Ferromagnetic Proximity Effect in a Ferromagnet–Quantum-Dot–Superconductor Device

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The ferromagnetic proximity effect is studied in InAs nanowire based quantum dots strongly coupled to a ferromagnetic (F) and a superconducting (S) lead. The influence of the F lead is detected through the splitting of the spin-1/2 Kondo resonance. We show that the F lead induces a local exchange field on the quantum dot, which has varying amplitude and sign depending on the charge states. The interplay of the F and S correlations generates an exchange field related subgap feature.

Superconductor-ferromagnet (S/F) heterostructures have been investigated extensively in the past and they have a potential impact on the field of spintronics and quantum computation. Because of the different spin ordering of an s-wave superconductor and a ferromagnet, a large variety of interesting phenomena can be studied at a S/F interface: probing the spin polarization by Andreev reflection [1], π-junction behavior [2,3], ferromagnetically induced triplet superconductivity [4], or ferromagnetically assisted Cooper pair splitting [5].

Here we focus on a novel S/F hybrid device where a quantum dot (QD) is incorporated between a S and a F lead. Recently, the modification of the Andreev reflection of such hybrid systems has been calculated [6,7]. Also configurations, where a QD is strongly coupled to two F leads (F-QDot-F) attracted increasing theoretical [8–11] and experimental [12–15] attention. It has been verified [13–15] that in this situation the spin-↑ and ↓ energy levels of the QD can split by an exchange energy, E_{ex} due to the hybridization between the QD states and the F lead. This proximity ferromagnetism has the same effect on the QD as the presence of an external magnetic field, B. Thus, the exchange splitting is often characterized by the so-called local magnetic exchange field (B_{ex}) in the literature, where $E_{ex} = g\mu_B B_{ex}$. As has been proposed by Martinek *et al.* [16], B_{ex} can be gate dependent and even change sign. Hence, electric controlled reversal of the spin occupation of the QD can be achieved. This has very recently been shown in carbon nanotubes with two F contacts [15].

In this Letter, we show the influence of F and S correlations on the transport properties of F-QD-S devices. We demonstrate that already a single F lead introduces a local exchange field which strongly varies for different QD levels. Furthermore, it even can change its sign within the same charge state. On the other hand, the presence of the S contact serves as a spectroscopic tool. Concentrating on the subgap transport, a novel minigap feature is observed, which is related to B_{ex}.

A typical device is shown in Fig. 1(a). We use high-quality molecular beam-epitaxy grown InAs nanowires

(NWs). NWs are dissolved in 2-propanol and deposited on doped Si substrates with 400 nm insulating SiO_2. Afterwards, Ohmic contacts at spacings of 300–500 nm are fabricated. The F contacts consist of a Ni/Co/Pd trilayer (15 nm/80 nm/10 nm), whereas the S ones consist of a Ti/Al bilayer (10 nm/110 nm). Prior to metal evaporation, argon gun sputtering is used to remove the native oxide layer from the nanowires. In agreement with our previous measurements, QDs form between these contacts, which can be tuned by the voltage applied on the back gate, V_{BG} [17,18]. Measurements are performed at a temperature of 25 mK. A B field is applied parallel to the easy axis of the F contact which is defined by shape anisotropy [see Fig. 1(a)]. The B field allows us to switch the orientation of the F stripe and to control the size of the superconducting gap. To magnetize the F lead the external field is ramped to $B = +300$ mT and then back to $B = 0$ mT before measuring.

In Fig. 1(b) the energy diagram of our devices is shown schematically. The F lead induces an asymmetry of the tunnel coupling (Γ) of spin-↑ and spin-↓ electrons to the QD, described by the tunneling spin polarization
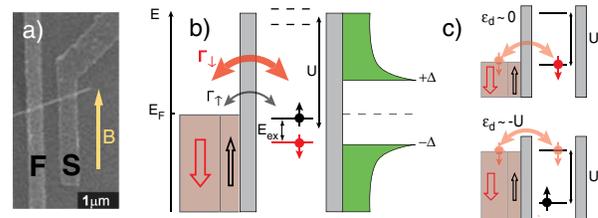

FIG. 1 (color online). (a) A scanning electron microscopy picture of a device. The right lead is a Ti/Al bilayer superconductor, while the left longer one is a Ni/Co/Pd trilayer ferromagnet. The B field is applied parallel to easy axis of the F lead. (b) Schematic view of the F-QDot-S system. The spin degeneracy on the QD is lifted by an exchange splitting induced by the ferromagnetic proximity effect. (c) Because of the spin polarized charge fluctuations, the spin ground state of the QD is opposite when the occupation of the highest orbital level fluctuates between 1 and 0 ($\epsilon_d \approx 0$) or 1 and 2 ($\epsilon_d \approx -U$).

$P = (\Gamma_\uparrow - \Gamma_\downarrow)/(\Gamma_\downarrow + \Gamma_\uparrow)$. This asymmetry is caused by the difference of the spin-↑ and spin-↓ electron density of the F lead at the Fermi energy and by the tunneling matrix elements of these electrons. E.g., in Ni two bands contribute at E_F with opposite spin imbalance and very different tunneling matrix elements to the QD [19]. By hybridization, the spin dependent tunnel coupling generates a spin imbalance on the QD, described as an exchange field. The S lead is represented by the BCS density of states with its energy gap, Δ.

In order to investigate the ferromagnetic proximity effect, the QD is operated in the strongly coupled regime, which allows the study of the cotunneling induced many body spin-1/2 Kondo resonance [20] at odd occupation number of the QD. Kondo resonances split into a doublet in a B field according to the Zeeman energy $E_{\downarrow/\uparrow} = \mp 1/2 g \mu_B B$, thus being also a sensitive tool to visualize B_{ex}. Probing the exchange splitting by the Kondo effect has been demonstrated in C60 molecule [13] and carbon nanotube based QDs [15]. InAs NW QDs have the advantage that the g factor can be comparable to the bulk value of $|g| \approx 15$ [17], making them particularly sensitive for studying the level splitting created by B_{ex}. Much smaller external fields are needed to access the regime, where the exchange energy and the Zeeman splitting are comparable.

The ferromagnetic proximity effect and the therewith connected ground state transition on the QD can be described in a simple model. Using perturbative scaling analysis for a flatband band structure with spin dependent tunneling rates and including finite Stoner splitting of the leads, an analytical formula for the energy splitting of the spin-↑ and the spin-↓ is found [16]:

$$eV_{sd} = g \mu_B B + \Delta_0 + (P\Gamma/\pi) \ln(|\epsilon_d|/|U + \epsilon_d|). \quad (1)$$

Here P is as defined earlier, Γ is the coupling to the F lead ($\Gamma = \Gamma_\uparrow + \Gamma_\downarrow$), U the charging energy of the QD, and ϵ_d the level position of the QD, tunable by V_{BG}. $g\mu_B B$ is the Zeeman splitting due to an external magnetic field, and Δ_0 is a Stoner splitting induced shift. Elaborated numerical renormalization group method calculations also support the result of Eq. (1) [16]. Interestingly, based on Eq. (1), the spin ground state of the QD is different for ϵ_d close to 0 than for ϵ_d close to $-U$, if Δ_0 or the Zeeman term are not too big. This change in the ground state of the QD can be explained by the charge fluctuations between the QD and the F lead [see Fig. 1(c)]. Electrons with majority tunneling spin orientation dominate the charge fluctuations. Thus, when the QD occupation fluctuates between 1 and 0 ($\epsilon_d \approx 0$), the majority tunneling spin occupies the QD preferably. However, when the occupation fluctuates between 1 and 2 ($\epsilon_d \approx -U$), the remaining (nonfluctuating) spin on the QD has the minority spin orientation of the tunneling electrons [15,16]. The transition between these opposite ground states is described by the sign change of the exchange field in Eq. (1). However, if Δ_0 is the dominant term, a roughly constant splitting of the Kondo resonance is expected within a charge state.

In Fig. 2(a) the differential conductance as a function of V_{BG} and source drain voltage V_{sd}, $G(V_{BG}, V_{sd})$, of several charge states of a studied F-QD-S device is presented. Measurements were performed with a standard lock-in technique with an ac excitation of 4 μeV. In accordance with the spin-1/2 Kondo effect, pronounced conductance is seen around ($V_{sd} = 0$ V) in every odd charge state. However, in these states (labeled with numbers 1, 3, 5, 7) the Kondo resonance shows different signatures due to correlations induced by the F lead. State 7 demonstrates a spin-1/2 Kondo situation with a single resonance line at $V_{sd} = 0$ V. As it is shown in Fig. 2(b) this zero bias Kondo resonance splits up linearly with B. In contrast, state 3 exhibits a clear signature of F correlations; i.e., the Kondo resonance has a finite and roughly constant splitting at $B = 0$ mT (see black cross section). Figure 2(c) shows that this splitting is compensated by $B \approx 64$ mT and split again at higher B fields. Thus B_{ex} has an opposite sign as the B field. Another type of B dependence of the Kondo ridge is presented in Fig. 2(d) (measured at $V_{BG} = 2.31$ V), where the zero field splitting of the resonance is further increased by an applied B field. It means that B_{ex} is parallel to B for this state. The three markedly different B field behaviors [Figs. 2(b)–2(d)] demonstrate that B_{ex} strongly depends on the QD level. Even in a small back gate range the amplitude and the sign of B_{ex} varies. This observation highlights the particular importance of the coupling of the QD state to the F lead for the charge fluctuations induced local ex-
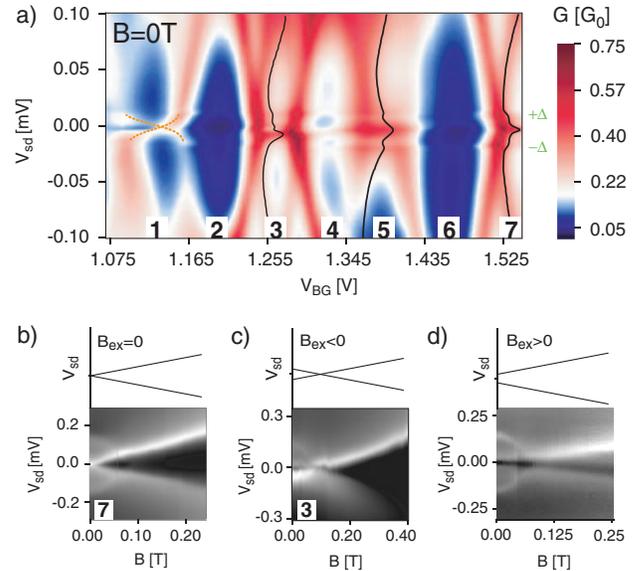

FIG. 2 (color online). (a) Differential conductance as a function of V_{BG} and V_{sd} of a F-QD-S device at $B = 0$ T. B_{ex} modifies the Kondo resonances (odd numbered states) differently. The S lead induces peaks in the conductance at $V_{sd} = \pm\Delta$. B dependence of different charge states: (b) with no signature of B_{ex} [state 7 in Fig. 2(a)], (c) with $B_{ex} < 0$, which is compensated by external B [state 3 in Fig. 2(a)], (d) with $B_{ex} > 0$, which is enhanced by B. Panel (d) is measured in a charge state at $V_{BG} = 2.31$ V.

change field [16]. Note that the different B dependencies also prove that the observed effect cannot solely be described by a stray field, since the stray field cannot depend on V_{BG}.

The most interesting class among the different exchange field manifestations is the one of state 1 [see Fig. 2(a)]. The Kondo resonance is also split for this state. However, the size of the splitting strongly varies with V_{BG}. [The resonance lines are highlighted with dashed lines in Fig. 2(a).] Interestingly, the split Kondo resonance lines also cross the $V_{sd} = 0$ mV value inside the charge state. This dependence suggests that within this charge state B_{ex} first gets smaller as V_{BG} is increased. With further increasing V_{BG} it changes its sign and increases further in the opposite direction. This situation corresponds to the electrically controlled ground state transition on the QD described by Eq. (1) and Fig. 1(c).

Since B_{ex} in situations like state 1 of Fig. 2(a) is strongly gate dependent, a measure for the exchange splitting at different gate voltages can be obtained by measuring the compensation field for which the Kondo peak is restored. Thus, if one measures $G(V_{BG}, B)$ at $V_{sd} = 0$ mV, at each V_{BG} signatures of high conductance are expected if $B = -B_{ex}(V_{BG})$ due to the restored Kondo resonance. Such a plot is presented in Fig. 3(a). The lower or upper ridge (see dashed lines) defines the resonance positions when the occupation of the QD level changes between $0 \leftrightarrow 1$ and $1 \leftrightarrow 2$. The white arrow indicates the sweep direction of B. The high conductance lines with finite slope show the evolution of the Kondo resonance (marked with a white line). At each B field the gate position of the restored Kondo resonance defines the border between the spin-↑ and the spin-↓ ground states. At high external magnetic fields (i.e., $B \gtrsim 0.3$ T) the Zeeman term dominates [see Eq. (1)]. Therefore, considering that the g factor of InAs is negative, the spin-↑ is the ground state for all gate values in high field and the Kondo resonance coincides with the border at $\epsilon_d = 0$. As the B field decreases the restored Kondo peak moves towards $\epsilon_d = -U$, opening back gate regions where spin-↓ is the ground state. At $B = 0$ mT the spin-↓ state dominates, and spin-↑ remains only in a small gate region around $\epsilon_d = -U$, the preferable spin orientation. The gate voltage value as a function of B, where the spin ground state change takes place, can be expressed with Eq. (1) using the condition of $eV_{sd} = 0$. The white line in Fig. 3(a) shows such a curve, giving a reasonable agreement with the measurement using the parameters of $|g| = 12.3$, $\Delta_0 = -90$ mT, and $P\Gamma = 0.22$ meV.

As the polarization of the F lead is switched to the opposite direction by a B field larger than the coercive field of the contact, B_{ex} also changes its sign. This appears as a step in the $G(V_{BG}, B)$ measurement [see Fig. 3(a)] since the splitting turns from $g\mu_B(B + B_{ex})$ to $g\mu_B(B - B_{ex})$. Because of the high g factor of the InAs QD state, this is clearly seen in Fig. 3(a) at the position of the black arrow. Note that in the vicinity of the $0 \leftrightarrow 1$ border at $B = 0$ mT, the ground state is spin-↓. At this border the preferable
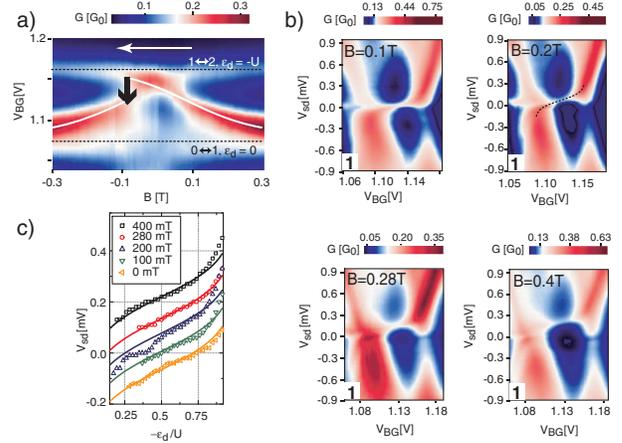

FIG. 3 (color online).   Measurements on state 1. (a) Differential conductance as a function of V_{BG} and B measured at $V_{sd} = 0$ mV. The two horizontal ridges (dotted lines) are the charge state boundaries. The high conductance ridge is the restored Kondo resonance, where $B = -B_{ex}(V_{BG})$. This ridge separates the regions where spin-↑ or spin-↓ is the ground state. The white line is a plot based on Eq. (1) using the parameters obtained from the fits shown in (c). The white arrow shows the sweeping direction of B. The black arrow points out the position where the magnetization of the F lead changes sign. (b) Color scale plots at different magnetic fields. The dashed line highlights the position of the tilted Kondo resonance at $B = 0.2$ T. (c) The evolution of the Kondo resonance is presented for different B fields by symbols. The lines are fits using Eq. (1).

spin orientation on the QD is the majority tunneling spin orientation [see Fig. 1(c)] [15,16]. Since the F lead has been previously polarized into a spin-↓ state by a positive B field, we can conclude that the polarization of the F lead and the polarization of tunneling electrons are the same.

Equation (1) describes the energy difference between the spin-↑ and the spin-↓ states as a function of V_{BG}. This is equivalent to the energy (eV_{sd}), where the Kondo ridge appears. $G(V_{BG}, V_{sd})$ of state 1 is also measured in different magnetic fields. As is seen in Fig. 3(b), the position of the more pronounced split Kondo line moves to higher source drain voltage values as B is increased. The (V_{BG}, V_{sd}) coordinates of this line are read out from the measurements; see, e.g., the dotted line for $B = 0.2$ T. Figure 3(c) summarizes the measured position of the Kondo ridge by symbols at different B fields. The theory described by Eq. (1) nicely fits the experiment (see lines) with the parameters used for the plot in Fig. 3(a).

Since our device has a novel hybrid configuration, i.e., a QD connected to one F and one S lead, the interplay of S and F correlations can be studied. Up to this point, charge states with Kondo physics were investigated, where the Kondo resonance dominates the low energy behavior also for the S state. For such states the S lead induces conductance maxima at $V_{sd} = \pm\Delta$ related to the singularities of the S density of states, which diminish with increasing B field. However, in several charge states of different devices, where spin-1/2 Kondo resonances are not present (neither
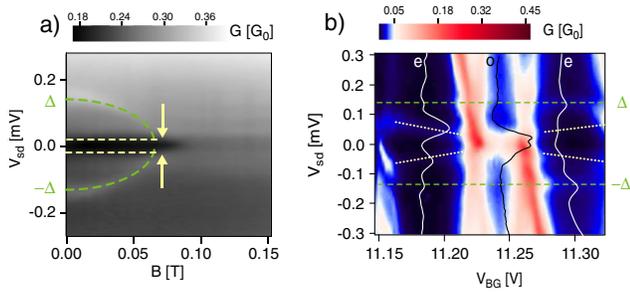
FIG. 4 (color online). (a) $G(B, V_{sd})$ measurement: A subgap feature appears (horizontal dashed lines) at the energy scale of the exchange field. The feature is suppressed above the critical field of the superconductor. (b) $G(V_{BG}, V_{sd})$ at $B = 0$ mT showing the relation of the new subgap structure to B_{ex}. Inside the superconducting gap additional resonance lines appear in even charge states (see dotted lines). These resonances change their position with gate voltage and merge into the exchange split Kondo resonance lines at the border of the odd charge state. The line graphs show $G(V_{sd})$ cuts in the middle of the three charge states.

in the S nor in the normal state), an interesting subgap feature appears. This is shown in Fig. 4(a), where the $G(B, V_{sd})$ measurement of such a state is presented. The conductance shows in a bias window of ≈ 50 μV significantly smaller values than in the rest of the superconducting gap (see horizontal dashed line). This minigap is clearly connected to superconductivity since it is suppressed above the critical field of the S lead. On the other hand the energy scale of the novel subgap feature coincides with the exchange energy observed in the split Kondo states [see, e.g., Fig. 2(d)]. It suggests that the subgap is also exchange field related. Further evidence for such a relation is shown in the $G(V_{BG}, V_{sd})$ measurement at $B = 0$ mT on another device [see Fig. 4(b)]. The middle charge state with odd electron filling (o) shows a slightly split Kondo resonance, while the two even states (e) demonstrate the new subgap feature. As is shown by the dotted lines, the minigap lines of the even charge states merge into the split Kondo resonance at the border between the even and the odd states. Thus, superconductivity induced transport processes in charge states with an even electron number are presented, which seem to be correlated with the exchange splitting.

In conclusion, our measurements demonstrate that the ferromagnetic proximity effect is indeed present in InAs NW based F-QD-S devices. A single F lead induces a local exchange field on the QD, which is strongly level dependent: it even changes sign for a single charge state. A demonstration of controlling the spin ground state of the QD by electric means makes the F-InAs QD-S system a promising building block for spin correlation studies, if implemented into a Cooper pair splitter device [21,22].

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