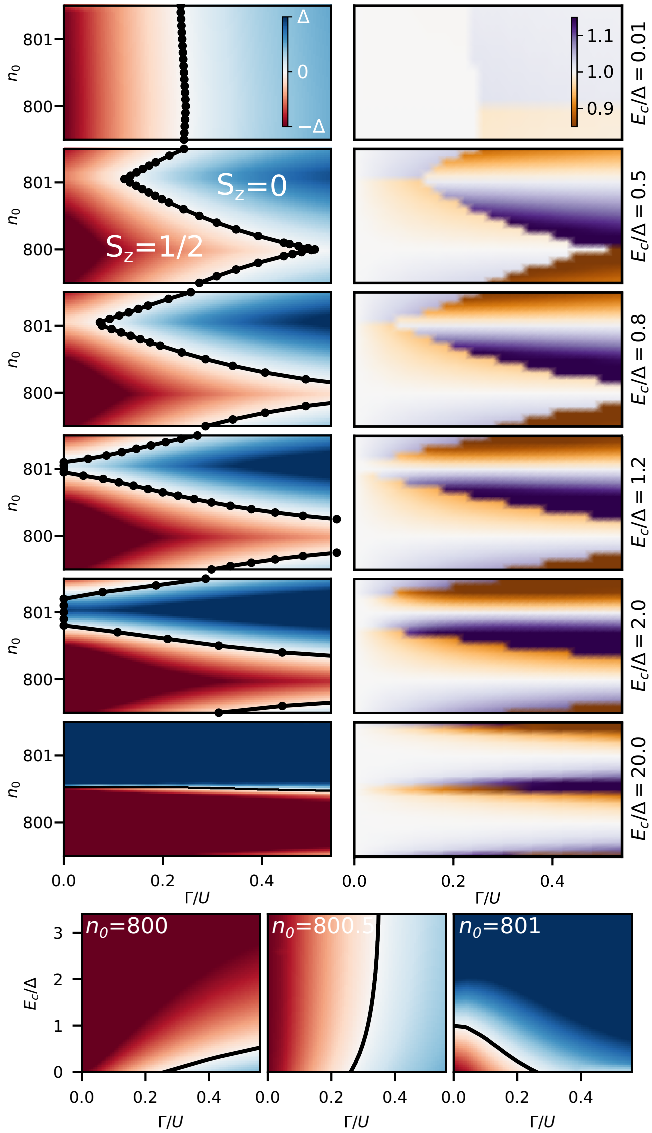

FIG. 2. Evolution from the YSR regime to the CB regime for $\nu = 1$. Energy difference $E^D - E^S$ between the lowest-lying singlet and doublet states in the (Γ, n_0) plane for a range of E_c (left panels), and in the (Γ, E_c) plane for even, half-integer, and odd n_0 (bottom panels). Red, doublet; blue, singlet; black line, quantum phase transition. Right panels: QD occupancy variation.

screening, pairing correlations, and Coulomb interaction. The latter also leads to a strong charge redistribution between the QD and SC in the singlet GS; see right panels in Fig. 2. Figure 3 shows the E_c dependence of impurity occupancy, occupancy (charge) fluctuations, and spin correlations at fixed Γ, for even and odd SC tuning. As E_c increases, the charge in the state penalized by the Coulomb term is redistributed, while the charge fluctuations generally decrease, as expected, except for the doublet state in the odd-n_0 case. The Kondo coupling decreases (increases) with increasing E_c for even (odd) parity of n_0, which is reflected in the spin-spin correlations of the spin-singlet states.

A striking consequence of the Coulomb repulsion is the lack of symmetry in the subgap peak positions except for special points (e.g., $\nu = 1$ and even n_0); see Fig. 4. This is a significant departure from the conventional case with $E_c = 0$, where the peaks are *always* located exactly at $\omega = \pm E_{\mathrm{YSR}}$, so that the spectra take the form of symmetric eye-shaped loops. For $E_c \neq 0$, the states (± 1) have in general different excitation energies $E^{\pm}$ leading to drastic changes in the spectral shapes even for small E_c [see, e.g., black arrows in Fig. 4(f)]. In particular, this leads to *discontinuous* changes in the spectrum when the total occupancy in the GS changes. For instance, as ν increases, E^+ decreases until reaching zero, at which point the former $(+1)$ state becomes the new GS. At this point the former (-1) is no longer spectroscopically visible (i.e., it "disappears"), since it has two electrons fewer than the new GS. The same holds for decreasing ν for E^-. An example of such discontinuous changes in the spectrum is indicated by vertical green arrows in Fig. 4(b). The spectrum behaves even more remarkably for odd n_0 [Figs. 4(e)–4(h)]. For moderate $E_c/\Delta = 0.2$, one observes valence skipping (occupancy jump from 800 to 802, then back to 801) due to a redistribution of charge between the SC and the QD, experimentally visible as a two-sided discontinuity [Fig. 4(f), purple arrows], while for $E_c \lesssim \Delta$ the excitations are pinched at $\nu = 1$. For large $E_c > \Delta$, the spectra eventually transform into straight lines typical of CB systems.

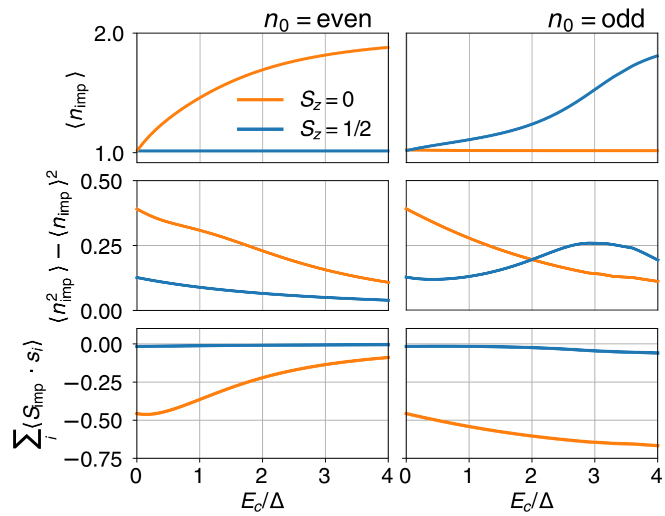

FIG. 3. E_c dependence of subgap state properties at $\nu = 1$.

Discussion. The nature of the subgap states at $E_c \gtrsim \Delta$ is revealed in Fig. 5, where we show the properties of (0), (+1), and (−1) as a function of ν in Fig. 5(a) and the Γ dependence of the excitations at $\nu = 1$ in Fig. 5(b). We first discuss the case of $\nu = 1$. For even n_0, the GS is a decoupled local-moment state, while the states (+1) and (−1) have impurity occupancies differing by more than *half an electron* compared with the ground state due to the cost E_c of changing the SC occupancy, but they still carry some local moment that aligns antiferromagnetically with respect to SC electrons. The excitations detach from the continuum edge at small Γ and shift toward the midgap region with increasing Γ; see Fig. 5(b). The (+1) and (−1) are hence somewhat similar to conventional YSR singlets, although their impurity local moment is reduced not only through the Kondo mechanism, but also by a very large charge transfer to or from the superconductor.

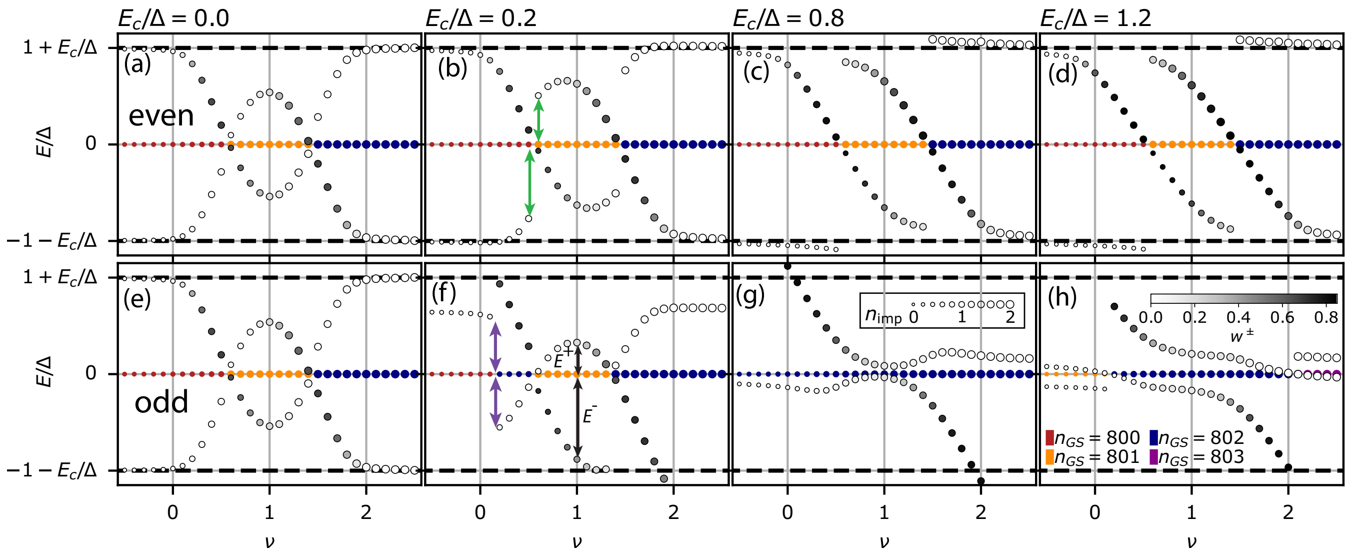

FIG. 4. Subgap spectral functions for even [(a)–(d), $n_0 = 800$] and odd integer [(e)–(h), $n_0 = 801$] tuning of the superconductor occupancy, as a function of the gate voltage applied on the quantum dot. The positions $E^+ = E^{(1)} - E^{(0)}$ and $E^- = E^{(-1)} - E^{(0)}$ indicate the excitation energies of particlelike (+1) and holelike (−1) states; the grayscale shows the corresponding spectral weights $w^\pm$ [see grayscale bar in (h)]. The dots at $E = 0$ provide information about the ground state: The color encodes the GS charge sector, and the size encodes the GS impurity occupancy $n_{\rm imp}$ [see legend in (h)]. The size of gray dots denotes the impurity occupancy in the corresponding excited states [see legend in (g)]. The charge gap for even n_0 and $\Gamma \to 0$ is $\Delta + E_c$ (dashed lines).

The excitations may thus be characterized as being YSR-like, sharing some but not all features of the conventional YSR states at $E_c = 0$. For odd n_0, the states (0), (+1), and (−1) are all similar to each other and carry a local moment at the impurity site. They differ mostly in the presence or absence of the lone Bogoliubov quasiparticle in the SC: (0) has the quasiparticle, while (−1) and (+1) do not; (+1) differs from (−1) by the presence of one additional Cooper pair. Adding an electron to the GS costs E_c and disrupts the singlet, which further costs an energy of order $J_K \propto \Gamma$; however, at the same time a Cooper pair is formed, and the energy Δ is gained. Indeed, the results for $n_0 = 801$ in Fig. 5(b) show for small Γ approximately linear behavior with a zero intercept at $E_c - \Delta = 0.2\Delta$. We thus conclude that for odd n_0 the subgap states are doublets which result from the disruption of the strong local QD-SC singlet formed by the electron in the QD and the lone quasiparticle in the SC (see also Fig. S4 of the Supplemental Material for details [42]); these states have no counterparts at all in $E_c = 0$ systems. Figure 5(b) shows that for even n_0 the excitations have a large weight on the QD, while the opposite is the case for odd n_0, where the electron is mostly added to the SC island.

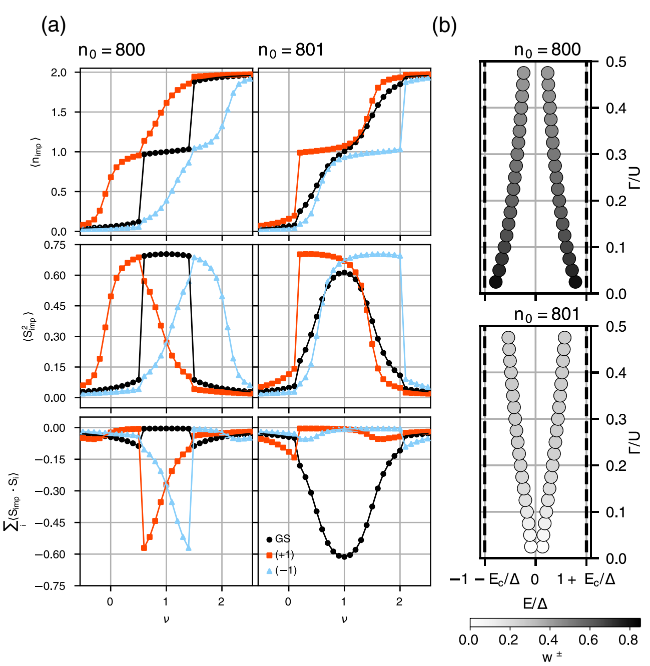

FIG. 5. Subgap state properties for $E_c/\Delta = 1.2$ [Figs. 4(d) and 4(h)]. (a) Expectation values for (0), (+1), and (−1) states vs gate voltage. (b) Γ dependence of the subgap spectra at $\nu = 1$.

The dispersion (ν dependence) of excitations for large E_c (e.g., $E_c \gtrsim 0.8\Delta$) is strongly affected by the charging terms, and it follows the variation of the difference in the impurity occupancy between the ground and excited states, as revealed by comparing Figs. 5(a) and 4. For even n_0 around half filling, the GS occupancy is mostly flat as a function of ν, while the (+1) and (−1) occupancies vary rapidly. This is reflected in an equally rapid variation of the excitation spectrum at the same parameters. Along the same lines, for odd n_0 and away from half filling, the occupancies of (0) and (+1) are similar for $\nu > 1$, while those of (0) and (−1) are similar for $\nu < 1$, and again, this is reflected in the spectral shape (in this case as flat sections).

Conclusion. Subgap states persist in the presence of large charging energy, but they have properties quite unlike those of YSR states [48]. The proposed method can also address the question of quasiparticle poisoning in Majorana islands [17,54], gate sensing of charge-tunneling processes [55,56], superconducting islands on surfaces [57–60], topological superconductivity [61], and the existence of Majorana zero modes beyond mean field [62]. Generalization to multiple bands will find application in multichannel and topological Kondo effects [63–65].

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