Electron Spin in Single Wall Carbon Nanotubes

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Abstract

We review aspects of electrical transport in metallic single wall carbon nanotubes (SWCNT) related to the spin of the conductance electrons. For large contact resistances, \( R \approx h/2e^2 \), a SWCNT exhibits Coulomb blockade, and transmission can only occur, when a gate voltage leads to an energy degeneracy for two different numbers of electrons in the SWCNT. The Coulomb blockade gate voltage change is directly proportional to the addition energy for single electron tunnelling. In certain ideal cases every second of the populated electronic states has a higher addition energy, indicating that two spindegenerate electrons are roomed at each orbital state. A low addition energy therefore corresponds to approaching an even number of electrons. The odd-even alternation can be checked in a magnetic field, since then the odd additional electron may enter in one of two Zeeman states. If the high resistance contact is a tunnel junction, the transmission reflects the density of states. This leads to a direct detection of the so-called Luttinger liquid of the electrons. Ferromagnetic contacts to the SWCNT leads to a conduction which depends on the orientation of the magnetic domains in the contacts. The magnetoresistance effect can be much larger than expected from a simple spin-valve phenomenon. For any intermediate normal metal (Au) contact resistances, \( R \approx h/2e^2 \), the Coulomb blockade may still separate the single electron states in the SWCNT with odd and even numbers of electrons. However, at the lowest temperatures the transmission only shows Coulomb blockade for even number of electrons. In the situations with odd number of electrons a coherent tunnelling process dominates. This shortage of the blockade is rooted in the Kondo states formed in the two Au electrodes by exchange interaction due to the spin state in the SWCNT. This tunnelling process is a result of a net spin on the SWCNT and consequently a spin degeneracy. A triplet state is forced into degeneracy with the singlet state in a suitable magnetic field. The situation in a magnetic field is particularly simple in a SWCNT, in contrast to conventional quantum dots, because the tiny diameter of the SWCNT practically speaking precludes orbital effects.

1. Introduction

After the discovery of multi wall carbon nanotubes and 2 years later single wall carbon nanotubes both by Iijima et al. [1] by use of transmission electron microscopy a considerable activity started in order to explore other physical properties. Some of the most spectacular results have been electrical transport data at low temperatures on multi wall carbon nanotubes by Ebbesen [2] and most recently on single wall carbon nanotubes (SWCNT) by Dekker and McEuen [3,4]. This combination of the structural discoveries and the subsequent transport experiments on carbon nanotubes resulted in the prestigious Agilent (Hewlett Packard) Prize of the European Physical Society in 2001/2002 to Iijima, Ebbesen, Dekker and McEuen. In the following we will highlight that part of the electrical transport properties of SWCNTs, which is related directly to the electron spin.

Our paper describes some astonishing electrical transport phenomena connected to spin states in SWCNTs, some of which are new and some of which have been published by us or others over the last three years [see the reviews 5,6,7,8].
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2. High resistance metallic contacts: Coulomb blockade and addition energies

The SWCNTs and the Au contact pads are placed on a 300 nm thick SiO$_2$ layer on top of a highly n-doped Si substrate, which acts as a conducting backgate right down to mK temperatures. The contact to the SWCNT seems to be mostly at the edge of the Au film as judged from the eigenenergies (see later). If the contact is a tunnel junction and the resistance is high, i.e. 100 kΩ or more, the SWCNT exhibit Coulomb blockade effects in two-terminal conductance measurements [3–5]. Figure 1 shows the conductance, \(dI/dV\), at small bias voltages as a function of the backgate voltage at several temperatures. At thermal energies smaller than the Coulomb energy single electron tunnelling prevails as seen as the pronounced oscillations. Two capacitances must be distinguished to understand single electron tunnelling. The capacitance, \(C\), between the backgate and the SWCNT, determines how much change in backgate voltage, \(\Delta V = e/C\), is required to add one more electron to the SWCNT. The corresponding energy shift, which is imposed on the isolated SWCNT has partly an electrostatic origin determined by the total capacitance, \(C_T\), to all surrounding electrodes, \(\epsilon^2/2C_T\) and the eigenenergy shift, \(\Delta E\), of the state into which the electron enters. The sum of these is the so-called addition energy, which is related to the required gate voltage by a constant of proportionality, \(\alpha\). In order to find the scaling of the gate voltage in terms of SWCNT energy, the conductance \(dI/dV\) is measured as a function of bias voltage for various gate voltages. This is shown as a grey scale plot for one of our SWCNT in Fig. 2 (top), where dark is high conductance. The bias voltage is along the \(y\)-axis and the backgate voltage is along the \(x\)-axis. The addition energy is determined by the ordinate taken to the top point of the diamonds in the diagram. The added electron, which creates the difference between the middle and the left diamond may actually enter into several possible eigenstates and this is seen as the faint dark lines in the figure. As seen, tunnelling into the first excited state in the SWCNT looks in the bias spectroscopy plot Fig. 2 as rather independent on the number of electrons \((n \text{ or } n-1)\) on the tube. This indicates a high degree of regularity of the potential landscape in the SWCNT for the chosen backgate voltage regime. The eigenstates are consistent with having rigid tube becomes metallic. There is about a 1:2 chance for the SWCNT to be metallic. Due to the large gap introduced by the new periodic (perimeter) boundary condition, the SWCNT is an ideal 1-dimensional system even at room temperature.

The SWCNTs used in the investigations presented in this overview are made by either chemical vapour deposition or by laser ablation. Another successful method, which is often used, is the simple discharge technique often used for the fullerenes. The first two methods are primarily known to produce high quality SWCNTs. The laser ablation method sends a high power laser beam into a carbon electrode with a small amount of Ni implanted or otherwise inserted. The evaporated carbon is taken by an Ar gas stream and deposited on a cold finger somewhere else in the chamber. The SWCNT grow quite abundantly with Ni apparently acting as a catalyst for the process. These tubes are then scraped off and suspended in 1,2-dichloro-ethane. Sometimes they are first treated in ultrasound in a strong acid to shorten them, but this may lead to defective SWCNTs; a disentanglement of the interwoven tubes can be obtained by ultrasound. A drop of these suspended tubes is put on a SiO$_2$ substrate and after the SWCNTs have fastened to the surface the 1,2-Dichloro-ethane is rinsed away again with isopropanol. Under lucky circumstances a coverage with SWCNTs is obtained which is neither too diluted (difficult to find them) nor too dense (impossible to contact just one). The tubes are then studied by an AFM and selected nanotubes are located relative to alignment marks, which were made prior to the deposition of the SWCNTs. The e-beam exposure of PMMA resist and the final Au deposition take advantage of the position of the nanotubes relative to the alignment marks. Another method is to place contacts by random with the hope for success. In order to have some control over where the nanotubes appear on the surface of the substrate a CVD method has grown very popular over the past few years (see Ref. [7] for details). In this process e.g. methane disintegrates in a catalytic reaction at 900°C. The islands for the catalytic material are produced by e-beam lithography using PMMA resist. The catalyst is Fe(NO$_3$)$_3$ (with Mo) suspended on alumina particles. The carbon nanotubes seem to grow from the (reduced) Fe nanoparticles. The subsequent Au film is defined by electron beam lift off technique leaving SWCNTs reaching across the e.g. 300 nm gap between two Au electrodes. For both fabrication methods some nanotubes are inspected by a transmission electron microscope to check that the tubes are single walled. The contact resistances come out quite large. There is a tendency of lower resistances by using pure Au, though it sticks poorly to the SiO$_2$ surface. Often the contacts are in the MΩ class or higher. The 1-dimensional nature of the metallic SWCNT precludes any tube-resistances smaller than \(h/4e^2 = 6.5\, \text{kΩ}\). [9]
based on a constant interaction model and a paired electron occupation. An unpaired electron is responsible for the transmission between source and drain of the SWCNT. The parts of diamonds to the left and right in the plot correspond to even and odd. An external magnetic field of 6 T causes a visible Zeeman splitting as seen in Fig. 2 (bottom) for the states where the transport goes via an unpaired electron. As an example let us start from a finite bias, $V = 5 \text{ mV}$, in the middle diamond and move left in Fig. 2 (bottom). Crossing the Zeeman split differential conductance maximum corresponds to opening a channel for an electron through the SWCNT. We are adding an electron from left (say) and extracting it to the right in the voltage bias window. The number of electrons, which are stuck in the SWCNT in the middle diamond must be even in order for this charging/discharging of the tube to exhibit the shown Zeeman splitting. The Zeeman splitting corresponds to a $g$-factor of 2 as expected for graphite and for the almost free electrons in a carbon nanotube.

3. Odd-even alternation. Bimodal distribution

The Pauli principle forbids two electrons in a metal to enter into the same eigenenergy state. For quantum dots with few electrons Hund’s rules dominate, but for many electrons the exchange energy to all the rest of the electrons becomes too large to allow higher spin states unless the sample is very irregular or undergoes a magnetic ordering. Thus the picture, as given in any elementary solid state textbook on the free electron model, emerges, where each level is degenerately filled with two paired electrons. Surprisingly, this has never been directly observed for individual electrons in semiconductor quantum dots with many electrons or in metal islands. It is a sign that the exchange interaction in these other cases depends on the number of electrons because the wavefunction of each individual state, is irregular and dependent on the geometry. Density functional theory predicts in fact that even spin states higher than $\frac{1}{2}$ may occur in quantum dots [10]. In SWCNT, the first experiments [11] indicated that higher than $\frac{1}{2}$ spin states existed. These observations were probably related to inhomogeneity in the tubes. In regular SWCNT with few impurities a pairing effect is generally seen. Figure 3 (bottom panel) shows a single electron tunnelling sequence of a SWCNT, which is carefully selected for its regularity. Other similar sequences have been

Fig. 2. A grey-scale plot of $dI/dV$ vs. $V_g$ and $V$, where $V$ is the bias voltage across the SWCNT. Darker grey scale corresponds to higher conductance. $T = 100 \text{ mK}$. Upper panel (a) is taken at $B = 0$ and the lower panel (b) is taken at $B = 6 \text{ T}$. Zeeman splitting, $g\mu_B H$, of some states (dark grey lines) is seen in (b). The low conductance diamond in the middle region of (a) and (b) is the result of Coulomb blockade due to an even number of electrons on the SWCNT. The parts of diamonds to the left and right in the plot correspond to odd number of electrons. For a finite bias, $V$, between source and drain of the SWCNT a window for transmission opens for the conductance. In this region several eigenenergies of the SWCNT may contribute to the conductance; this is seen as a number of skew grey lines in the $V-V_g$ plane. The level spectrum of the SWCNT can therefore be directly read off from the grey scale diagram. In strong magnetic field (b) some of the skew lines are Zeeman split; these lines correspond to peaks in transmission where an unpaired electron is responsible for the transmission between source and drain. The interpretation of such grey-scale plots for SWCNT can be based on a constant interaction model and a paired electron occupation.

boundary conditions where the tube enters under the Au film. From a bias spectroscopy diagram as the one shown in Fig. 2 (bottom), it is possible to deduce whether $n$ is even or odd. An external magnetic field of 6 T causes a visible
reported [12, 13]. In Fig. 3 the conductance vs. backgate voltage is taken at very small dc bias voltage (0.5 mV) and at a temperature of 4.2 K. Both the height and the addition energy of the Coulomb blockade peaks come in an odd-even alternation in the gate voltage range $-0.1$ V to $+0.1$ V. This is clearly displayed in the top panel, where the addition energies are plotted. In the gate voltage regime $-1.0$ V to $-0.3$ V, the addition energy is constant. This is then representing the bare Coulomb energy $e^2/2C_T$. This behaviour is essentially what is observed in large 2D or metallic quantum dots, where the variation in the eigenenergy is insignificant compared to the Coulomb blockade energy. For the SWCNT shown in Fig. 3 in the gatevoltage regime $-0.3$ V to $+0.1$ V there is a strong and rather regular alternation in the additional energy. The strong odd-even effect is most likely related to the electron spin and one possibility is that a magnetic impurity talks to the electrons at the Fermi energy in the region between $-0.3$ to $0.1$ V, whereas it is not overlapping with the electronic states at the Fermi surface in the region $-1.0$ V to $-0.3$ V. The sequence we see is likely to reflect the odd even alternation of electrons on the SWCNT. The alternation between $S=0$ and $S=1/2$ we observe may be due to an exchange interaction with a magnetic ion sitting on the SWCNT and therefore a strongly amplified odd-even effect compared to what one might see for a regular SWCNT [12,13].

The level statistics for semiconductor quantum dots and metal particles has been much studied in later years. The expected odd-even bimodal distribution has been suspiciously absent. Recent studies [14], however, have demonstrated odd-even structure in the transmission through small lateral semiconductor Aharonov-Bohm rings exhibiting Coulomb blockade effects. The bimodal distribution we observe, clearly brings the SWCNT into a role of a particularly regular quantum dot system, which might also be expected in very symmetric quantum dots. The fact that the single electron tunnelling exhibits clear odd-even effect for a metallic SWCNT with about 20000 electrons is gratifying and shows that the Pauli principle is sufficient in 1 dimension as expected. However there are other indications that a SWCNT, which is isolated from source and drain, has properties in accordance with a Luttinger liquid [15]. Thus the spin degeneracy is observed, but the semiclassical model does not yield the full story, as we shall now discuss.

4. Tunnel contacts and the Luttinger liquid

One of the first and most immediate surprises in the electrical resistance of metallic SWCNT (and the two contacts) is that the resistance increases as the temperature is lowered as we have already demonstrated in Figure 1. At the lowest temperatures, where $k_B T$ becomes comparable to the Coulomb blockade energy, single electron tunnelling appears. What is not so immediately apparent is a nonlinearity at voltages higher than $k_B T/e$ and lower than the thermal run-away voltages (100 mV) [15,16]. These power-law dependences have particular exponents, which are in good agreement with theoretical prediction for the so-called Luttinger liquid. Theory predicts that $G T^{-2}$ can be expressed in the form $G T^{-2} = f(eV/k_B T)^2$, where $f$ is a characteristic function, $G$ is the conductance through the tunnel barrier between the Au contact and the SWCNT. The deduced exponent $z$ is in good agreement with experiments. This was the first demonstration of the existence of the Luttinger liquid and it means a revival of Tomanoga’s and Luttinger’s theories from 1950 and 1963 (see Ref. [17] for a review). This gives quite strong conclusions about the breakdown of the Fermi liquid model and the semiclassical free electron model for strictly 1-dimensional systems isolated from any influencing contacts or external disturbances. The powerlaw now found for the conductance of the contacts to SWCNT is in very convincing agreement with the theory of Luttinger and Tomonaga. The really surprising fact is that the Luttinger liquid involves changes in the quasiparticle energies, which are not even swamped by room temperature thermal energies. Another surprising observation is, that it can be observed also in multi wall carbon nanotubes, in spite of the much weaker 1-dimensionality there [18]. The observation of the Luttinger liquid effect confirm theories [17], however we would still like to see other consequences of this new liquid and understand why, for instance, the odd-even behaviour, described above, is not affected. No semiclassical and intuitive picture of the Luttinger liquid exists. The small screening of spin and charge presumably gives rise to spinwave and plasma excitations without an energy gap in their dispersion. This may be a precursor to a magnetic, superconductivity or Wigner lattice transition at low temperatures. There are in fact indications that magnetic order or superconducting proximity in SWCNT exist at very low temperatures. In the following section we describe some surprising results on spin injection into SWCNT.

5. Ferromagnetic contacts

The facts that the SWCNTs are well described in terms of eigenstates determined by the full length (500 nm) between contacts, and that odd-even structure is occasionally observed indicate that a 500 nm SWCNT acts as a coherent interface between our source and drain, and shows that the Pauli principle is sufficient in 1 dimension as expected. However there are other indications that a SWCNT, which is isolated from source and drain, has properties in accordance with a Luttinger liquid [15]. Thus the spin degeneracy is observed, but the semiclassical model does not yield the full story, as we shall now discuss.

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hysteretic magnetoresistance has been observed below about 30 K leading to quite dramatic effects at the lowest temperature (300 mK). Figure 4 shows biasspectroscopy plots at a weak magnetic field \( B = 0.08 \) T and at strong magnetic field \( B = 2 \) T. The weak field is chosen so that, the conductance is minimum. In fact, the conductance is too small to be measured at small bias values and a finite conductance can only be reliably measured at a few mV bias. The bias spectroscopy plot yields a Coulomb blockade diamond corresponding to a Coulomb energy around 5 meV. At the strong field the conductance is much larger and in Fig. 4 (right panel) the Coulomb blockade diamond is resolved, though with strong asymmetry corresponding to unequal contact resistances at source and drain. A dramatic influence of the magnetic field is displayed in Fig. 5 where the conductance through the SWCNT is shown when sweeping the magnetic field. A hysteresis is observed for this sample as well as others. The detailed variation is however different from sample to sample. A number of features should be noticed: (a) There is an asymmetry of the magnetoresistance around zero field. For increasing magnetic field from \(-1\) T to 0 T the conductance gradually decreases. The minimum in conductance is at 0.08 T. When the field increases further the conductance goes through a sharp maximum \((G = 0.3 \mu \text{S})\) around 0.5 T and another sharp but smaller maximum at 0.8 T after which the conductance saturates at a value of about 0.2 \(\mu\)S. These features reflect the properties of the contact with the highest conductance, which can be located by knowing the direction of the asymmetry of the biasspectroscopy plot. When the sweep direction of the magnetic field is reversed the conductance curve is mirror reflected around \(B = 0\). The high and the low conductance peaks are now moved to the corresponding negative field values. These features are clearly related to the change in magnetisation of the Fe domains in the contacts of the SWCNT. (b) The large variation in the observed magnetoresistance is unexpected and far exceeds what is thought to be possible (32% for Fe) [22]. In order to explain this, we may consider an exchange influence on the SWCNT, which, however, is small because of the high contact resistance. From the results displayed in Fig. 4, we know that the largest resistance must be close to \(5 \text{ M}\Omega\). The other contact resistance can be considerably lower. The exchange effect may then be large because we are influencing the tube with a ferromagnet with perhaps \(10^{10}\) polarized electrons. Since the coherent Kondo properties (see below) of the SWCNT have dramatic effects, a very small induced magnetisation of the SWCNT may also have quite dramatic effect on the transmission through the tube.

6. Coherent tunnelling through the Coulomb blockade – The Kondo effect

As the contact conductance increases, the Coulomb blockade is smeared and the electron number in the SWCNT becomes a more and more ill-defined quantity. The determining parameter is the lifetime of a given charge number state given by the \( RC \) time constant, \( \Gamma^{-1} \), of the tube, where \( R \) is the parallel combination of the source and drain contact resistances. If \( h\Gamma \gg e^2/2C \) the Coulomb blockade and the distinction between the integral electron charge states becomes irrelevant. For \( h\Gamma \sim e^2/2C \) there may still be a rough charge counting at low temperatures, yet a significant overlap between the electronic system in the Au contacts and the tube. The strong overlap of the electronic states in the contacts and in the SWCNT has the consequence that we cannot solve the many-body electron problems of the contacts separated from the tube. While this in general is a mean field problem, in the mentioned situation we may have a odd number of electrons in the SWCNT and therefore a total electronic spin of \( \frac{1}{2} \). Exchange interactions are very powerful and as soon as we introduce unpaired spins, large energies are in play, which directly couple magnetic screening Kondo states in the source and drain coherently, as was first observed in semiconductor quantum dots [23]. An uncompensated spin in a SWCNT will be screened by the Au contacts. This can only occur through spin rearrangement at the Fermi surface and an extra exchange energy is added to the potential energy below the Fermi energy and subtracted above, concentrating many \( k \)-states close to the chemical potential. So, as a consequence of the strong exchange coupling a maximum in the density of
states at the fermi energy gives rise to a coherent state together with the unpaired electron in the SWCNT. If we were to measure the resistivity in a small region of the Au contact within a region given by the Kondo length, $h/\sqrt{mk_B T_K}$, close to the contact point, we would presumably measure an increased resistance because the dispersion relation at the chemical potential becomes flat and the group velocity becomes close to zero causing severe scattering. This constitutes the well-known Kondo effect in metals (here Au) apart from the fact that in conventionally observed Kondo resistance anomaly a magnetic impurity atom causes the extra scattering, not as here an $S=\frac{1}{2}$ quantum dot. The first observation of a Kondo effect in carbon nanotubes was with a magnetic impurity present [24,25]. In our experiment [26] we studied the transmission of electrons between source and drain, and it is the spin $\frac{1}{2}$ in the SWCNT, which acts as a magnetic impurity. The exchange coupling between the tube and the drain and source contact, couples source and drain coherently together and we observe a one-channel conductance through the $S=\frac{1}{2}$ eigenstate of the SWCNT. This is a perfectly adiabatic conduction channel between the many electron coherent states in the Au source and drain. The conductance is therefore in the ideal case at zero temperature simply $4e^2/h$ similar to a quantum point contact. We expect $2 \times 2e^2/h$ due to the two uncoupled, but crossing 1-dimensional bands in the metallic SWCNT. We never reach this value, though. The described transmission between source and drain is principally different from the conventional Kondo effect. It is a second order process, where at the same time a spin up (down) electron enters the SWCNT spin $\frac{1}{2}$ state from the source, as a spin down (up) electron leaves the SWCNT for the drain [27,28]. One might expect this transmission to be of a much lower probability than the transmission when the SWCNT spin $\frac{1}{2}$ state is aligned with the chemical potential in source and drain. The coherence all the way from the source through the tube to the drain enhances this transmission in much the same way as Cooper pairs in superconductors enter through a Josephson barrier. Whether this analogy can be taken even further and lead to a Josephson-like oscillation of the current through the spin $\frac{1}{2}$ SWCNT state with a frequency $eV/h$, remains to be seen.

We have measured the conductance through a SWCNT as a function of the voltage on a backgate. Due to the rather small contact resistance (15 kΩ), the Coulomb blockade is not well developed and rather irregular, even here at 100 mK. Much of the irregularities are related to imperfections and impurities along the SWCNT, but we do find rather regular sequences as we have also seen in the samples with high resistance contacts. One noticeable fact is that the conductance in its peak values only in a few perfect and symmetric cases [29] reach the theoretical limit $4e^2/h$. One reason for this could be a difference between the contact resistances, which is revealed in the bias spectroscopy plots; another reason could be a possible lifting of degeneracy of the two 1-dimensional bands in the metallic carbon nanotubes. Figure 6 shows bias spectroscopy grey scale plots of a small part of the gate voltage axis for a SWCNT with intermediate contact resistances to source and drain. The temperature is 100 mK and the 4 plots are taken for different magnetic fields. The grey colour indicates the value of the conductance. The darkest region corresponds to a conductance of about $2e^2/h$. Reminiscent Coulomb blockade diamonds are seen, but they are more irregular and less well-defined than for the small contact conductances device in Fig. 2. This is as expected for the intermediate contact conductance, because the reflections along the SWCNT become more dominating relative to the end boundary. Essentially two types of equally abundant Coulomb diamonds are seen in the top panel in Fig. 6: One type has (like in Fig. 2) a diamond with a very small conductance in the interior. The diamond is truncated at high and low source-drain voltages giving the blockade region a hexagonal shape. The other type of diamond has a high conductance line running across at zero bias voltage (a dark ridge). We interpret these two blockade regions as representing an even and an odd number of electrons, respectively. The dark ridge

![Fig. 6. Grey-scale plots of dI/dV vs. Vg and V at T = 100 mK and for 4 magnetic field strengths (B = 0, 1, 2, 3 T) showing Kondo resonances. The light (low conductance) areas around $V = 0$ correspond to reminiscences of Coulomb blockades. For $B = 0$ T these areas are cut by a characteristic ridge at $V = 0$ V (for instance in a region around $V_g = 0.26$ V). This is a coherent transmission between Kondo states imposed on both the source and drain Au contacts due to exchange set up by the spin $\frac{1}{2}$ state in the odd electron SWCNT. The even electron Coulomb blockade has paired spins and gives no exchange coupling. In finite magnetic field the paired spin ($S = 0$) singlet state and the next excited triplet state can be forced into degeneration. This happens for $B = 3$ T, where a ridge at $V = 0$ is now formed in the gate voltage region around $V_g = 0.05$ V and $V_g = 0.2$ V, with a new Kondo state with even number of electrons. The half-width of the ridges give the Kondo temperature, which may also be extracted from the temperature dependence (not shown).]
in the odd electron region is a result of the Kondo state in each Au electrode at each end of the SWCNT and represents the coherent short-circuit of the otherwise blockaded region. The half-width of the ridge corresponds to the Kondo energy or temperature (here about 0.5 K). At temperatures below $T_K$ the height of the conductance ridge saturates. As the Kondo temperature goes down, thermal smearing takes over. The ridge becomes thermally smeared, when $k_B T > k_B T_K$.

The open truncated blockade region corresponds to an even number of electrons in the SWCNT and we have $S = 0$ and thus no generation of a Kondo state in the Au contacts. However as the magnetic field is increased the even electron singlet is approached by one of the higher energy triplet states and at around $3 \, \text{T}$ the two become degenerate and we have $S = 1$, 0 degeneracy leading again to a Kondo effect in the source and drain caused by the Kondo screening of the contacts. In this situation two electrons may tunnel via the forbidden state in the SWCNT from source to drain coherently. This leads again to a ridge in the grey scale conductance spectroscopy plot at zero bias, cutting through the Coulomb blockade diamonds (or hexagon) as clearly seen at $B = 3 \, \text{T}$ for the even electron gate voltage regions. This singlet-triplet Kondo effect has similar properties to the odd electron effect. The height of the conductance ridge (Fig. 6, lower panel) again saturates at temperatures lower than $T_K$. The value is in this case far from $4 e^2/h$. This may be related to the decoherence effect of the magnetic field used to generate the effect. The even electron Kondo state should be observable for a singlet ground state of an impurity atom in a paramagnetic matrix like Cu and in a high magnetic field, but such a Kondo system has so far not been studied. The singlet-triplet Kondo system was first observed in SWCNTs [26] and predicted for semiconductor quantum dots [27].

7. What is to come?

The recent reports about magnetism [30] and superconductivity [31] in fullerene crystals at $600 \, \text{K}$ and $117 \, \text{K}$, respectively, give expectations for anomalous behaviour in carbon nanotubes as well. It has in fact been reported that SWCNT can be superconducting (intrinsic or by proximity) at the lowest temperatures [32–34]. Our results on spin injection (Figs. 4 and 5) indicate some unknown magnetic properties of the tubes. One of the main obstacles for studying electrical transport in SWCNTs is the difficulty of getting low contacts resistances. The Au contacts seem presently a way to obtain the small resistance. Fe, Ni, Co which we have used for magnetic contacts, accidentally give good contact to the SWCNT. There are a number of alternative experiments on the magnetic coupling of a ferromagnet to a SWCNT, which might be interesting. Thus it might be worthwhile to investigate the influence of one or two extra ferromagnetic contacts on the SWCNT, while keeping the source and drain contact nonmagnetic (Au). To what extent will exchange interactions force a magnetic order into the SWCNT? Will this magnetic proximity be ferro or antiferromagnetic? Is the Fe-SWCNT-Fe a giant magneto-resistance system as Fig. 5 indicates?

It would also be interesting to pursue the Kondo experiments by not just measuring the transmission effect through the SWCNT, but also measure the influence the spin state of the SWCNT has on the electrical properties of the Au contact. The Au contact could be a narrow metal strip (300 nm) which would have an induced Kondo state right across its width. The Kondo effect would be like a local spin glass in a middle region of the Au strip and the Kondo resistance measured along the Au strip may for odd number of electrons on the SWCNT have an anomalously high value [35]. Another interesting route of investigations could be to test the coherent properties of the Kondo states. This could be achieved by measuring a single electron ac Josephson-like effect between the two Kondo states at each Au contacts. The triplet-singlet Kondo effect created by a magnetic field transmits pairs of electrons from source to drain and might have a voltage-frequency relation $v = 2eV/h$ as for Cooper pairs in superconductors.

The spin properties of SWCNT is a very interesting field, both on its own right, but also as a tool to give rise to a controllable single electron spin ($\frac{1}{2}$) exchange effect on the surrounding electrodes. It is a challenge to develop the growth techniques to be able to grow SWCNT with a particular chirality. By Raman spectroscopy [36] it is already possible to determine this parameter, and one particular chirality has been suggested to give rise to a characteristic linear magnetoresistance in an axial magnetic field [37]. An ac drag of single electrons in a SWCNT, using surface acoustic waves, has recently been suggested and would in combination with injection of spin polarised electrons be an interesting target for quantum information processing [38].

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