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Single Electron Spintronics

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Single electron electronics is now well developed, and allows the manipulation of electrons one-by-one as they tunnel on and off a nanoscale conducting island. In the past decade or so there have been concerted efforts in several laboratories to construct single electron devices incorporating ferromagnetic components in order to introduce spin functionality. The use of ferromagnetic electrodes with a non-magnetic island can lead to spin-accumulation on the island. On the other hand, making the dot also ferromagnetic leads introduces new physics such as tunnelling magnetoresistance enhancement in the cotunnelling regime and manifestations of the Kondo effect. Such nanoscale islands are also found to have long spin lifetimes. Conventional spintronics makes use of the average spin-polarisation of a large ensemble of electrons: this new approach offers the prospect of accessing the quantum properties of the electron, and is a candidate approach to the construction of solid-state spin-based qubits.

Keywords: Spintronics; nanomagnetism; nanoelectronics

1. Introduction

In this review we will describe recent progress in bringing together two subfields of nanoelectronics: spintronics [1, 2] and single electron electronics [3]. There is by now a well-established science base for both [4], but commercialisation of technologies built on them is at a very early stage, making the conjunction of the two especially timely. The outcome will be novel device designs that will offer new opportunities in both post-CMOS nanoelectronics and potentially open the door to solid-state quantum information technologies based on spin (see the review by Ardavan & Briggs [5] in this issue).

Indeed, the post-CMOS age is almost upon us: it is expected in the International Technology Roadmap for Semiconductors (http://www.itrs.net) that devices will soon shrink to the level where, at current doping levels, there will be an average of only one dopant within the channel of a transistor. However, the actual number of dopants in a device will be randomly distributed with Poisson statistics, and such randomness in device performance is unacceptable within the conventional CMOS technology paradigm. It is therefore better to work with devices that are designed to deal with individual carriers from the outset. Single electron electronics is based around nanoscale elements so small that addition of a single carrier increases the charging energy by an amount larger than $k_{\mathrm{B}}T$, preventing further carriers passing simultaneously [3]. Coulomb blockade (CB)-based devices have been realised in a

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number of different semiconductor [6], metallic [7, 8], and organic materials systems [9, 10]. Wafer-scale parallel fabrication of devices showing room temperature single electron effects has recently been demonstrated by Ray *et al.* [11].

Meanwhile, spintronics hit the headlines in 2007 when the Nobel Prize for physics was awarded to Fert and Grünberg. Their independent discoveries of giant magnetoresistance (GMR) [12, 13] represented the birth of the field of spintronics, in which the generation, manipulation, and detection of spin-polarised currents are used to store and process information [1, 2]. Evidence for the scientific impact is found in the high number of research articles in leading journals (the 1988 Physical Review Letter of Baibich *et al.* reporting the discovery of giant magnetoresistance (GMR) is the sixth most highly cited of all time), whilst the massive investment of information technology companies in spintronics-based research is based on the huge revenue (billions of $ p.a.) that they are already reaping from early devices exploiting the basic GMR spintronic effect such as hard disc spin-valve heads, and the revenue that is expected from future technological breakthroughs. The most basic spintronic devices, spin-valves, have found commercial application in hard disk playback heads and magnetic random access memories. To date, a logic gate operating on these principles has eluded researchers since the on/off ratios of current spin-valves are too low: the use of spin blockade physics in single electron devices offers significantly higher values. Moreover, these currently commercialised devices operate using the ensemble-averaged spin of a current of electrons or holes. Hence the quantum properties of the spin are only indirectly accessed. In order to move towards quantum information technologies, spintronics at the single spin level is required.

This review concerns the efforts to unify these two vibrant research areas in nanoelectronics. Using these systems it is possible to study a wide range of novel aspects to the problem, including Kondo physics, Coulomb and Pauli spin blockade, tunnel magnetoresistance enhancement, spin accumulation and non-equilibrium magnetisation, CB anisotropic magnetoresistance in dots with strong spin-orbit effects, and enhanced spin-lifetimes.

2. Single Electron Electronics

Coulomb interaction effects are always present in electrical devices, but it is only recent advances in nanofabrication techniques that have allowed devices to become small enough that the Coulomb effects can dominate. Small electrically isolated conducting islands give rise to low capacitances, $\sim 10^{-18}$ F, which consequently give large charging energies $E_{\mathrm{C}} = e^2/2C$, where C is the capacitance of the nanoparticle. This classical electrostatic energy associated with the addition or removal of electrons from the island can cause suppression of transport through the island, this is known as the CB. The CB can dominate transport if $E_{\mathrm{C}} \gg (k_{\mathrm{B}}T, |eV|)$, i.e. for low temperature T and low bias voltage V. In that case a single tunnel event to the metallic island can increase the electrostatic energy of the system sufficiently that the bias voltage is not great enough to allow another electron to enter the dot from the source until the original one has departed for the drain [14]. These electron tunnelling events (known as sequential tunnelling) are thus forced to be discrete. Upon increasing the bias it is possible to inject further electrons each time eV exceeds an integer multiple of E_{C}, causing steps in the current voltage-curves $I(V)$ as

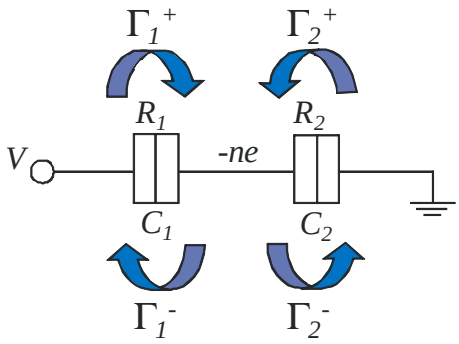

Figure 1. Schematic of the generic device layout for orthodox theory of Coulomb blockade transport. The system comprises two tunnel junctions, labelled $i = 1, 2$, with resistance R_i and capacitance C_i. The particle flow directions Γ are shown with curved arrows. After Maekawa *et al.* [19].

more electrons can simultaneously traverse the island due to the increased applied bias, known as the Coulomb staircase. An excellent pedagogical introduction to the relevant physics is given by Nazarov & Blanter [15].

These systems are often discussed using the orthodox theory, of which we review the main results here. This is a simplified Fock space treatment where all the excitations are integrated incoherently. The orthodox theory was initially described by Likharev [3, 16], and Averin & Nazarov [17]. Following this work Hanna & Tinkham [18] derived the theory in an intuitive manner to model $I(V)$ curves incorporating the resistance, R, capacitance C, temperature T, number of electrons in the system n, and the tunnelling rates onto (Γ^+) and off (Γ^-) the central island. A schematic of the system is shown in figure 1. When a gate electrode is added, the device is known as a single electron transistor (SET).

From the golden rule calculation, the tunnelling rate for the ith junction $\Gamma_i^{\pm}(n)$ can be written as

$$\Gamma_i^{\pm}(n) = \frac{1}{R_i e^2}\left(\frac{-\Delta E_i^{\pm}}{1-\exp(\Delta E_i^{\pm}/k_{\mathrm{B}}T)}\right), \tag{2.1}$$

where ΔE is the energy change as the electron tunnels across the system. From electrostatic considerations we can then write

$$\Delta E_1^{\pm} = \frac{e}{C_\Sigma}\left(\frac{e}{2} \pm (ne - Q_0) \pm C_2 V\right), \tag{2.2}$$

and

$$\Delta E_2^{\pm} = \frac{e}{C_\Sigma}\left(\frac{e}{2} \mp (ne - Q_0) \pm C_1 V\right), \tag{2.3}$$

where $C_\Sigma = (C_1 + C_2)$ and Q_0 is the background charge of the island. The current through the island is then calculated using the master equation approach,

$$I(V) = e\sum_{n=-\infty}^{\infty} p(n)[\Gamma_2^+(n) - \Gamma_2^-(n)] = e\sum_{n=-\infty}^{\infty} p(n)[\Gamma_1^-(n) - \Gamma_2^+(n)], \tag{2.4}$$

where $p(n)$ is the probability of finding n excess electrons on the island.

Despite the approximations made in the model, a good fit to experimental data is often obtained [see e.g. 20–22]. The assumptions made are: the electron energy quantisation is ignored, the electron tunnelling time is assumed to be negligible in comparison with other time scales, and cotunnelling (correlated transfer of more than one electron) events are ignored [3].

3. Spin Electronics

The idea of spin-polarised conduction in ferromagnets can be traced back to Mott & Jones [23] and a summary of the state of the art in homogeneous materials was given by Fert & Campbell [24]. In the Stoner [25] picture of itinerant electron ferromagnetism that describes the transition metal ferromagnets Fe, Co, and Ni, the $3d$ bands are spin-split by the exchange energy ($\sim$ 1-2 eV), giving rise to markedly different band structures for spin-$\uparrow$ and spin-$\downarrow$ electrons at the Fermi level. This difference in properties such as the density of states and Fermi velocity leads to a difference in conductivity for the two spin species. Mott's insight was that these two spin populations can be treated as two separate conduction channels in parallel. (This relies on the spin-flip lifetime $\tau_{\mathrm{sf}} \gg$ any other relevant timescale, which is the case in most, but not all, common situations.) Hence, when an electric field drives a current through the metal, it will be carried predominantly by electrons of the most conducting spin channel, which form a shunt, partially short-circuiting the other spins. Spintronics is essentially the use of these spin-polarised currents that can be generated and detected by ferromagnets in heterostructured devices.

This knowledge of the spin-polarised conduction in ferromagnets was exploited by Baibich *et al.* [12] and Binasch *et al.* [13] in their study of Fe/Cr multilayer heterostructures grown by molecular beam epitaxy and the discovery of the GMR. The basic unit of a GMR device is the so-called spin-valve, first reported by Dieny *et al.* [26], and consists of a pair of ferromagnetic layers separated by a non-magnetic metal layer. Transport takes place in the diffusive regime. The layers are capable of being switched into states where the two magnetic moments are parallel or antiparallel. When the moments are parallel the spin labels in the two layers match, and the shunt channel, with weak scattering, is preserved: all of the strong scattering is in the other channel. When the moments are antiparallel, the spin labels do not match, and there is significant scattering of carriers in both spin channels, leading to a higher overall resistance. Since this switching can be accomplished with an applied field, this resistance change is the GMR. The fractional change in resistance, $\Delta R/R$, is $\sim$ 10 % in spin-valve trilayers, and can be $\sim$ 100 % in multilayer superlattices.

Related physics obtains when one considers a tunnel junction between two itinerant ferromagnets. Meservey & Tedrow [27] give a review of their pioneering contributions to the measurement of tunnelling spin-polarisation of ferromagnetic metals using superconductor/ferromagnet junctions. The first attempt to make a ferromagnet/ferromagnet tunnel junction was made by Julliere [28], who found magnetoresistance only at low temperatures, again found when switching the two magnetic electrodes in the junction from an antiparallel to a parallel state. He also derived the now famous Julliere formula

$$\frac{\Delta R}{R} = \frac{2P^2}{1-P^2}, \tag{3.1}$$

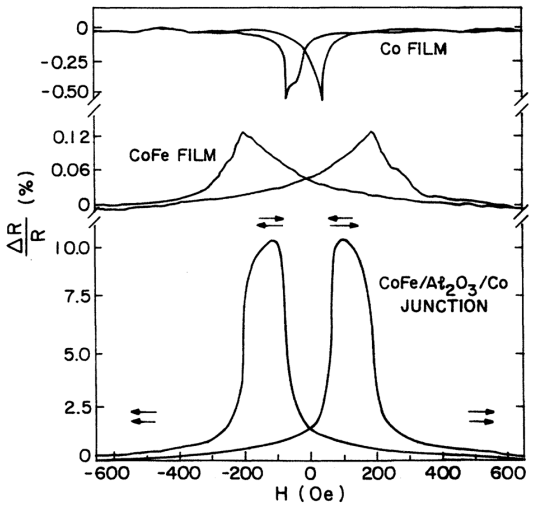

Figure 2. Resistance of a $CoFe/Al_2O_3/Co$ junction plotted as a function of magnetic field H in the film plane, measured at 295 K. Also shown is the variation in CoFe and Co film resistance. The arrows indicate the relative directions of the magnetic moments in the two layers. After Moodera *et al.* [30].

which relates the tunnelling magnetoresistance (TMR) to the tunnelling spin polarisation of the electrodes, P.

A significant breakthrough was made by Miyazaki & Tezuka [29] and Moodera *et al.* [30] (see figure 2), who found sizeable room temperature effects in the 1990s, using optimised barriers of post-growth oxidised aluminium, which yields a disordered alumina layer. This lead to an explosion of activity in laboratories across world on such junctions, reviewed by Tsymbal *et al.* [31]. The initial values were ~ 10 %, but 80 % at room temperature has recently been achieved by Wei *et al.* [32] using CoFeB electrodes.

Just after that review was published, predictions based on *ab initio* band structure calculations of huge TMR in epitaxial Fe/MgO/Fe junctions by Mathon & Umerski [33] and Butler *et al.* [34] were experimentally confirmed. Yuasa *et al.* [35] grew their junctions by molecular beam epitaxy, whilst Parkin *et al.* [36] used magnetron sputtering. Both groups obtained room temperature TMR ratios ~ 200 %, and values exceeding 600 % have recently been achieved [37]. Much of the work on CB spintronics is based on the introduction of conducting islands into the centre of the barriers of such tunnel junction devices, either with alumina or MgO barriers.

Another spintronic effect that plays a role in the experiments that we shall review is spin accumulation, which occurs when a current flows between a ferromagnet and an unpolarised material. The phenomenon was originally demonstrated by Johnson & Silsbee [38] and described theoretically by van Son *et al.* [39] as a non-equilibrium splitting of the spin-dependent chemical potentials in the unpolarised material. The essentials of the effect are that the interface has different transparency to electrons of different spin due to the same type of spin-dependent scattering processes that give rise to spin-polarised currents in the bulk of the ferromagnet. Suppose that the current is being driven into the normal metal from

the ferromagnet. One spin will pass more easily through the interface and so an out-of-equilibrium spin density of that species will build up there. The other species cannot pass through so easily and will build up density on the ferromagnetic side of the interface. The opposite occurs for the current flow in the opposite direction. The net result is that a weak magnetisation, proportional to the current density, is induced in the normal metal, compensated for by a small demagnetisation of the ferromagnet.

Both effects are localised close to the interface on a lengthscale called the spin diffusion length, ℓ_{sf}. This can be hundreds of nm in a normal metal like Cu, but only perhaps ten nm in a ferromagnet like Co. Spin accumulation forms the basis for the semiclassical understanding of GMR in the vertical current-perpendicular-to-the-plane geometry [40], is important for non-linear GMR effects [41] and spin-transfer torque effects in nanopillars [42], and plays a key role in spin-injection into semiconductors [43].

4. Spintronics in the Coulomb blockade limit

(*a*) *Theoretical beginnings*

There is a great deal of published theory concerning single-electron devices embedded in magnetic tunnel junctions [44]. One of the most important predictions is that an oscillatory dependence of TMR on junction voltage bias should be observed in the CB regime. This oscillation can be used to characterise the spin lifetime. This topic has been studied for some years by Barnaś and co-workers. In collaboration with Fert he made the first prediction of an oscillatory TMR in a landmark paper in PRL [45]. The issue of a long spin relaxation time, leading to spin accumulation on the island, was addressed by the same authors [46]. This spin accumulation is important since it can generate new effects, including negative differential resistance, inverse TMR, and the presence of TMR for islands of non-magnetic material. More recently the relative contributions of sequential and co-tunnelling have been calculated for bias voltages below the CB threshold [47], and the effects of gate voltages have also been treated [48]. Takahashi & Maekawa [49] independently predicted the oscillatory TMR in these structures, and also treated gating in the same early paper. Brataas *et al.* [50] showed how measurements of the TMR in the CB regime provide unambiguous evidence for spin accumulation, as well as showing how high speed measurements could give direct access to the spin lifetime. This spin accumulation is crucial to this whole enterprise, since it is vital that spin information is not lost within the island.

(*b*) *Early experimental progress*

Although early experimental efforts to study the interplay of CB and spin-polarised transport were made as long ago as the 1970s by Gittleman *et al.* [51] and Helman & Abeles [52] in cermet films, experimental progress has generally lagged behind theory. Nevertheless, some recent experiments have made remarkable progress in starting to demonstrate the range of fascinating phenomena predicted by the theoretical models. Schelp *et al.* [53] measured Co / Al_2O_3 / Co junctions with Co nanoclusters embedded in the barrier: a low temperature I-V characteristic

that could not be fitted with the conventional Simmons [54] theory was cited as evidence for CB effects, but single electron steps were not observed.

The Ralph group at Cornell have studied tunnelling through particle-in-a-box states using metallic [55] and magnetic [56] nanoclusters for a number of years using self-assembled clusters addressed by tunnelling through a nanofabricated pinhole in a Si_3N_4 membrane. This method has been used to examine the magnetic anisotropy of different quantum levels [57], filter spins for polarisation measurements [58], and determine spin-orbit scattering [59] and g factor anisotropies in noble metal nanoparticles [60].

Oscillatory conductance and TMR with voltage bias was observed by Nakajima *et al.* [61] in similar junctions with $Co_{80}Pt_{20}$ nanoparticles within the barrier. Ernult *et al.* [62] also made an early observation of these oscillations in an epitaxial Fe/MgO/Fe MTJs. Coherent tunnelling through an Fe-MgO core-shell nanoparticle has recently been observed [63], but using non-magnetic electrodes.

(*c*) *Spin accumulation in non-magnetic islands*

Spin accumulation was introduced in part 3 and involves the injection of non-equilibrium spin density into a nonmagnetic material by passing a spin-polarised current into it from a ferromagnet. Whilst a mainstay of conventional spintronics devices, spin injection into zero dimensional objects has only been explored by a few groups. Nevertheless, the type of structure discussed here and shown schematically in figure 1 is almost ideal to see the basic mechanism behind the effect. Consider an island between ferromagnetic electrodes which have antiparallel moments, and let us suppose that the tunnel rate is spin-dependent and larger for majority spins. This will mean that the electrons entering the island from the source electrode predominantly have majority spin, as they may tunnel in more easily. However, it is harder for them to leave into the drain, whilst minority spins quickly do so. Majority spins will therefore accumulate in the dot, and equilibrium will only be restored when the bias voltage is switched off and the current drains away. Such devices were studied theoretically by Brataas *et al.* [50], Barnaś & Fert [64], and Imamura *et al.* [65].

Chen *et al.* [66] fabricated superconducting single electron transistors with an Al island connected to Co leads. When these were set into an antiparallel state, the superconducting gap of the Al was dramatically reduced, in proportion to the source-drain voltage, consistent with accumulated spins breaking Cooper pairs. Zaffalon & van Wees [67] also detected spin accumulation in an Al zero dimensional dot connected to four Co electrodes via tunnel contacts, fabricated using lateral angle-evaporation technology, but that structure was too large to show CB effects.

Bernand-Mantel *et al.* [68] studied a device with a single 2.5 nm Au island between Co electrodes. They observed clear CB peaks in the differential conductance below temperatures of about 60 K. TMR measurements were carried out at 4 K, and an easily measurable TMR signal observed in both the blockaded ($V = 20$ mV) and sequential ($V = 110$ mV) tunnelling regimes at 4 K, as shown in in figure 3, compelling evidence of the preservation of spin information accumulated on the Au island. It is noteworthy that in both cases the TMR was negative in sign, a possibility admitted by the theory of Martinek *et al.* [69]. A similar structure, but with MgO barriers, was studied by Mitani *et al.* [70]. They observed negligible TMR at

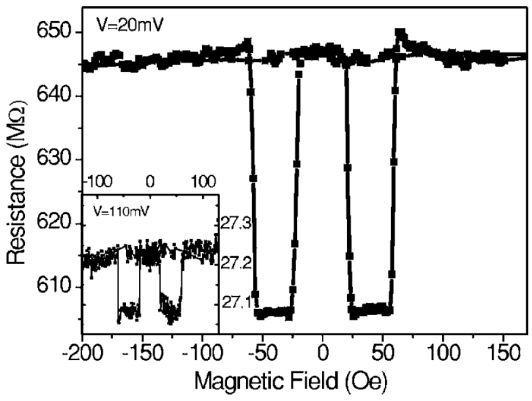

Figure 3. Resistance vs magnetic field obtained at $V = 20$ mV and 4 K for Au nanoisland contacted by Co leads. Inset: same sample at $V = 110$ mV. At both bias voltages the TMR is inverted. After Bernand-Mantel *et al.* [68].

low bias, but found that it appeared and was enhanced as V was increased, *viz.* as more spins were accumulated on the dot.

Birk *et al.* [71] studied the effects of non-collinearity between the accumulated spins in an Al dot and an external applied field, which lies at an angle α to the parallel or antiparallel magnetisations of the Permalloy leads. This means that the electron spinor $|\uparrow(\downarrow)\rangle'$ in the dot must be decomposed into the basis states in the ferromagnets $|\uparrow\rangle$ and $|\downarrow\rangle$ according to $|\uparrow(\downarrow)\rangle' = \cos(\alpha/2)|\uparrow(\downarrow)\rangle + \sin(\alpha/2)|\downarrow(\uparrow)\rangle$. One can then obtain the total tunnel rate between a ferromagnetic lead and the state $|\uparrow(\downarrow)\rangle'$ as $\Gamma_{|\uparrow(\downarrow)\rangle'} = \Gamma(1 \pm P)\cos^2(\alpha/2) + \Gamma(1 \mp P)\sin^2(\alpha/2) = \Gamma[1 \pm P\cos\alpha]$. The spin polarisation P of the electrode is hence transformed into an effective polarisation $P\cos\alpha$. It is to be noted that this is somewhat different physics to the Hanle effect that applies to injected spins in larger structures [67]. The same group then went on to study an Al nanoparticle connected to one permalloy and one Al lead [72]. Just as spins can accumulate at a single ferromagnet/normal metal interface [39], here they do so on the Al dot. The crucial point is that in the dot the quantisation axis is defined by the external field, which need not necessarily be collinear with the magnetisation in the permalloy lead. This leads to small but easily measurable magnetoresistance effects.

(*d*) *Spin lifetimes*

The question of the spin lifetime $\tau_{\rm sf}$ on the islands in these structures is an important one, intimately related to spin accumulation, since spins cannot accumulate if they can relax before leaving the dot. The experiments of Ernult *et al.* [62] on Fe dots and Bernand-Mantel *et al.* [68] on Au ones seem to satisfy this condition but there was no attempt to quantify the spin lifetime made in their analysis.

This was done by Wei *et al.* [73], who fabricated lateral structures where a single Al nanograin is connected to Permalloy leads through alumina tunnel barriers. They found that the change in current ΔI upon switching from a P to an AP state hardly changed beyond a critical bias voltage. This can be used to infer the spin lifetime, which was estimated to be ~ 1 μs, and to scale with the electron-phonon relaxation

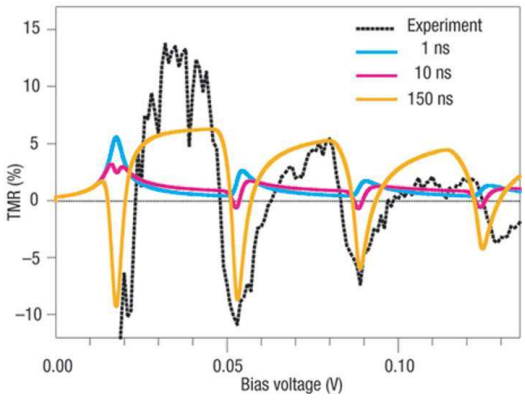

Figure 4. V dependence of the TMR through a Co nanoparticle. The blue, pink and orange curves show the calculated TMR for $\tau_{\rm sf} = 1$ ns, 10 ns and 150 ns, respectively. The dotted curve shows the experimentally observed TMR. The calculated curve assuming the spin-relaxation time of 150 ns well reproduces the experimental one. After Yakushiji *et al.* [20].

rate according to the Elliot-Yafet mechanism [74]. This relatively long lifetime is consistent with Al being a light element where spin-orbit interactions are weak. This can be compared with the work of Mitani *et al.* [70], which suggested $\tau_{\rm sf} \sim 10$ ns on Au dots from the onset current of TMR. This is nevertheless still very long compared to the estimate of 150 ps in the bulk.

Spin lifetime enhancements may also be found in ferromagnetic materials when going from bulk crystals to nanoparticles. Yakushiji *et al.* [20] studied planar Co/AlO_x/Al junctions, with Co nanoparticles embedded in the alumina barrier. In this paper a very clear observation of the oscillating TMR predicted by Barnaś & Fert [45] was made, as shown in figure 4. The fit to these data requires a very long spin lifetime, of the order of 150 ns, which is some 10^4 times longer than is usually found in bulk Co material in the diffusive regime. This is a very exciting development, but basic understanding and technological exploitation are both held back by the degree to which all these these researchers fabrication methods rely on chance due to their self-assembly fabrication method for the nanoparticles. When a factor of 10^4 improvement in spin lifetime is discovered serendipitously, even better results may be found by a systematic search. Indeed, an extremely long value of $\tau_{\rm sf} = 10$ μs was recently measured in a MnAs nanoparticle self-assembled in a GaAs matrix by Hai *et al.* [75].

(*e*) *TMR enhancement by cotunnelling*

In 1997, Ono *et al.* [76] studied a lateral Ni/ NiO/ Co/ NiO/ Ni device, and observed a roughly tenfold enhancement of the TMR on entering the CB regime. Brückl *et al.* [77] also observed a TMR enhancement at low bias in nanoscale Co/AlO_x/NiFe junctions prepared by variable angle evaporation, whilst Zhu & Wang [78] observed a similar effect in a sputtered Fe-Al_2O_3 granular film. In all these cases, the effect is explained by the coherence in electron motion to be found

in the higher order tunnelling processes that are still possible when the usual sequential tunnelling process is forbidden.

As described above in part 2, below the Coulomb gap ($E_C/k_B > T$ and $E_C/e > V$), sequential tunnelling processes are blocked by the Coulomb charging effects. Nevertheless, a small current may still flow, even in the ideal case of no leakages, by considering higher order processes [79], the most commonly discussed of which is known as cotunnelling. There are two versions of this effect. Inelastic cotunnelling can be viewed as two electrons that tunnel simultaneously in a coherent process: one arrives on the island from the source at the same time as another departs to the drain. Elastic cotunnelling is a related second order process where the same electron tunnels through the system.

It was predicted by Takahashi & Maekawa [49] that in the cotunnelling regime the TMR can be substantially enhanced when the magnetisation of the central island is switched against the outer electrodes. According to their paper, in the cotunnelling regime, the resistance of this double magnetic tunnel junction can be written as

$$R_{co} = \frac{3e^2}{2h}\left(\frac{E_C}{k_B T}\right)^2 R_T^2, \tag{4.1}$$

whilst at higher temperatures, the appropriate expression for the thermally assisted sequential regime is

$$R_{seq} = 2\left(1 - \frac{E_C}{3k_B T}\right) R_T, \tag{4.2}$$

where R_T is the resistance of a tunnel junction between an outer electrode and the island, which will depend on its magnetic configuration through the TMR effect. Whilst in the sequential regime the resistance of the double junction is essentially just the sum of the two individual junctions, in the cotunnelling regime it is the product, reflecting the fact that it is a second order process. When R_T increases on magnetic switching into an antiparallel state, we can see from a comparison of equations 4.1 and 4.2 that the fractional gain in resistance of the whole structure will be larger when cotunnelling: this can be quantified by making use of equation 3.1, and it can be shown that the TMR in the cotunnelling regime should be enhanced by a factor of $2/(1 - P^2)$. The predicted enhancement is shown in figure 5.

These effects were explored in some detail by Sukegawa *et al.* [80] using a conventional double junction stack with granular CoFe deposited between two outer pinned CoFe electrodes. The tiny size of the self-assembled CoFe grains yielded Coulomb-gapped $I(V)$ characteristics below ~ 50 K in this case, a substantial increase over the < 1 K temperatures required in the early work of Ono *et al.* [76]. At 7 K the TMR ratio was 24 % at zero bias, roughly double the value measured beyond the gap at $V = 100$ mV. The effective spin polarisation P was estimated to be 32 % from the non-enhanced TMR, leading to a prediction from the $2/(1 - P^2)$ Takahashi & Maekawa expression of an enhanced TMR of 23 %, in excellent agreement with the observed value.

A similar low V, low T enhancement in TMR has been seen by Yang *et al.* [81] in double junctions with CoFe islands, at least for island sizes exceeding an equivalent CoFe layer thickness of about 1 nm. Below this size a TMR suppression at low T was seen attributed to Kondo physics (to be discussed below, in Part f). Most recently, a huge TMR exceeding 1000 % was observed by Jiang *et al.* [82] in

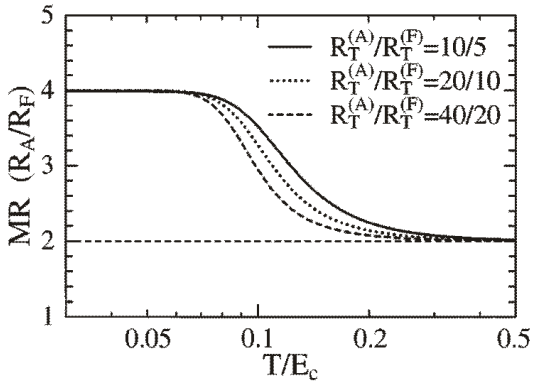

Figure 5. Predicted enhancement of TMR (defined here as the ratio of resistance in the antiparallel state $R_{\rm A}$ to that in the ferromagnetically aligned parallel state $R_{\rm F}$) when entering the cotunnelling regime. The prediction is made for various values of the single junction tunnel resistance $R_{\rm T}$ in these two states with a fixed ratio between the two states. As the temperature T is lowered relative to the charging energy $E_{\rm C}$, the Coulomb blockade regime is entered and the TMR enhanced. After Takahashi & Maekawa [49].

a double MgO barrier MTJ structure with a 1.2 nm thick CoFeB central electrode, below the thickness required for a continuous CoFeB layer. The huge TMR appears at the edges of a region of suppressed differential conductance at low bias, which has a width that changes substantially with magnetic configuration: giving a double peak in TMR(V) that is inconsistent with the predictions of the simple version of the theory described above. We shall return to this very striking result later, in Part 5.

In these three studies the magnetic configuration of the device is one where the ferromagnetic islands are switched between a pair of outer electrodes. Dempsey *et al.* [83, 84] have recently studied double MTJs with conventional free and pinned outer electrodes, with superparamagnetic NiFe nanoparticles forming the central electrode sandwiched between alumina barriers. Whilst the TMR in the CB regime is a little lower than that for a control sample lacking the NiFe islands, the cotunnelling enhancement in the TMR is clearly observed at biases small enough to lie within the Coulomb gap for the double MTJ, as shown in figure 6. This shows that spin information can be propagated through the fluctuating NiFe island moments. This is reasonable since typical tunnelling times are of the order of fs, much shorter than typically superparamagnetic fluctuation lifetimes, which are $\sim$ 0.1-1 ns. The TMR significantly exceeds that arising from NiFe islands alone [85].

(*f*) *Kondo effect in double magnetic tunnel junctions*

The Kondo effect is one of the most interesting phenomena in condensed matter physics. In addition to the CB, the Kondo effect can also arise in quantum dots or nanocluster-based systems, like double magnetic tunnel junctions (DMTJs), where there is strong coupling to the electrodes. This effect is important for single-electron conduction systems since it can compete with CB in determining the TMR. The

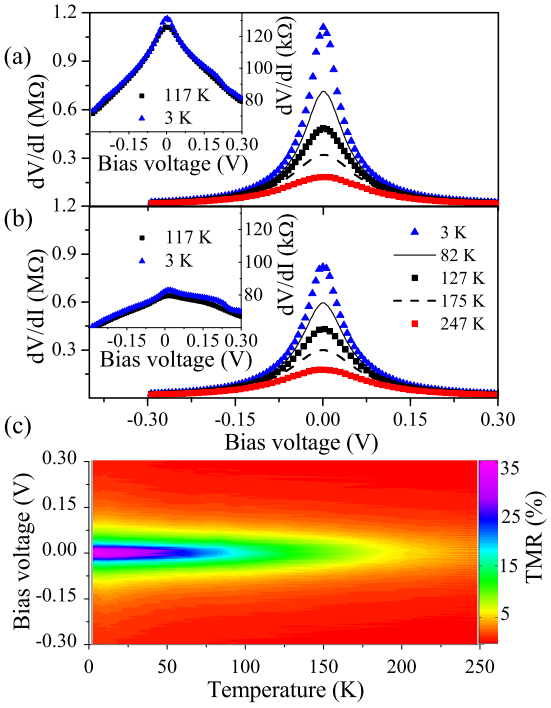

Figure 6. Cotunnelling through superparamagnetic Permalloy nanoparticles embedded in an alumina-based magnetic tunnel junction stack. (a) Typical differential resistance vs bias voltage (R vs V) data for a double barrier magnetic tunnel junction in the anti-parallel outer electrode arrangement, at various temperatures. Inset: R vs V for a comparable single barrier junction at two temperatures. (b) Typical R vs V data for the double barrier junctions in the parallel outer electrode arrangement, various temperatures. Inset: R vs V for the single barrier junction at two temperatures. (c) TMR vs bias voltage and temperature, derived from the full set of I-V curves. After Dempsey *et al.* [84].

Kondo effect cannot only avoid the TMR enhancement achieved in the cotunnelling regime, but suppress it.

The "classical" Kondo effect arises when doping a conductor, such as copper, with a magnetic impurity [86]. In that case, the spin of the impurity interacts with the free electrons forming a singlet state and, as a result, the conductivity saturates when reducing the temperature. The maximum value of the conductivity is achieved for the so called Kondo temperature (T_{K}). This value directly depends on the number of defects introduced in the conductor.

It was Jun Kondo in 1964 who gave the first explanation of these experimental results [87]. He realised that, in the perturbation theory used to study the scattering of electrons due to magnetic impurities, the second-order term can be much larger than the first-order one. Below T_{K}, the scattering cross section of the impurity is enhanced. This only happens when the total spin of all the electrons within the impurity is non-zero. A better understanding of the effect can be achieved through

the Anderson model where the magnetic impurity is modelled as a single quantum state for an electron bounded locally. In principle, this local state can be occupied by two electrons with opposite spins. However, the charging electrostatic energy plays its role making energetically favourable the state to be occupied by just one electron. Only free electrons with opposite spin can tunnel in and out of the impurity. This causes an effective antiferromagnetic coupling at low temperature of the free electrons with those inside the impurity. In other words, the origin of the anomalous increase of the resistance below T_K is due to an exchange process that flips the spin of the free electrons around the impurity. A full description of this physics can be found in Hewson [88].

Surprisingly, in tunnel junctions with magnetic nanoclusters in the insulating layer, the Kondo effect can produce the enhancement of the resistance or its reduction depending on the exact location of the clusters within the layer. Kondo resonances were suggested to be responsible for the increase of the resistance at low temperature and low bias when introducing thin Cr(< 0.4 nm)/Co(< 0.6 nm) impurities in one of the electrode/barrier interfaces of MTJs [89]. Later, Lee *et al.* [90] studied the resistance and TMR of MTJs with nanoclusters in the interface lead/insulator, produced by over-oxidising the alumina layer in $Co_{84}Fe_{16}/Al_2O_3/Co_{84}Fe_{16}$ tunnel junctions. They found that, upon increasing the oxidation time to produce the alumina layer, the TMR was suppressed while the resistance was enhanced at low temperature and low bias. Studying the scaling behaviour of both parameters in the junction, they concluded that it was due to Kondo impurities in the electrode/barrier interface. The impurities act as scattering centres, and they block the conduction of electrons from one electrode to the other.

On the other hand, the Kondo effect produces an increase of the conductance if the insulating tunnel barrier is doped with a magnetic material [91] or some magnetic nanoclusters are placed within this layer [92]. It can be explained with the Anderson model when the clusters are placed within the insulator layer. Classically the electron is confined in the impurity if the energy of the state is below the Fermi level of the leads. However, quantum mechanics allows the electron to tunnel into the lead for a short period of time of about h/ϵ due to the Heisenberg uncertainty principle. Here h is Planck's constant and ϵ the energy of the level in the impurity. During this short period of time, the electron can tunnel into the leads and another electron–maybe with opposite spin–can tunnel from the leads to the impurity. The result is a change of the energy spectrum of the DMTJ due to the spin exchange. When considering this process happening many times, the spin exchange produces a new state close to the Fermi energy called a Kondo resonance. The new state is a conductive path for the free electrons of the leads to easier tunnelling from one lead to the other. It decreases the resistance of the DMTJ.

When studying DMTJs for single-electron applications, taking advantage of the CB, the impurities in the lead/insulator interface can be avoided by correctly engineering the materials. This is not the case when the nanoclusters are intentionally placed within the insulator. Some theoretical work [93, 94], and recent experimental results [81, 95], show that the Kondo effect in TMR can be important even if the magnetic moments have large magnetic anisotropy [96]. Due to the Kondo Effect, TMR can achieve negative values much larger than those estimated by Julliere's model, as measured in DMTJs using nickel electrodes and C60 molecules [97]. Thus, the Kondo effect is of great importance in DMTJs, and its competition with CB

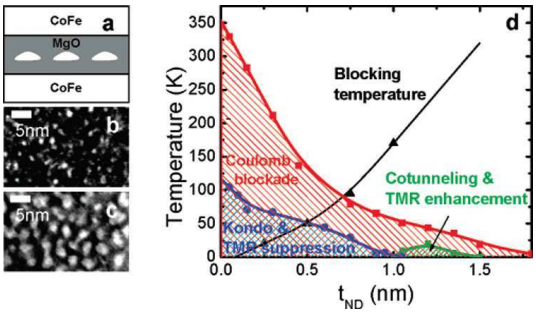

Figure 7. (a) Schematic of a DMTJ with a discontinuous magnetic layer in the middle of the tunnel barrier, forming a layer of nanodots. (b,c) Plan view TEMs for the thickness of this layer being 0.25 nm and 0.75 nm. White regions correspond to nanodots while black regions indicate MgO. (d) Phase diagram showing the dependence of the superparamagnetic blocking temperature T_{B}, the Kondo temperature T_{K}, and the CB temperature on discontinuous layer thickness t_{ND}. After Yang *et al.* [81].

crucial for single-electron applications. Despite its importance, few works have been addressed to study the interplay between both effects. A theoretical discussion of the Kondo effect in single-electron devices can be found in Weis [98].

Yang *et al.* [81] studied the crossover between Kondo TMR suppression and co-tunneling enhancement of TMR in MgO DMTJs. The DMTJs were formed by two CoFe free layers with CoFe nanoclusters within an MgO barrier. The CoFe clusters were produce by growing a layer with a thickness below 1.75 nm. For all the CoFe thicknesses the tunnelling is dominated by sequential tunnelling at high temperature and bias voltage. However, when the temperature is reduced, some typical Kondo signatures can be observed for thicknesses below 1 nm (see figure 7): a zero bias anomaly (ZBA) in the conductance versus bias voltage curve; the temperature dependence of the ZBA; and suppression of the TMR at low bias below a particular temperature. From each of these experimental features, the Kondo temperature can be determined. The crossover from the two effects, Kondo and CB, was found to be correlated with the fluctuations of the magnetic moments of the nanoclusters.

In this research, the temperature dependence of the conductance G is key to determining the presence of the different effects. (See Goldhaber-Gordon *et al.* [99] and references therein for a description of the temperature dependence of G in the presence of Kondo Effect.) It has been theoretically deduced, and experimentally found, that the temperature dependent part of the conductance $G \sim T^{-2}$ when $T \ll T_{\mathrm{K}}$, viz. Fermi liquid behaviour, and that $G \sim -\ln T$ when $T \geqslant T_{\mathrm{K}}$. For intermediate temperatures:

$$G(T) = G_0 \left(\frac{(T'_{\mathrm{K}})^2}{T^2 + (T'_{\mathrm{K}})^2} \right)^S \tag{4.3}$$

where $G_0 = 2G(T_{\mathrm{K}})$ and $T'_{\mathrm{K}} = T_{\mathrm{K}}/(2^{1/S} - 1)$. Here, S is a parameter that depends on the spin of the impurity in the Kondo effect.

Another important issue is the dependence of Kondo effect with the magnetic anisotropy. It has been studied in isolated magnetic atoms in a surface by scanning

tunnelling microscopy. It was found that the magnetic anisotropy determines both the temperature in which Kondo resonance arises and its behaviour under magnetic fields [100, 101]. However, this experiment has not been carried out in solid-state DMTJs yet.

To sum up, the Kondo effect is of fundamental importance in DMTJs and single-electron devices. In DMTJs, the Kondo effect competes with CB providing a limitation of the TMR enhancement due to co-tunnelling. Some theoretical and experimental research is still needed in order to properly understand the role of different parameters like the nanoclusters shape, size, and magnetic anisotropy in the interplay of Kondo and CB physics.

(*g*) *Chemical potential effects*

Modifications of the chemical potential μ in nanodots can have a significant effect on transport through them. This is, after all, the basis of the operation of a gate electrode in a SET. The magneto-Coulomb effect, discovered by Ono *et al.* [76, 102] was studied by theoretically by van der Molen *et al.* [103]. The central issue is that the application of a magnetic field H can modulate the μ in a ferromagnet through the flux density B it gives rise to. Consider a ferromagnetic nanoparticle with spin-resolved densities of states at the Fermi level $g_{\uparrow(\downarrow)}(E_{\mathrm{F}})$, which lead to a thermodynamic spin polarisation $P = [g_{\uparrow}(E_{\mathrm{F}}) - g_{\downarrow}(E_{\mathrm{F}})]/[g_{\uparrow}(E_{\mathrm{F}}) + g_{\downarrow}(E_{\mathrm{F}})]$. This P is very different to the ones involved in spin-polarised tunnelling, which also depends on the spin-resolved tunnelling matrix elements [104]. Application of a field leads to an Zeeman energy shift of $\pm\frac{1}{2}g\mu_{\mathrm{B}}B$, where g is the Landé g-factor and μ_{B} is the Bohr magneton. Since the spin-resolved densities of states differ, there must be an overall change $\Delta\mu = -\frac{1}{2}Pg\mu_{\mathrm{B}}B$ in order to conserve the number of electrons on the dot. Hence, for ferromagnetic dots, a magnetic field may play an equivalent role to a gate electrode, inducing a charge on the dot, which will change discontinuously when the ferromagnetic electrodes switch. This was shown, within the orthodox theory of CB, to produce hysteretic and spin-valve-like $G(H)$ curves, as shown in figure 8, with zero spin accumulation on the dot. Great care must be taken to separate this magneto-Coulomb effect from spin accumulation in experiment.

CB anisotropic magnetoresistance, reported by Wunderlich *et al.* [105], is another effect related to manipulation of μ in a ferromagnetic dot, in this case one with a large spin-orbit coupling. The system studied was a SET fabricated from the dilute magnetic semiconductor (Ga,Mn)As, with the quantum dot comprising a puddle of electrons formed in the disorder left from an almost fully etched channel. The most clear evidence for chemical potential manipulation is found when the channel resistance is measured as a function of gate voltage in a saturating ($B = 5$ T) field applied along different directions, where field-angle dependent shifts in the CB oscillations were observed, with a π-periodicity: the chemical potential shifts are related to the uniaxial magnetocrystalline anisotropy, which arises from spin-orbit coupling, and the magnetisation angle plays the role of a gate voltage. The effect is related to, but distinct from, the tunnelling anisotropic magnetoresistance, TAMR, which is found in ferromagnet/normal metal tunnel junctions [106, 107] with large spin-orbit effects [108]. This spin-orbit coupling deforms the band structure anisotropically around the magnetisation direction. When the magnetisation

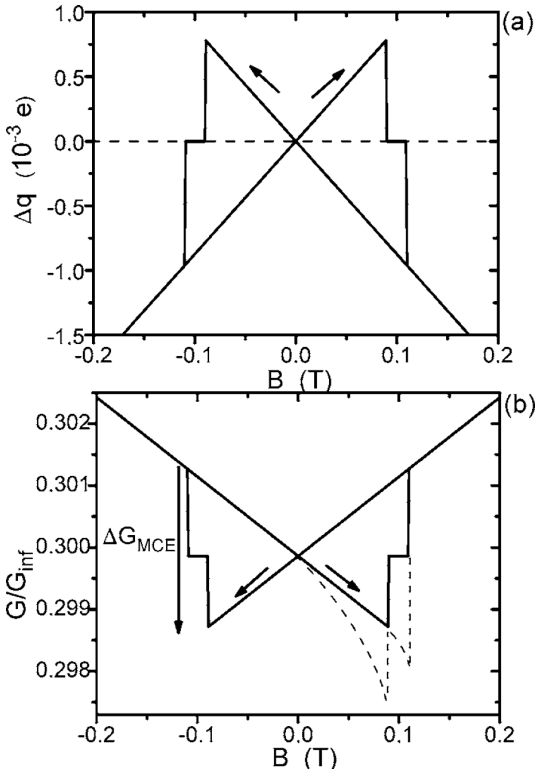

Figure 8. (a) Induced charge on an ferromagnetic nanoisland island, Δq, vs B field. Δq varies linearly with B, except at the switching fields of the ferromagnetic electrodes, where steps are seen. The curve ignores the demagnetising field. (b) Calculated conductance G vs B field, showing spin-valve-like curves. Solid line: demagnetisation field ignored. The sum of the steps is defined as ΔG_{MCE} Dashed line: qualitative effect of the rotation of the demagnetisation field. After van der Molen *et al.* [103]

is then re-oriented by an applied field, the nature of the states presented to the barrier is changed and so the tunnelling rate is affected.

Another related effect was very recently reported by Bernand-Mantel *et al.* [22], who studied transport through a single Au nanoparticle connected to Co leads. The $I(V)$ characteristics of their device could be fitted well using the orthodox theory of CB, with the commonplace requirement of a local charge offset Q_0 reflecting the local electrostatic environment of the nanoparticle. Again, this can be seen as a chemical potential shift $\mu = Q_0 e/C$, with C the capacitance of the dot and e the electronic charge. Remarkably, this charge was found to vary simply with the direction of magnetisation of the two Co electrodes in a saturating field: the moment direction is coupled through spin-orbit interactions to the chemical potential of the dot and hence charge transfer from the leads. A 90° rotation corresponded to a change in background charge $\Delta Q_0 = 0.033e$, giving rise to large conductance changes near the Coulomb steps in the $I(V)$ curve. As with the other magneto-Coulomb effects described above, this essentially allow spintronic SETs to

be constructed where the transistor action is gated by a magnetic, rather than an electric, field.

5. Closing Remarks

It has not been possible to cover every possible topic in this review, and the list of references is far from exhaustive. Many aspects have been covered in the prior reviews of Martinek & Barnaś [109], Seneor, Bernand-Mantel & Petroff [4], Ernult, Yakushiji, Mitani & Takanashi [110], and Barnaś & Weymann [44]. For instance, there have also been studies of quantum dots formed in carbon nanotubes contacted by ferromagnets [see e.g. 111], a topic which we have not covered here. With spin injection demonstrated into graphene [112, 113], a material in which high quality quantum dots may be formed [114, 115], the prospect of single spin devices in this material is in sight [116]. Nevertheless, the overall area of single electron spintronics continues to be a very active area of research. A few of the most recent advances are discussed here.

A very exciting recent result is the generation of an electromotive force by magnetisation dynamics reported by Hai *et al.* [117] in a CB structure based on magnetic semiconductor materials. Similar effects have been predicted for DW motion [118], and observed in Permalloy nanowires by Yang *et al.* [119], although there a clever modulation scheme was needed in order to detect the very small signals. In the work of Hai *et al.* [117], the signals were large, persisted over many minutes, and resulted in TMR ratios exceeding 100,000 %. There have yet to be further studies of this effect reported.

There have also been recent theoretical advances. A "giant" CB MR has been predicted by Zhang *et al.* [120], who put forward a mechanism based on the collective CB picture of Stafford & Das Sarma [121]. The essentials of the theoretical picture are that E_C may be modulated by a magnetic field. This takes place when the magnetic moments of the nanoclusters are brought into alignment, increasing the conductance between them through the usual TMR effect. This allows the collective CB effect to take place, where small groups of nanoclusters group together to form larger effective clusters, reducing the charging energy. This is potentially an explanation for the very large TMR observed by Feng *et al.* [122], Jiang *et al.* [82], and Tan *et al.* [123]. It is worth noting that in principle, only the nanoclusters themselves need be ferromagnetic, the outer electrodes may be made from any metal: there is scope here for new experiments.

Other new theoretical predictions yet to be addressed by experimenters include quantum criticality in the destruction of the Kondo effect[124], self-excited oscillations in spin/charge accumulation at GHz frequencies due to negative differential conductance in double junctions with a single ferromagnetic lead [125], TMR beating effects in double dot devices [126], and all-electrical generation of pure spin currents and spin filtering using quantum dots with high spin-orbit coupling [127].

Almost all of the structures discussed in this article are essentially two-terminal devices. In spite of very extensive theoretical treatment [see 44, and references therein], there have been very few experimental studies of gated structures [see, for instance, 97, 105, 128, 129], which may be described as true spintronic single electron transistors. There is great scope for experimental progress here. Three-terminal structures with wrap-around gates showing single electron effects at room

temperature have very recently been demonstrated using non-magnetic materials, using processes compatible with the usual industrial planar processing methods [11]. It is now possible to prepare magnetic nanoparticles with very well-defined sizes under ultrahigh vacuum conditions [130, 131], ideal for incorporation into spintronic versions of such structures, offering a realistic prospect of useful devices based on single spin effects.

Of course, single electron spintronic devices allow proper access to the quantum properties of the spin, essential for realising any quantum information technology, and we can envisage that at some point in the not-too-distant future all spintronic devices will be fully "quantum", perhaps with quantum computers based on the type of architecture proposed by Loss & DiVincenzo [132]. Rather than discuss this here, we refer the reader to the article in this issue by Ardavan & Briggs [5].

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