Electric-field controlled spin reversal in a quantum dot with ferromagnetic contacts
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Manipulation of the spin states of a quantum dot by purely electrical means is a highly desirable property of fundamental importance for the development of spintronic devices such as spin filters, spin transistors and single spin memories as well as for solid-state qubits1–6. An electrically gated quantum dot in the Coulomb blockade regime can be tuned to hold a single unpaired spin-1/2, which is routinely spin polarized by an applied magnetic field7. Using ferromagnetic electrodes, however, the quantum dot becomes spin polarized by the local exchange field8–11. Here, we report on the experimental realization of this tunnelling-induced spin splitting in a carbon–nanotube quantum dot coupled to ferromagnetic nickel electrodes with a strong tunnel coupling ensuring a sizeable exchange field. As charge transport in this regime is dominated by the Kondo effect, we can use this sharp many-body resonance to read off the local spin polarization from the measured bias spectroscopy. We demonstrate that the exchange field can be compensated by an external magnetic field, thus restoring a zero-bias Kondo resonance, and we demonstrate that the exchange field itself, and hence the local spin polarization, can be tuned and reversed merely by tuning the gate voltage.

Since their discovery, carbon nanotubes (CNTs) have been intensively studied for their unique electrical properties. Their high Fermi velocity and low content of nuclear spins make them particularly well suited for spintronics applications using the transformation of spin information into electrical signals. Spin-valve effects in nanojunctions with a CNT spanning two ferromagnetic electrodes have already been observed12–15, and a strong gate dependence of the tunnel magnetoresistance has been demonstrated for CNT quantum dots in both the Coulomb blockade and the Fabry–Perot regime16.

In the intermediate coupling regime, odd-numbered quantum dots exhibit the Kondo effect, seen as a pronounced zero-bias conductance peak at temperatures below a characteristic Kondo temperature16–18, \( \sim T_K \). This effect relies on the conduction electrons being able to flip the spin of the dot during successive cotunnelling events and is therefore expected to be sensitive to spin polarization of the electrodes. As pointed out by Martinek et al.17, quantum charge fluctuations, the electrons ability to tunnel on and off the dot, will renormalize the single-particle energy levels. In the case of ferromagnetic electrodes, this energy renormalization will be spin dependent and break the spin degeneracy on the dot, causing the spin states and thereby the zero-bias Kondo peak to split in two. This tunnelling-induced exchange-field splitting of the Kondo peak was observed by Patsapathy et al.18 in an ungated electromigrated Ni gap holding a \( \text{C}_{60} \) molecule. In contrast, the measurements here were made on a CNT quantum dot coupled to ferromagnetic Ni electrodes and, most importantly, with a back gate that enables us to tune the energy levels on the quantum dot. It is the presence of this back gate that now grants us experimental access to the interesting area of electrical spin control.

A singly occupied level residing just below the Fermi energy of the electrodes is strongly shifted by electrons tunnelling off and then back onto the dot, whereas a level deep below the Fermi energy (by almost the charging energy) is shifted by tunnelling of electrons onto and then off the dot. For spin-polarized electrodes with a difference in the density of spin-up and spin-down states, this implies a spin splitting of the dot level where sign and size depend on the applied gate voltage, that is, the position of the level below the Fermi energy. At the particle–hole symmetric point between the empty and doubly occupied states, the spin degeneracy is expected to be intact19. In a material such as Ni, however, we expect the band structure to be energy dependent and to have a Stoner splitting; this will break the particle–hole symmetry and shift the spin degeneracy point away from the middle of the diamond19,20. Basically, all of the theoretical predictions for this pronounced gate dependence of the local spin states are experimentally verified by the transport measurements presented below.

Our sample geometry is shown in Fig. 1a with an external magnetic field applied in the direction of the Ni electrodes. We observe a clear even–odd effect with a zero-bias anomaly in every second Coulomb diamond, indicating a spin-1/2 Kondo effect with a typical Kondo temperature, \( T_K \sim 1 \text{ K} \). Many of the observed Kondo anomalies showed a gate-dependent splitting and here we discuss two different devices for which this dependence is particularly clear; two other examples are shown in the Supplementary Information.

Magnetic force microscopy images of devices similar to the ones measured indicate that the CNT quantum dot is most likely coupled to one single domain in both the source and the drain electrodes. Applying a strong magnetic field serves partly to align the two contact domains and partly to provide a Zeeman splitting of the local spin. Figure 2 shows the conductance versus bias voltage and external field, measured for a gate voltage tuned to the middle of an odd-occupied Coulomb blockade valley in the Kondo regime. Device 1 (Fig. 2a,b) shows a simple linear behaviour in which the single-domain magnetization and hence the exchange field, \( B_{\text{ex}} \), is aligned with the external field, \( B \). The exchange field can therefore be completely compensated and the zero-bias Kondo peak is seen to be restored at \( B \sim \pm 1.12 \text{ T} \), giving an indirect measure of the exchange field at this gate voltage. In device 2...
Γ can be used to estimate the coupling asymmetry. In terms of the history of the external magnetic field. The difference in splitting is clearly visible at small negative (positive) fields. A similar splitting is found for the two contact domains. At $B > 12$ T, the external field is compensating the exchange field and restoring the zero-bias Kondo peak. A sudden change in $dI/dV$ is observed at $B < -80$ mT due to a switching of one of the two contact domains. At $B > 80$ mT, the other domain has flipped and the device is back in a parallel configuration. The plot of the peak positions from a, offset with $V_g = 5 \mu$V to symmetrize the plot. The black and red lines are fits to the peaks and have a slope of $\pm 1.04 \times 10^{-4}$ V T$^{-1}$ corresponding to $g \approx 1.8$. c, Plot as in a, measured on device 2, sweeping from $-2$ to $2$ T. In this device, the exchange field is never completely compensated, indicating a misalignment of the domain magnetization direction and the applied magnetic field. d, A domain switch causes a jump in the splitting at $B \leq 60$ mT. e, Peak positions from c, offset by $V_g = 12 \mu$V. The black and red lines are fits to the plots where the domain magnetization direction is at an angle of 25$^\circ$ to the external field (see text). f, Plots of the spin-valve behaviour measured on devices 1 and 2 respectively.
tunnelling amplitude to the source (drain), it is expected\cite{8,20} that $g e^2 B_{cc}^{(AP)} = \Delta \epsilon_{cc}^{(AP)} = a P (\Gamma_u + \Gamma_d)$. Comparing the exchange fields in the parallel and antiparallel configurations, for device 1, we deduce that $\Gamma_u / \Gamma_d \sim 3$, which corresponds to a zero-bias Kondo peak height of $(2e^2 / h) 4 \Gamma_u \Gamma_d / (\Gamma_u + \Gamma_d)^2 = 1.5 e^2 / h$ roughly consistent with the value of 1.4 $e^2 / h$, measured in the middle of the Coulomb blockade diamond for a fully compensating magnetic field near $B = \pm 1.12 \ T$.

In a Coulomb blockade diamond with an odd number of electrons, the left and right charge-degeneracy points correspond to emptying the dot or filling it by one extra electron, respectively. With a finite spin polarization in the electrodes, tunnelling of majority spins, spin-up for example, will be favoured by the higher density of states and a dot state of spin-up will therefore be shifted further down in energy than a spin-down state, as long as one is closer to the left-hand side of the diamond. Owing to the Pauli principle, the majority spins can only tunnel into the dot if the residing electron is in a spin-down state. It is therefore the spin-down state that is lowered the most near the right-hand side of the diamond. This simple mechanism of level renormalization, shown in Fig. 3a–c, is encoded in the exchange field, given to good accuracy by

$$\Delta \epsilon_{cc} (|\epsilon_\delta|) = e \Delta_0 (\Gamma / \pi) \ln (|\epsilon_\delta| / |U + \epsilon_\delta|)$$

(1)

to leading order in the tunnelling amplitude\cite{11} and assuming a constant density of states. $U$ is the charging energy and $\epsilon_\delta$ is the dot level position which is proportional to the gate voltage. Note the strong negative and positive logarithmic corrections for $\epsilon_\delta$ close to 0 or $-U$, respectively (corresponding to the left and right borders of the diamond). In the middle of the diamond, $\epsilon_\delta = - U / 2$, the exchange field is zero except for a constant term, $\Delta_0$, which we ascribe partially to a Stoner splitting between the spin-up and spin-down bands\cite{10,11}, and partially to the spin splitting arising from spin-dependent interfacial phase shifts picked up by dot electrons on reflection on the ferromagnetic electrodes\cite{21}. Note that possible stray fields from the magnetic contacts would also contribute with a gate-independent spin splitting (see the Supplementary Information).

Depending on the magnitude of $\Delta_0$, equation (1) predicts a change of the ground-state spin direction as the localized level is moved from $\epsilon_\delta = 0$ to $\epsilon_\delta = - U$. Such a spin reversal can be observed in Fig. 3d–g and occurs at the conductance peak where the Kondo ridges cross and the spin states are degenerate. The movement of the conductance peak (red dot) as function of magnetic field, seen in Fig. 3d–g, confirms that the ground-state spin can indeed be reversed by changing the gate voltage. Furthermore, the direction of the motion shows that it is predominantly spin-up (that is, the spin along the
white lines, corresponding to the Coulomb peaks. The plot has a
gratifying resemblance to the theory plots shown in refs 10,11.
Inverting equation (1) and using the fitting parameters
derived from Fig. 4a, we arrive at the parameter-free fit seen as the black
line in Fig. 4b.

METHODS

EXPERIMENTAL

The CNTs were grown by chemical vapour deposition on a SiO2 wafer with a
highly doped Si back gate. The ferromagnetic electrodes were made of pure
Ni in strips of thickness ~60 nm, width ~300–1,000 nm and separated by
200–400 nm. All measurements were made in a 1He/D helium dilution
refrigerator, with a base temperature of 30 mK, corresponding to a minimum
electron temperature of 80 mK, and using a standard a.c. set-up with asymmetric bias.

NOTE

Note that the plots in Fig. 3d–g have been recorded before a switch in device 1,
whereas all other measurements taken on device 1 were recorded after. We show
these plots because they bring out most clearly the movement of the degeneracy
point. The corresponding plots recorded after the gate switch are shown in the
Supplementary Information and are qualitatively similar.

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external magnetic field) electrons that tunnel on and off the dot,
giving a ground state with spin-up at the left-hand side of
the diamond and spin-down at the right-hand side. This confirms
earlier observations in conventional superconducting Ni tunnel
junctions22,23 indicating that the tunnelling electrons in Ni have
majority spin, that is positive polarization. Most likely, the more
mobile s electrons taking part in the tunnelling are spin polarized
by their hybridization with the localized d electrons24,25.

To substantiate the detailed gate-voltage dependence of the
exchange field, we have extracted the peak positions from plots such
as those in Fig. 3d–g and fitted them by
\[ \Delta V_{\text{ex}} \] given by equation (1). The result is shown in Fig. 4a, where
the inset shows \( \Delta V_{\text{ex}} - g \mu_B B \) as a function of \( B \). The slope of the
line corresponds to \( g = 2 \) as expected for a CNT. Figure 4b maps out
the movement of the point where the spin states are degenerate as a
function of magnetic field and gate voltage. The degeneracy point
is seen as the white ridge moving between the two nearly horizontal

Figure 4 Gate dependence of the exchange field for different applied magnetic
fields. a, Scatter plot of the dominant Kondo peak position, in bias voltage, as a
function of level position \( \varepsilon \) (in \( V_g \)) gate voltage for device 1. Peak positions are
read off from plots similar to those in Fig. 3d–g (see the Supplementary Information).
The different colours correspond to different magnetic fields. The gate axis has been
displaced and normalized with the charging energy such that the dot is emptied at
\( \varepsilon = 0 \) and becomes doubly occupied at \( \varepsilon = -U \). Solid lines are fits to the data,
using \( (\Delta \varepsilon_{\text{ex}} - g \mu_B B) / e \) defined by equation (1) and with \( g = 2 \). All three curves are
fitted by \( PT / \pi = 0.113 \text{ mV} \). Inset: A fit to the gate-independent term
\( \Delta g \mu_B B / e \) with \( \Delta g = 0.18 \text{ mV} \). b, \( dI/dV \) as a function of gate voltage and
magnetic field measured at zero bias voltage. The two nearly horizontal lines are
Coulomb peaks and the white line moving from the lower to the upper peak as \( B \)
is lowered is a Kondo peak, thus mapping out where the spin states are degenerate.
The black line is described by the function \( \varepsilon = U / (\mu_B B + B + PT / \pi) - c \), where
\( c \) is a constant determined by the first Coulomb peak and \( U \) is the charging energy.
\( c \) and \( U \) are read off from the plot, whereas the other constants are determined from
the fits in a. Note that, like in Fig. 2, a domain switch is seen at \( |B| \lesssim 80 \text{ mT} \).


References


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