Zeeman Tunability of Andreev Bound States in van der Waals Tunnel Barriers

Tom Dvir,[1] Marco Aprili,[2] Charis H. L. Quay,[2] and Hadar Steinberg[1,*]
[1]The Racah Institute of Physics, The Hebrew University of Jerusalem, Givat Ram, Jerusalem 91904 Israel
[2]Laboratoire de Physique des Solides (CNRS UMR 8502), Bâtiment 510, Université Paris-Sud/Université Paris-Saclay, 91405 Orsay, France

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Quantum dots proximity coupled to superconductors are attractive research platforms due to the intricate interplay between the single-electron nature of the dot and the many body nature of the superconducting state. These have been studied mostly using nanowires and carbon nanotubes, which allow a combination of tunability and proximity. Here we report a new type of quantum dot which allows proximity to a broad range of superconducting systems. The dots are realized as embedded defects within semiconducting tunnel barriers in van der Waals layers. By placing such layers on top of thin $NbSe_2$, we can probe the Andreev bound state spectra of such dots up to high in-plane magnetic fields without observing the effects of a diminishing superconducting gap. As tunnel junctions defined on $NbSe_2$ have a hard gap, we can map the subgap spectra without a background related to the rest of the junction. We find that the proximitized defect states invariably have a singlet ground state, manifest in the Zeeman splitting of the subgap excitation. We also find, in some cases, bound states that converge to zero energy and remain there. We discuss the role of the spin-orbit term, present both in the barrier and the superconductor, in the realization of such topologically trivial zero-energy states.

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Introduction.—In a quantum dot (QD) residing at close proximity to a superconductor, the excitation spectrum is governed by an interplay between induced superconductivity, charging energy, and a chemical potential. Such coupling was initially studied by integrating dots into Josephson junctions ("S-QD-S" devices) which may be studied in the strong or weak coupling regime [1]. In the alternative "N-QD-S" geometry, subgap energies are probed directly. Here, the dot is weakly coupled to a normal electrode on one side, and strongly coupled to a superconductor on the other. Charge transfer through the dot and into the superconductor is carried through Andreev processes involving transitions between the ground and excited states [1–5]. These transition energies appear as features in the tunneling spectra, below the superconducting gap Δ. "N-QD-S" systems were realized by evaporating contacts on top of carbon nanotubes [6,7], self-assembled dots [8], and semiconducting nanorwires (NWs) [9–11]. These systems allow for gate tunability of the dot chemical potential, generating a transition between two distinct ground states: an even parity, Cooper-pair-like singlet, and an odd parity, single-electron doublet [4,9,10,12,13]. Tuning the ground state into the doublet ground state is also possible by the application of an in-plane magnetic field. In this case, the doublet state energy is Zeeman split, with the lower energy branch crossing the singlet energy at a finite applied field [9].

In recent years, a major research drive has been aimed at probing the spectra of Majorana excitations, predicted to appear and observed as a zero-bias spectral feature in NWs proximity coupled to superconductors [14–18]. Following these works, it became apparent that dots coupled to superconductors can also exhibit a peak similar to the expected Majorana signal at near-zero energies. This happens when the dot is characterized by a strong spin-orbit coupling (SOC) term [19–21]. More generally, understanding how the Andreev bound states (ABS) spectrum develops at the presence of a SO term is important for distinguishing between trivial and topological states.

A trivial system can exhibit a zero-energy spectral feature when the lower spin branch of the Zeeman split doublet state becomes degenerate with the singlet. Observing this crossover is an experimental challenge: it requires the energy scale $g\mu_B H$ (where g is the Landé g factor, μ_B is the Bohr magneton, and H is the magnitude of the field) to become significant while the superconductor retains a finite gap. To observe such splitting, some studies employed materials with a high g factor [9], although results might be obscured by the diminishing of Δ with H. Here we use an alternative—to couple a quantum dot to an ultrathin superconductor—such as $NbSe_2$. $NbSe_2$ is a van der Waals superconductor, which remains superconducting at the ultrathin limit. Coupling QDs to $NbSe_2$ has two advantages: First, the superconducting gap of ultrathin $NbSe_2$ experiences very little change up to fields of a few Tesla in the plane [22]. Second, $NbSe_2$ and related transition metal dichalcogenide (TMD) materials are characterized by strong Ising SO coupling. It is of interest to consider the role of such SO terms on proximitized dot spectra.

In this Letter we assemble and measure tunnel devices consisting of TMD barriers placed on top of $NbSe_2$, as reported in our earlier works [22,23]. Van der Waals (vdW) layers are known to host atomic defects such as naturally occurring vacancies [24]. These introduce bound states which are often observable via photoluminescence [25,26] or may have the electronic transport signature of quantum dots [27–29]. Here we find that the tunneling spectra through TMD barriers exhibit Andreev bound states which we associate with such dots. The spectra undergo clear Zeeman splitting at the presence of in-plane magnetic field, and are tracked up to 9 T. The majority of dots studied exhibit continuous spectral evolution, with a singlet to doublet crossover at some finite field. In some cases, however, we find a field-dependent transition to zero energy peaks. We suggest these are of trivial topology, and discuss their possible origin.

Results.—Observation of subgap states:

Figure 1(a) shows a sketch of the devices reported in this Letter. Normal-insulator-superconductor (NIS) tunnel junctions were created using the dry transfer technique, by placing a few layer semiconductor TMD (WSe_2) on top of a flake of $2H$-$NbSe_2$ ($NbSe_2$) of thickness ranging between 2 and 50 nm. Normal counter electrodes were created using standard e-beam lithography methods as

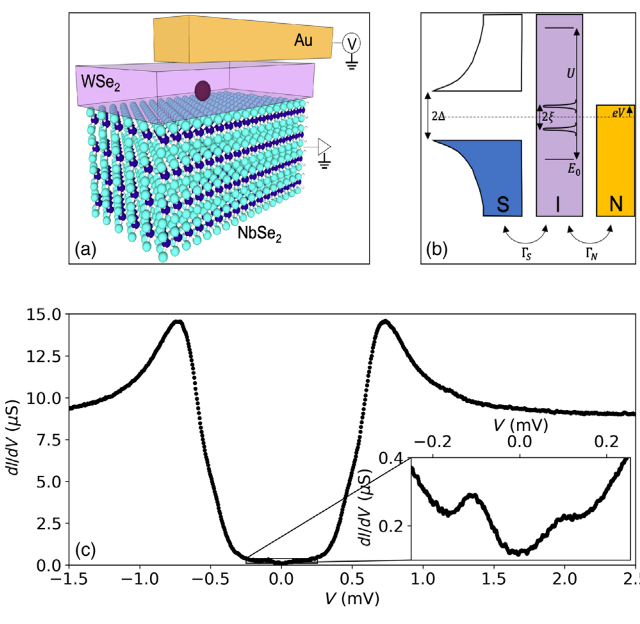

FIG. 1. Subgap states in van der Waals tunnel junctions: (a) Device schematics: a layer of $NbSe_2$ is connected to the ground and is covered by a thin barrier in which a quantum dot is formed. A Ti/Au electrode is evaporated above and connected to a voltage source. (b) Schematic model for tunneling between a superconductor to a normal metal through a quantum dot. Details in main text. (c) Differential conductance of Device 1 at base temperature at zero magnetic field. Inset: magnification of the subgap region showing the two Andreev peaks formed symmetrically around zero.

reported earlier [23]. Typical junction dimensions are in the order of 1–2 μm^2 and barriers are 2–3 nm thick. Measurements are conducted using a standard lock-in technique, where a bias voltage V is applied to the Au counter electrode and the current I and differential conductance dI/dV are measured through a current preamp in Ohmic contact with the $NbSe_2$ bulk. Measurements were conducted at base temperature below 70 mK, with ac excitation in the range of 20–50 μeV.

The problem of a quantum dot that is coupled to a superconductor and to a normal metal is usually formulated in terms of the Anderson impurity model. This model accounts for tunneling between the dot and respective electrodes, and for the on site electrostatic repulsion on the dot. The full solution of the model should also include the dot-lead exchange interaction [30–34], which accounts for Kondo correlations and the formation of Shiba states. This requires sophisticated computational tools and is beyond the scope of this Letter. However we gain sufficient intuition by considering the case of a "superconducting impurity" [4,32]: when the coupling of the superconducting electrode Γ_S is much larger than the coupling to the normal electrode Γ_N and the intrinsic superconducting gap Δ is the largest parameter in the system, an effective on site interaction forms on the dot whose magnitude equals $\Delta_d = \Gamma_S/2$. The effective Hamiltonian then reads:

$$H_{QD} = \sum_{\sigma} E_0 d_\sigma^\dagger d_\sigma - \frac{\Gamma_s}{2}(d_\downarrow^\dagger d_\uparrow^\dagger + \text{H.c.}) + U n_{d\downarrow} n_{d\uparrow} \quad (1)$$

where E_0 is the energy level of the dot, $d_\sigma^\dagger, d_\sigma$ are the creation and annihilation operators for spin σ, $n_\sigma = d_\sigma^\dagger d_\sigma$ is the number operator for spin σ, and U is the electrostatic repulsion energy of the dot, as depicted in the scheme shown in Fig. 1(b). Diagonalization of this Hamiltonian is straight forward: there are two degenerate doublet eigenstates, $|\uparrow\rangle$, $|\downarrow\rangle$ with the energy E_0 and two "singlet" eigenstates that consist of the superposition of zero occupancy state, $|0\rangle$, and the double occupancy state, $|\uparrow\downarrow\rangle$:

$$|\Psi_-\rangle = u_d|0\rangle - v_d|\uparrow\downarrow\rangle, \quad (2)$$

$$|\Psi_+\rangle = v_d|0\rangle + u_d|\uparrow\downarrow\rangle, \quad (3)$$

$$E_\pm = \left(E_0 + \frac{U}{2}\right) \pm \sqrt{\left(E_0 + \frac{U}{2}\right)^2 + \left(\frac{\Gamma_s}{2}\right)^2}. \quad (4)$$

The ground state of the system can be either the doublet or Ψ_-, depending on the interplay between U, E_0, and Γ_s. Tunneling experiments probe the energies corresponding to transitions where the number of electrons in the system is changed by one. In superconducting dots this process requires a transition between singlet and doublet states, with the energies: $\pm\xi = (U/2) - \sqrt{[E_0 + (U/2)]^2 + (\Gamma_s/2)^2}$. While the simplified picture presented here is correct only in the limit of large superconducting gap, and doesn't take into account Kondo correlations, we believe that it qualitatively accounts for the observed data.

Figure 1(c) shows the differential conductance as measured with Device 1. The observed density of states shows a clear superconducting gap with quasiparticle peaks at energies of $\approx 800\ \mu V$ as discussed elsewhere [22]. The subgap spectrum shows two peaks with energies of $\approx 100\ \mu V$ above a parabolic background. Such subgap peaks, observed in many of the tunnel junctions created, are the subject of this Letter, and represent the singlet to doublet transition energy ξ.

Magnetic field dependence: An important knob for the control and study of dot-bound states is a magnetic field. The application of a field lifts the degeneracy between the two doublet states, with a Zeeman energy $E_z = \pm g\mu_B H$. Lifting of the doublet degeneracy allows the distinction between a singlet and a doublet ground state. When the ground state is a doublet, application of a field shifts its energy downwards, increasing the doublet-singlet energy difference and thus merely increasing the observed excitation energy. If, however, the ground state is the singlet state, the excitation energy splits, eventually leading to a crossover when the Zeeman energy equals ξ. Figure 2(a) shows the spectrum of Device 2 at in-plane magnetic fields between 0 and 0.3 T. While this range of fields is insufficient for the observation of Zeeman effect, it is enough to allow for penetration of vortices whose spectroscopic signature overwhelms any other subgap feature [23].

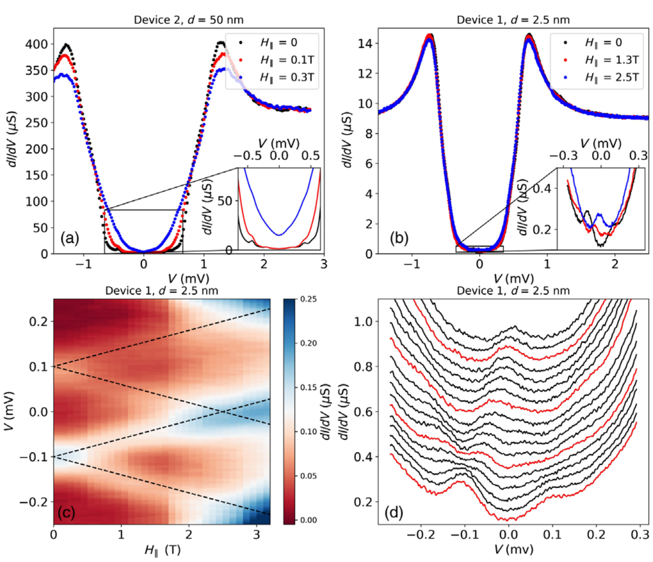

FIG. 2. Subgap states in van der Waals tunnel junctions. (a),(b) Differential conductance of Device 2 (50 nm thick) and Device 1 (2.5 nm thick) at increasing in-plane magnetic field. Insets: enlargement of the subgap spectrum. (c) Color map of the subgap conductance of Device 1 with increasing in-plane magnetic field. Dotted black lines trace the evolution of the subgap peaks with a slope of 40 μeV/T, equivalent to a g factor of 1.3. (d) Shifted differential conductance curves from the subgap region of Device 1, the curves are shown at intervals of 0.2 T. Red curves are shown at intervals of 1 T.

To overcome this issue, we recall that few layer $NbSe_2$ has a strong spin-orbit coupling; thus the application of in-plane magnetic fields has a negligible effect on the spectrum of such superconductors [22]. Figure 2(b) shows the spectrum of Device 1, consisting of a tunnel junction into a four-layer $NbSe_2$ flake, at in-plane magnetic fields ranging between 0 and 2.5 T. While the major features of the superconducting gap seem almost untouched by the field, the subgap spectrum changes significantly, as seen in Fig. 2(c), which follows the evolution of the subgap spectrum of Device 1 with an in-plane magnetic field. It is clear that the two symmetric subgap peaks split. The black traces fit the peak position according to a Zeeman energy with a g factor of 1.3. This observation, of splitting of the subgap peaks, repeats itself in all of the devices measured with g factors in the range of 1.3 to 2 (see the Supplemental Material [35] for subgap spectra of all of the devices measured).

This observation lends support to the claim that the zero field ground state of the proximitized dot is the singlet state. This was previously observed in quantum dots formed at the edges of nanowires and carbon nanotubes, with careful control over E_0 using dielectric gates. Compared with these systems, the proximitized dots formed in the vdW barriers tend to have a singlet ground-state, which points to small charging energy or to E_0 in the close vicinity of the Fermi energy. Furthermore, the observed magnitude of the g factor points to atomic defect, rather than a large quantum dot with an internal band structure that renormalizes g. For such an atomic defect, the broken inversion symmetry that leads to Ising spin-orbit coupling is of no importance. The consistent degeneracy at $H = 0$ shows that the ground state has no preferred magnetic orientation, and the spins of the electrons on the dot are free to interact with the in-plane magnetic field.

When the Zeeman energy equals the zero field ξ, a degeneracy between the singlet and the lower energy doublet state occurs, giving rise to a zero energy conductance peak. Further increase of the magnetic field beyond this crossover field, leads to a shift of the ground state to the lower doublet state, a crossover whose spectroscopic signature is the disappearance of the higher energy split peaks [9]. This is accompanied by a crossing of the lower energy split peaks. Since the conduction in the junctions reported here consists of both tunneling through the quantum dot and tunneling directly between the normal metal and the superconductor, the higher energy peaks tend to merge with the above-gap conduction, thus hindering the observation of the former spectroscopic signal. The latter signal—the crossing of the split peaks at zero energy—was evident in many of the measured devices (Supplemental Material, Fig. S2 [35]).

Zero bias conductance peaks (ZBCP): Figure 3 shows the differential conductance [Figs. 3(a) and 3(c)] and subgap conductance [Figs. 3(b) and 3(d)] of Devices 3

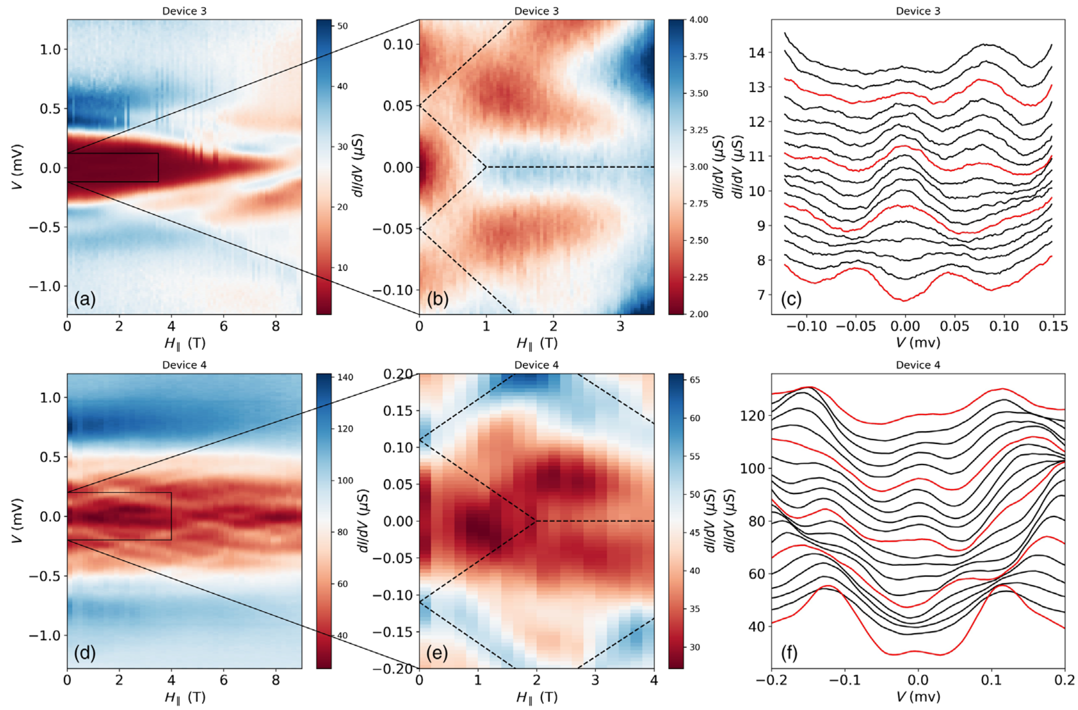

FIG. 3. Formation of zero energy states. (a) Color map of the subgap conductance of Device 3 with increasing in-plane magnetic field. (b) Close up on the subgap region showing the formation of a stable zero bias peak. (c) Shifted differential conductance curves from the subgap region of Device 3, the curves are shown at intervals of 0.2 T. Red curves are shown at intervals of 1 T. (d), (e), (f) Same for Device 4

and 4, respectively. In both devices a stable zero bias peak is formed at fields higher than the crossover field. This feature is stable for approximately 2.5 T, much higher than expected from the spectral width of the subgap peaks. While in Device 3, the ZBCP is merged with the increasing background conductance beyond 3.5 T, in Device 4 the ZBCP splits at 4.5 T, only to reappear at 6.5 T.

ABS pinning to zero energy, beyond some critical in-plane field, is a feature repeatedly seen in proximitized nanowires [16,17,36]. While such ZBCPs are often associated with Majorana fermions, which appear due to nontrivial topology of the superconducting state, recent experimental [37] and theoretical [19–21] studies point to trivial origins of zero energy pinning, calling for extra scrutiny of such results. A different source of zero bias peaks, originating in coupling between the quantum dot and the superconductor, has also been reported in proximitized nanowires [10,38]. In what follows, we discuss alternative interpretations for the emergence of topologically trivial ZBCPs, in the NIS van der Waals system.

In principle, the system discussed here possesses the required ingredients for the formation of topological superconductivity that breaks time reversal symmetry and Majorana bound states: superconductivity, strong spin orbit coupling, and a magnetic field that is applied perpendicular to the direction of the SOC. Formation of topological superconductivity is unlikely, as topological systems require the proximitized region to be large in at least a single dimension, to enable the formation of edge states. Since the subgap features reported here are in all likelihood associated with atomic scale quantum dots, as evident in the observed low g factor, we can rule out the topological origin of the stable ZBCP that is discussed in the context of nanowires [14,15]. The combination of superconductivity, strong spin-orbit coupling and Zeeman field can form trivial nearly zero energy bound states in quantum dots, as recently shown in Refs. [19,20,39,40]. There, a nonsuperconducting quantum dot, in contact with a BCS superconductor, was theoretically shown to host such bound states when a magnetic field crosses a threshold, determined by the superconducting gap and the strength of the SOC. While the details of the discussed model and our van der Walls system are different, we believe that the phenomenon of pinning to zero bias is general. We study here a quantum dot embedded in a semiconducting barrier that hosts a strong intrinsic Ising SOC in addition to Rashba SOC, in proximity to an ultrathin Ising superconductor. This special type of proximitized quantum dot calls for further theoretical modeling.

A different possibility for the formation of zero bias peaks comes from the Kondo effect. It was shown that the ground state of a quantum dot coupled both to a superconductor and to a normal metal can be either the doublet or superconducting singlet as discussed, or a Kondo singlet that involves a superposition between the electrons in the dot and the electrons in the normal lead [10]. Application of magnetic field can induce a SC singlet to Kondo singlet transition as a result of reduction in the magnitude of the superconductor order parameter or filling of the SC gap [38]. Kondo peaks, however, are stable only in magnetic fields smaller than the Kondo temperature T_K, in which the Kondo resonance becomes apparent. Beyond such small fields, the Kondo degeneracy splits [8]. Such splitting is not observed here.

Furthermore, the actual reduction in the magnitude of the superconducting gap can pin the excitation energy of the dot to zero by level repulsion [10]. Few layer $NbSe_2$ is very resilient to the application of an in-plane field. In fields of the range reported in this Letter, the superconducting gap hardly changes [23], rendering both mechanisms—Kondo or repulsion from the gap—implausible.

Conclusions.—In summary, we show that vdW tunnel junctions using TMD barriers may serve as a platform to study the proximity between quantum dots and superconductors. Barrier-defect dots are significantly smaller than those defined by local depletion by gates. They confine carriers to the atomically sized regions, and in addition experience a strong Ising spin orbit coupling. Our results suggest that the zero field ground state of such dots is analogous to the BCS singlet state, which can be tuned by the application of in-plane magnetic field. Finally, the formation of stable zero bias spectral features at finite magnetic fields calls for further theoretical investigation.

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T. D. created the devices. C. Q. H. L., T. D., and M. A. performed the measurements. All authors contributed to data analysis and the writing of the manuscript.

*Corresponding author.
hadar@phys.huji.ac.il

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