Spintronics

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Origin, Development, and Future of Spintronics (Nobel Lecture)**

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From the Contents

1. Overview

Electrons have a charge and a spin, but until recently, charges and spins have been considered separately. In conventional electronics, the charges are manipulated by electric fields but the spins are ignored. Other classical technologies, such as magnetic recording, use the spin but only through its macroscopic manifestation, the magnetization of a ferromagnet. This picture started to change in 1988 when the discovery[1,2] of giant magnetoresistance (GMR) of the magnetic multilayers opened the way to efficient control of the motion of the electrons by acting on their spin through the orientation of a magnetization. This rapidly triggered the development of a new field of research and technology, which is today called spintronics and exploits the influence of the spin on the mobility of the electrons in ferromagnetic materials. Actually, the influence of the spin on the mobility of the electrons in ferromagnetic metals, first suggested by Mott,[3] had been experimentally demonstrated and theoretically described in my PhD thesis more than ten years before the discovery in 1988. The discovery of GMR was the first step on the road towards exploiting this influence to control an electrical current. Its application to the read head of hard disks greatly contributed to the fast rise in the density of stored information and led to the extension of hard-disk technology to consumer electronics. Then, the development of spintronics revealed many other phenomena related to the control and manipulation of spin currents. Today this field of research is extending considerably, with very promising new directions such as spin transfer, spintronics with semiconductors, molecular spintronics, or single-electron spintronics.

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2. From Spin-Dependent Conduction in Ferromagnets to Giant Magnetoresistance

GMR and spintronics have their roots in previous research on the influence of the spin on the electrical conduction in ferromagnetic metals.[3–5] The dependence of the conduction on spin can be understood from the typical band structure of a ferromagnetic metal (Figure 1 a). The splitting between the energies of the "majority spin" and "minority spin" directions (spin up and spin down in the usual notation) means that the electrons at the Fermi level, which carry the electrical current, are in different states for opposite spin directions and exhibit different conduction properties. This spin-dependent conduction was proposed by Mott[3] in 1936 to explain some features of the resistivity of ferromagnetic metals at the Curie temperature. However, in 1966, when I started my PhD thesis, the subject was still almost completely unexplored. My supervisor, Ian Campbell, proposed that I investigate it with experiments on Ni- and Fe-based alloys, and I had the privilege to be at the beginning of the study of this topic. I could confirm that the mobility of the electrons was spin-dependent and, in particular, I showed that the resistivities of the two channels can be very different in metals doped with impurities presenting a strongly spin-dependent scattering cross section.[4] Figure 1 b shows the example of the spin-up (majority spin) and spin-down (minority spin) resistivities of nickel doped with 1 % of different types of impurities. It can be seen that the ratio α of the spin-down resistivity to the spin-up one can be as large as 20 for Co impurities or, as well, smaller than one for Cr or V impurities, which is consistent with the theoretical models developed by Jacques Friedel for the electronic structures of these impurities. The two-current conduction was rapidly confirmed in other groups and extended, for example, to Co-based alloys by Loegel and Gautier[5] in Strasbourg.

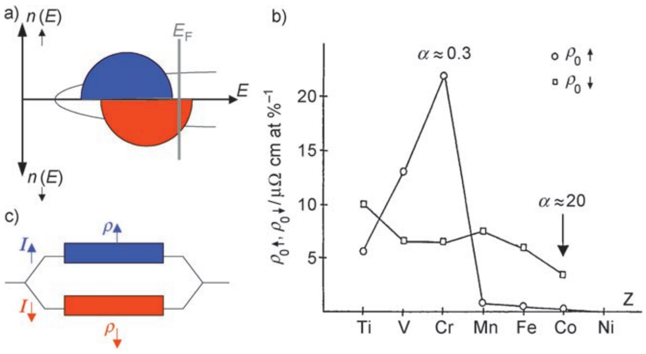

Figure 1. Basics of spintronics. a) Band structure of a ferromagnetic metal showing the spin splitting of the energy bands. $n(E)$ = charge carrier density at energy E; E_F = Fermi level energy. b) Resistivities of the spin-up and spin-down conduction channels for nickel doped with 1 % of several types of impurity (measurements at 4.2 K).[4] The ratio α between the resistivities $\rho_{0\downarrow}$ and $\rho_{0\uparrow}$ of the spin-up and spin-down channels can be as large as 20 (Co impurities) or smaller than one (Cr or V impurities). c) Spin-dependent conduction through independent spin-up and spin-down channels in the limit of negligible spin mixing ($\rho_{\uparrow\downarrow} = 0$ in the formalism of reference [4]).

In my thesis, I also worked out the so-called two-current model[4] for the conduction in ferromagnetic metals. This model is based on a picture of spin-up and spin-down currents coupled by spin mixing, that is, by momentum exchange. Spin mixing comes from spin-flip scattering, mainly from electron-magnon scattering, which increases with temperature and equalizes partly the spin-up and spin-down currents at room temperature in most ferromagnetic metals. The two-current model is the basis of spintronics today, but, surprisingly, the interpretation of spintronics phenomena is generally based on a simplified version of the model neglecting spin mixing and assuming that the conduction by two independent channels in parallel (Figure 1 c). It should be certainly useful to revisit the interpretation of many recent experiments by taking into account the spin-mixing contributions (note that the mechanism of spin mixing should not be confused with the relaxation of spin accumulation by other types of spin flips[6]).

As a matter of fact, some experiments of my thesis with metals doped with two types of impurities[4] were already anticipating GMR. This is illustrated by Figure 2. Suppose, for example, that nickel is doped with impurities of Co, which scatter strongly the electrons of the spin-down channel, and with impurities of rhodium, which scatter strongly the spin-up electrons. In the ternary alloy Ni(Co+Rh), which I call type #1, the electrons of both channels are strongly scattered either by Co or by Rh, so that the resistivity is strongly enhanced. In contrast, there is no such enhancement in alloys of type #2 doped with impurities (Co and Au for example) scattering strongly the electrons in the same channel and leaving the second channel open. The idea of GMR is the replacement of

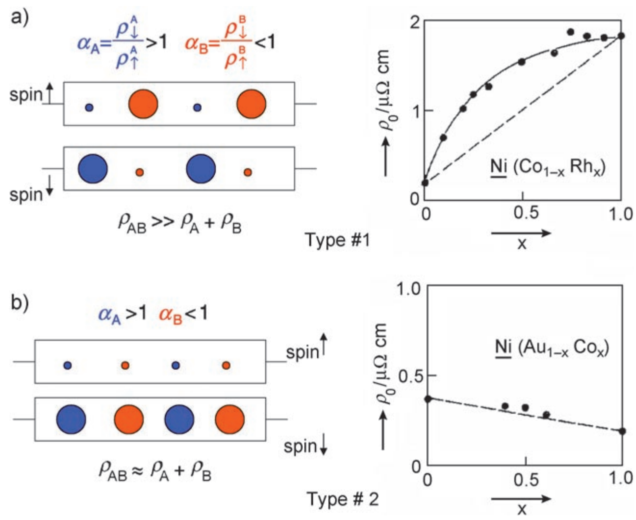

Figure 2. Experiments on ternary alloys based on the same concept as that of the GMR.[4] a) Schematic for the spin-dependent conduction in alloys doped with impurities of opposite scattering spin asymmetries ($\alpha_A = \rho_{A\downarrow}/\rho_{A\uparrow} > 1$, $\alpha_B = \rho_{B\downarrow}/\rho_{B\uparrow} < 1$, $\rho_{AB} \gg \rho_A + \rho_B$) and experimental results for Ni($Co_{1-x}Rh_x$) alloys. b) The same for alloys doped with impurities of similar scattering spin asymmetries ($\alpha_A = \rho_{A\downarrow}/\rho_{A\uparrow} > 1$, $\alpha_B = \rho_{B\downarrow}/\rho_{B\uparrow} > 1$, $\rho_{AB} \approx \rho_A + \rho_B$) and experimental results for Ni-($Au_{1-x}Co_x$) alloys. In GMR, the impurities A and B are replaced by multilayers; the situation in (a) and (b) corresponds to the antiparallel and parallel magnetic configurations, respectively, of adjacent magnetic layers.

the impurities A and B of the ternary alloy by layers A and B in a multilayer, whereby the antiparallel magnetic configuration of the layers A and B corresponds to an alloy of type #1 and parallel configuration corresponds to type #2. This opens the possibility of switching between high- and low-resistivity states by simply changing the relative orientation of the magnetizations of layers A and B from antiparallel to parallel. However, the transport equations tell us that the relative orientation of layers A and B can be felt by the electrons only if their distance is smaller than the electron mean free path, that is, practically, if they are spaced by only a few nanometers. Unfortunately, in the 1970s, it was not technically possible to make multilayers with layers as thin as a few nanometers. I put some of my ideas on ice and, in my team at the Laboratoire de Physique des Solides of the Université Paris-Sud, from the beginning of the 1970s until 1985, I worked on other topics such as the extraordinary Hall effect, the spin Hall effect, the magnetism of spin glasses, and amorphous materials.

In the mid-1980s, with the development of techniques such as molecular beam epitaxy (MBE), it became possible to fabricate multilayers composed of very thin individual layers, and I could consider trying to extend my experiments to ternary alloys to multilayers. In addition, in 1986, I saw the beautiful Brillouin scattering experiments of Peter Grünberg and co-workers[7] revealing the existence of antiferromagnetic interlayer exchange couplings in Fe/Cr multilayers. Fe/Cr appeared as a magnetic multilayered system in which it was possible to switch the relative orientation of the magnetization in adjacent magnetic layers from antiparallel to parallel by applying a magnetic field. In collaboration with the group of Alain Friederich at the Thomson-CSF company, I started the fabrication and investigation of Fe/Cr multilayers. The MBE expert at Thomson-CSF was Patrick Etienne, and my three PhD students, Frédéric Nguyen Van Dau first and then Agnès Barthélémy and Frédéric Petroff, were also involved in the project. This led us in 1988 to the discovery[1] of very large magnetoresistance effects that we called GMR (Figure 3a). Effects of the same type in Fe/Cr/Fe trilayers were obtained at practically the same time by Peter Grünberg at Jülich[2] (Figure 3b). The interpretation of the GMR is similar to that described above for the ternary alloys and is illustrated by Figure 3c. The first classical model of the GMR was published in 1989 by Camley and Barnas[8] and I collaborated with Levy and Zhang for the first quantum model[9] in 1991.

I am often asked if I was expecting such large MR effects. My answer is yes and no: on the one hand, a very large magnetoresistance could be expected from an extrapolation of my preceding results on ternary alloys; on the other hand, one could fear that the unavoidable structural defects of the multilayers, interface roughness, for example, might introduce spin-independent scattering that cancels the spin-dependent scattering inside the magnetic layers. The good luck was finally that the scattering by the roughness of the interfaces is also spin-dependent and adds its contribution to the "bulk" one (the "bulk" and interface contributions can be separately derived from CPP-GMR experiments).

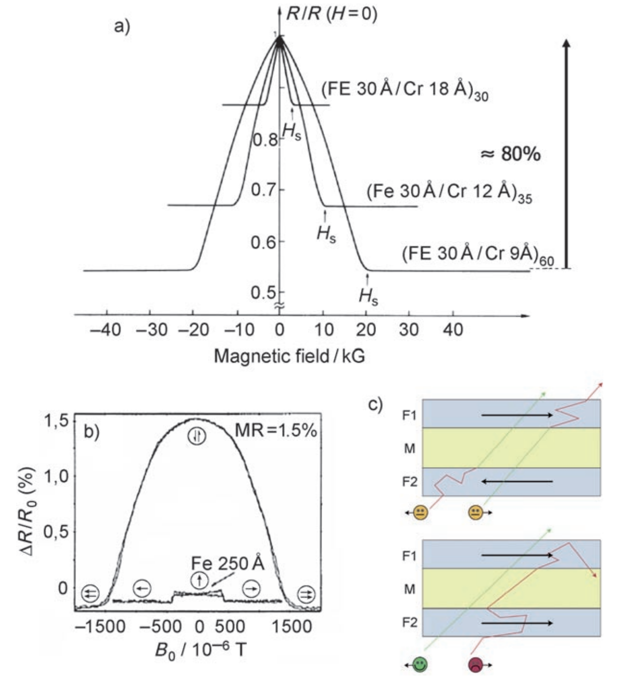

Figure 3. First observations of giant magnetoresistance. a) Fe/Cr(001) multilayers[1] (magnetoresistance ratio [$MR = 100\,(R_{AP} - R_P)/R_p$] for the Fe (3 nm)/Cr (0.9 nm) multilayer of MR = 85%). b) Fe/Cr/Fe trilayers.[2] c) Mechanism of GMR. In the parallel magnetic configuration (bottom), the electrons of one of the spin directions can go easily through all the magnetic layers and the short circuit through this channel lead to a small resistance. In the antiparallel configuration (top), the electrons of each channel are slowed down by every second magnetic layer and the resistance is high (from reference [18]).

3. The Golden Age of GMR

Our papers reporting the discovery of GMR quickly attracted attention for their fundamental interest as well as for the many possibilities of applications, and the research on magnetic multilayers and GMR became a very hot topic. In my team, reinforced by the recruitment of Agnés Barthélémy and Frédéric Petroff, as well as in the small but rapidly increasing community working in the field, we had the exalting impression of exploring a wide virgin country with so many amazing surprises in store. On the experimental side, two important results were published in 1990. Parkin et al.[10] demonstrated the existence of GMR in multilayers made by the simpler and faster technique of sputtering (Fe/Cr, Co/Ru and Co/Cr), and found the oscillatory behavior of the GMR caused by the oscillations of the interlayer exchange as a function of the thickness of the nonmagnetic layers. Also in 1990, Shinjo and Yamamoto,[11] as well as Dupas et al.,[12] demonstrated that GMR effects can be found in multilayers without antiferromagnetic interlayer coupling but composed of magnetic layers of different coercivities. Another important result, in 1991, was the observation of large and oscillatory GMR effects in Co/Cu, which became the archetypical GMR system (Figure 4a). The first observations[13] were obtained in my group by PhD student Dante

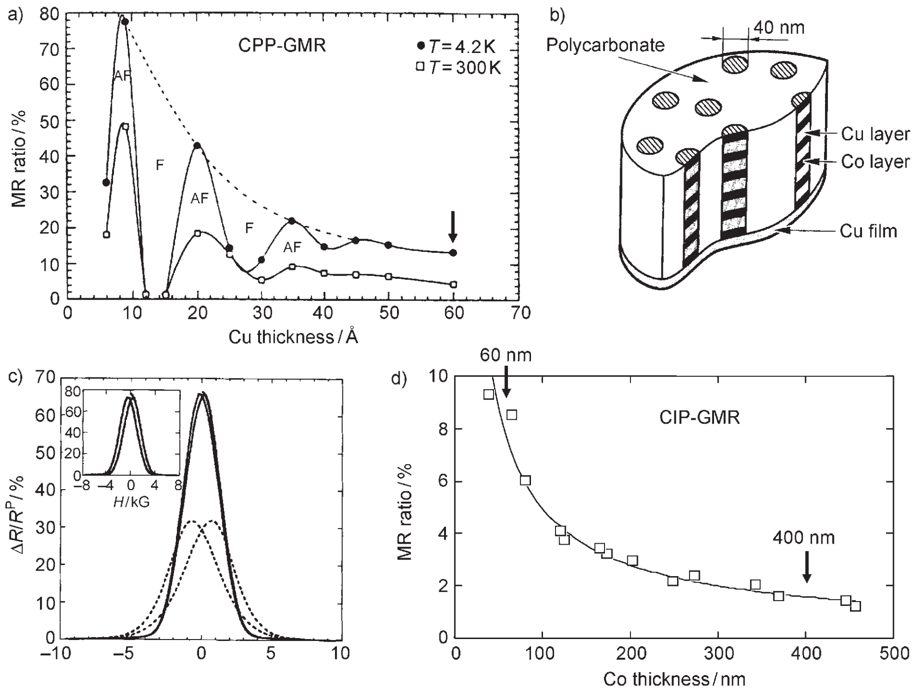

Figure 4. a) Variation of the GMR ratio of Co/Cu multilayers in the conventional current in plane (CIP) geometry as a function of the thickness of the Cu layers.[13] The scaling length of the variation is the mean free path (short). b) Structure of multilayered nanowires used for CPP-GMR measurements. c) CPP-GMR curves for permalloy (12 nm)/copper (4 nm) multilayered nanowires (solid lines) and cobalt (10 nm)/copper (5 nm) multilayered nanowires (dotted lines).[21] d) Variation of the CPP-GMR ratio of Co/Cu multilayered nanowires as a function of the thickness of the Co layers.[21] The scaling length of the variation is the spin diffusion length (long).

Mosca with multilayers prepared by sputtering at Michigan State University and at about the same time in the group of Stuart Parkin at IBM.[14] Also in 1991, Dieny et al.[15] reported the first observation of GMR in spin valves, that is, trilayered structures based on a concept of my co-laureate Peter Grünberg[16] in which the magnetization of one of the two magnetic layers is pinned by coupling with an antiferromagnetic layer while the magnetization of the second one is free. The magnetization of the free layer can be reversed by very small magnetic fields, so that the concept is now used in most applications.

Other developments of the research on magnetic multilayers and GMR at the beginning of the 1970s are described in the Nobel lecture of my co-laureate Peter Grünberg, with, in particular, a presentation of the various devices bases on the GMR of spin-valve structures.[17,18] In the read heads (Figure 5) of the hard-disk drives (HDDs), GMR sensors based on spin valves replaced AMR (anisotropic magnetoresistance) sensors in 1997. GMR, by providing a sensitive and scalable read technique, has led to an increase of the recording density by more than two orders of magnitude (from ca. 1 to ca. 600 Gbit inch^{-2} in 2007). This increase opened the way both to unprecedented drive capacities (up to 1 terabyte) for video recording or backup and to smaller HDD sizes (down to 0.85 inch disk diameter) for mobile appliances such as ultralight laptops or portable multimedia players. GMR sensors are also used in many other types of application, mainly in automotive industry and biomedical technology.[19]

4. CPP-GMR and Spin Accumulation Physics

During the first years of the research on GMR, experiments were performed only with currents flowing along the layer planes, in the geometry we call CIP (current in plane). It was only in 1993 that experiments of CPP-GMR were first performed, that is experiments of GMR with the current perpendicular to the layer planes. The first experiments were performed by Bass, Pratt, and Shroeder at Michigan State University,[20] who sandwiched a magnetic multilayer between superconducting electrodes, and, a couple of years later, by a collaboration of my group with Luc Piraux at the University of Louvain, by electrodepositing the multilayer into the pores of a polycarbonate membrane (Figure 4b–d).[21] In the CPP geometry, the

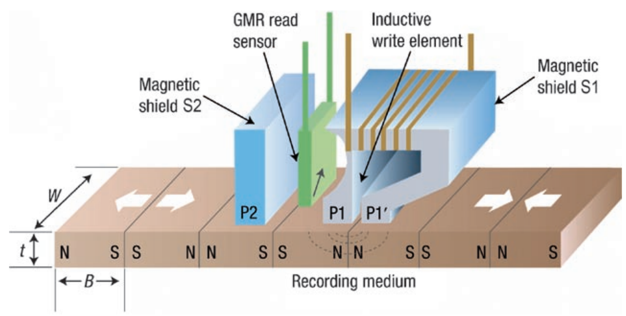

Figure 5. GMR head for hard-disk drives. Figure from Chappert et al.[18] W = track width, t = magnetic film thickness, B = bit length.

GMR is not only definitely higher than in CIP geometry (CPP-GMR will be probably used in a future generation of read heads for hard disks), but also subsists in multilayers with relatively thick layers, up to the micrometer range (Figure 4c–d).[21] In a theoretical paper with Thierry Valet,[22] I showed that, owing to spin accumulation effects occurring in the CPP-geometry, the length scale of the spin transport becomes the long spin-diffusion length in place of the short mean free path for the CIP geometry. Actually, CPP-GMR has revealed the spin-accumulation effects that govern the propagation of a spin-polarized current through a succession of magnetic and nonmagnetic materials and play an important role in all the current developments of spintronics. The diffusion current induced by the accumulation of spins at the magnetic/nonmagnetic interface is the mechanism driving a spin-polarized current at a long distance from the interface, well beyond the ballistic range (i.e. well beyond the mean free path) up to the distance of the spin-diffusion length (SDL). In carbon molecules for example, the spin-diffusion length exceeds the micrometer range and, as we will see in the section on molecular spintronics, strongly spin-polarized currents can be transported throughout long carbon nanotubes.

The physics of the spin-accumulation occurring when an electron flux crosses the interface between a ferromagnetic and a nonmagnetic material is explained in Figure 6. Far from the interface on the magnetic side, the current is larger in one of the spin channels (spin up in Figure 6), whereas, far from the interface on the other side, it is equally distributed in the two channels. With the current direction and the spin polarization in Figure 6, there is accumulation of spin-up electrons (and depletion of spin-down electrons for charge neutrality) around the interface, or, in other words, splitting between the Fermi energy levels (chemical potentials) of the spin-up and spin-down electrons. This accumulation diffuses from the interface in both directions to the distance of the SDL. Spin flips are also generated by this out-of-equilibrium distribution, and a steady splitting is reached when the number of spin flips is just what is needed to adjust the incoming and outgoing fluxes of spin-up and spin-down electrons. To sum up, there is a broad zone of spin accumulation that extends on both sides to the distance of the SDL and in which the current is progressively depolarized by the spin flips generated by the spin accumulation.

Figure 6 is drawn for the case of spin injection, that is, for electrons going from the magnetic to the nonmagnetic conductor. For electrons going in the opposite direction (spin extraction), the situation is similar except that spin accumulation in the opposite direction progressively polarizes the current in the nonmagnetic conductor. In both the injection and extraction cases, the spin polarization subsists or starts in the nonmagnetic conductor at a long distance from the interface. This physics can be described by new types of transport equation[22] in which the electrical potential is replaced by a spin- and position-dependent electrochemical potential. These equations can be applied not only to the simple case of a single interface but also to multi-interface systems with overlap of the spin accumulations at successive interfaces. They can also be extended to take into account band bending and high current density effects.[23,24]

The physics of spin accumulation play an important role in many fields of spintronics, for example, in one of the most active field of research today, spintronics with semiconductors. In the case of spin injection from a magnetic metal into a nonmagnetic semiconductor (or spin extraction for the opposite current direction), the much larger density of states in the metal means that similar spin accumulation splittings on the two sides of the interface (as in Figure 6) lead to a much larger spin accumulation density and to a much larger number of spin flips on the metallic side. The depolarization is therefore faster on the metallic side and the current is almost completely depolarized when it enters

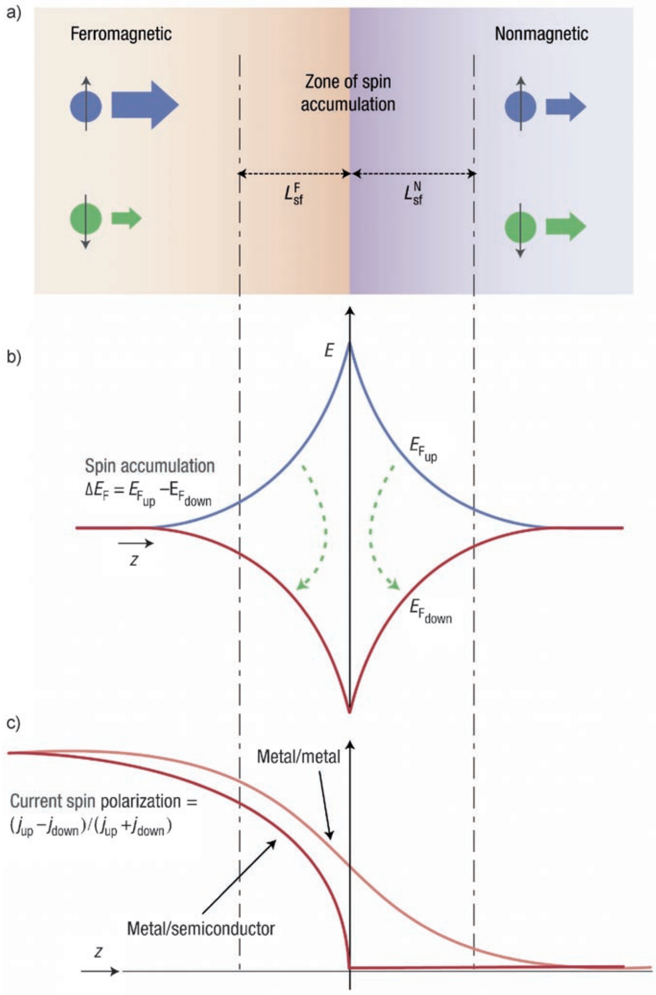

Figure 6. Schematic representation of the spin accumulation at an interface between a ferromagnetic metal and a nonmagnetic layer. a) Spin-up and spin-down currents far from an interface between ferromagnetic and nonmagnetic conductors (outside the spin-accumulation zone). b) Splitting of the chemical potentials $E_{F\uparrow}$ and $E_{F\downarrow}$ at the interface. The arrows symbolize the spin flips induced by the spin-split out of equilibrium distribution. These spin flips control the progressive depolarization of the electron current between the left and the right. With an opposite direction of the current, there is an inversion of the spin accumulation and opposite spin flips, which polarizes the current when it goes through the spin-accumulation zone. c) Variation of the current spin polarization when there is an approximate balance between the spin flips on both sides (metal/metal) and when the spin flips on the left side are predominant (metal/semiconductor without spin-dependent interface resistance, for example). Figure from reference [18].

the semiconductor, as shown in Figure 6c. This problem was first raised by Schmidt and co-workers.[25] I came back to the theory with my co-worker Henri Jaffrès to show that the problem can be solved by introducing a spin-dependent interface resistance, typically a tunnel junction, to introduce a discontinuity of the spin accumulation at the interface, increase the proportion of spin on the semiconductor side, and shift the depolarization from the metallic to the semiconductor side (the same conclusions appear also in a paper of Rashba).[26,27] Spin injection through a tunnel barrier has now been achieved successfully in several experiments, but the tunnel resistances are generally too large for efficient transformation of the spin information into an electrical signal.[24]

5. Magnetic Tunnel Junctions and Tunneling Magnetoresistance (TMR)

An important stage in the development of spintronics was the research on the tunneling magnetoresistance (TMR) of magnetic tunnel junctions (MTJs). MTJs are tunnel junctions with ferromagnetic electrodes, and their resistance is different for the parallel and antiparallel magnetic configurations of their electrodes. Some early observations of TMR effects, small and at low temperature, had already been reported by Jullière[28] in 1975, but they were not easily reproducible and actually could not be really reproduced for 20 years. It was only in 1995 that large (ca. 20%) and reproducible effects were obtained by Moodera's and Miyasaki's groups on MTJ with a tunnel barrier of amorphous alumina.[29,30] From a technological point of view, the interest in MTJs with respect to the metallic spin valves focuses on the vertical direction of the current and the resulting possibility of decreasing the lateral size to a submicrometer scale by lithographic techniques. MTJs are at the basis of a new concept of magnetic memory called MRAM (magnetic random access memory) and schematically represented in Figure 7a. MRAM is expected to combine the short access time of semiconductor-based RAM and the nonvolatile character of magnetic memory. In the first MRAM, put on the market in 2006, the memory cells are MTJs with an alumina barrier. The magnetic fields generated by "word" and "bit" lines are used to switch their magnetic configuration (Figure 7a). The next generation of MRAM, based on MgO tunnel junctions and switching by spin transfer, is expected to have a much stronger impact on the technology of computers.

Research on TMR has been very active since 1995, and the most important step was the recent transition from MTJs with amorphous tunnel barriers (alumina) to single-crystal MTJs and especially MTJs with MgO barriers. In the CNRS/Thales laboratory we founded in 1995, research on TMR was one of our main projects and, in collaboration with a Spanish group, we obtained one of the very first results[31] on MTJs with epitaxial MgO. However, the TMR we observed was only slightly larger than that found with alumina barriers and similar electrodes. The important breakthrough came in 2004, when researchers from Tsukuba[32] and IBM[33] found that very large TMR ratios (up to 200% at room temperature) could be obtained from MgO MTJs of very high structural quality. TMR ratios of about 600% have been now reached (Figure 7b).[34] In such MTJs, the single-crystal barrier filters the symmetry of the wave functions of the tunneling electrons,[35–37] so that the TMR depends on the spin polarization of the electrodes for the selected symmetry.

The high spin polarization obtained by selecting the symmetry of the tunneling waves with a single-crystal barrier is a very good illustration of what is meant by the word "spin polarization" in a spintronic experiment. In the example in Figure 8, taken from an article by Zhang and Butler,[37] one sees the density of states of evanescent wave functions of different symmetries (Δ_1, Δ_5, etc.) in a MgO(001) barrier between Co electrodes. The key point is that, at least for interfaces of high quality, an evanescent wave function of a given symmetry is connected to the Bloch functions of the same symmetry at the Fermi level of the electrodes. For Co electrodes, the Δ_1 symmetry is well represented at the Fermi level in the majority spin direction sub-band and not in the minority one. Consequently, a good connection of the slowly decaying channel Δ_1 with both electrodes can be obtained only in their parallel magnetic configuration, which explains the very high TMR. Other types of barrier can select other symmetries than the symmetry Δ_1 selected by MgO(001). For example, a $SrTiO_3$ barrier selects predominantly evanescent wave functions of Δ_5 symmetry, which are connected to

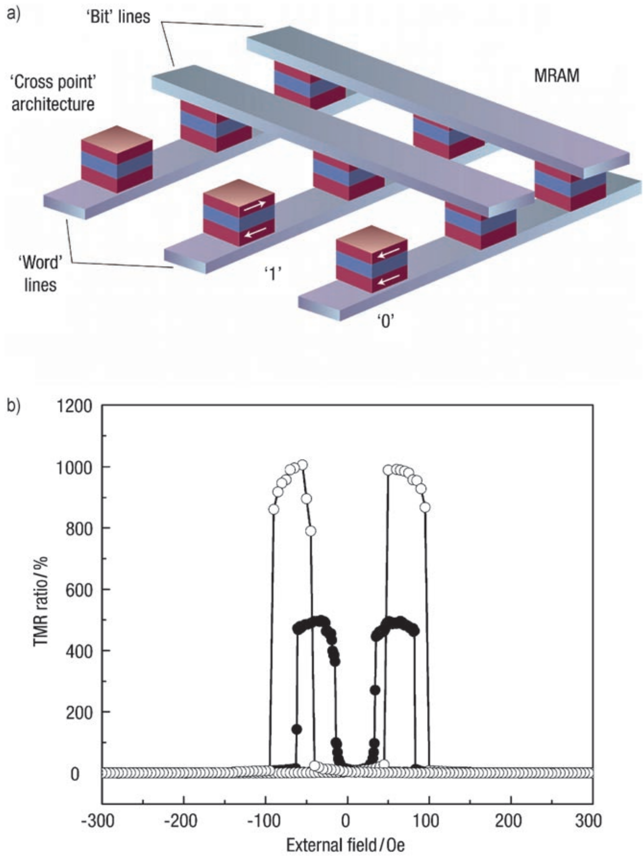

Figure 7. a) Principle of MRAM in the basic "cross-point" architecture. The binary information "0" and "1" is recorded on the two opposite orientations of the magnetization of the free layer of magnetic tunnel junctions (MTJs), which are connected to the crossing points of two perpendicular arrays of parallel conducting lines. For writing, current pulses are sent through one line of each array, and only at the crossing point of these lines is the resulting magnetic field high enough to orient the magnetization of the free layer. For reading, one measures the resistance between the two lines connecting the addressed cell. Reproduced from reference [18]. b) High magnetoresistance, $TMR = (R_{max}-R_{min})/R_{min}$, measured by Lee et al.[34] for the magnetic stack: $(Co_{25}Fe_{75})_{80}B_{20}$ (4 nm)/MgO (2.1 nm)/$(Co_{25}Fe_{75})_{80}B_{20}$ (4.3 nm) annealed at 475 °C after growth, measured at room temperature (black circles) and low temperature (open circles).

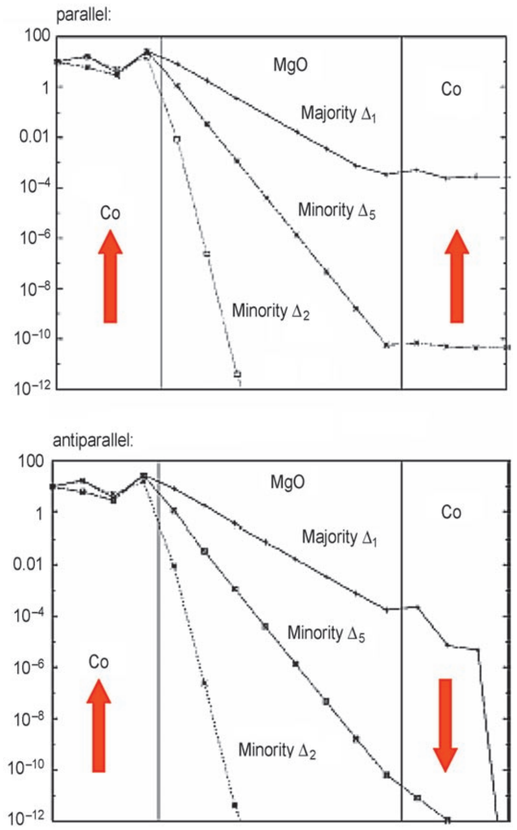

Figure 8. Physics of TMR illustrated by the decay of evanescent electronic waves of different symmetries in a MgO(001) layer between cobalt electrodes calculated by Zhang and Butler.[37] The Δ_1 symmetry of the slowly decaying tunneling channel is well represented at the Fermi level of the spin-conduction band of cobalt for the majority spin direction and not for the minority spin one, so that a good connection by tunneling between the electrodes exists only for the parallel magnetic configuration when a Δ_1 channel can be connected to both electrodes (above). In the antiparallel configuration (below), both the spin-up and spin-down Δ_1 channels are poorly connected on one of the sides, which explains the very high TMR of this type of junction.

minority spin states of cobalt.[38] This explains the negative effective spin polarization of cobalt we observed in $SrTiO_3$-based MTJs.[39] This finally shows that there is no intrinsic spin polarization of a magnetic conductor. The effective polarization of a given magnetic conductor in a MTJ depends on the symmetry selected by the barrier and, depending on the barrier, can be positive or negative, large or small. In the same way, the spin polarization of metallic conduction depends strongly on the spin dependence of the scattering by impurities, as illustrated by Figure 1 b.

There are other promising directions to obtain large TMRs, and experiments in several of them are now led by Agnès Barthélémy (much more than by myself) in the CNRS/Thales laboratory. First, we tested ferromagnetic materials that were predicted to be half-metallic, that is, metallic for one spin direction and insulating for the other (in other words, 100% spin-polarized). Very high spin polarization (95%) and record TMR (1800%) were obtained by our PhD student Martin Bowen with $La_{2/3}Sr_{1/3}MnO_3$ electrodes,[40] but the Curie temperature of this manganite (around 350 K) is too low for applications. It now turns out from recent results in Japan[41] that ferromagnets of the family of Heusler alloys also have very large TMR ratios of still 90% at room temperature.[41] Another interesting concept that we are exploring is spin filtering by tunneling through a ferromagnetic insulator layer.[42,43] This method can be described as the tunneling of electrons through a barrier of spin-dependent height if the bottom of the conduction band is spin-split, which gives rise to a spin dependence of the transmission probability (spin filtering). Very high spin-filtering coefficients have been found at low temperature with EuS barriers[42] by groups at MIT and in Eindhoven. Promising results with insulating ferromagnets of much higher Curie temperature have been recently obtained (see, for example, reference [43]). Some of the magnetic barriers we have recently tested in MTJs are also ferroelectric, so that the MTJs present the interesting property of four states of resistance corresponding to the P and AP magnetic configurations and to the two orientations of the ferroelectric polarization (Figure 9).[44]

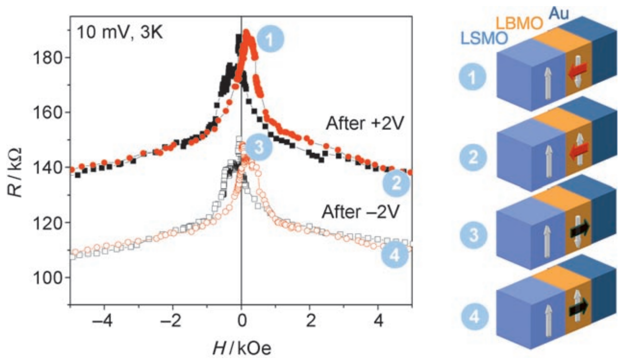

Figure 9. Four-state resistance of a tunnel junction composed of a biferroic tunnel barrier ($La_{0.1}Bi_{0.9}MnO_3$; LBMO) between a ferromagnetic electrode of $La_{2/3}Sr_{1/3}MnO_3$ (LSMO) and a nonmagnetic gold electrode. The states 1–4 correspond to the magnetic (white arrows) and electric (black arrows) polarizations represented on the right of the figure. From Gajek et al.[44]

6. Magnetic Switching and Microwave Generation by Spin Transfer

The study of the spin-transfer phenomena is one of the most promising new directions in spintronics today and also an important research topic in our group at the CNRS/Thales laboratory. In spin-transfer experiments, one manipulates the magnetic moment of a ferromagnetic body without applying any magnetic field but only by transfer of spin angular momentum from a spin-polarized current. The concept, which was introduced by John Slonczewski[45] and appears also in papers of Berger,[46] is illustrated in Figure 10. The transfer of a transverse spin current to the "free" magnetic layer F_2 can be described by a torque acting on its magnetic moment. This torque can induce irreversible switching of this magnetic moment or—in a second regime, generally in the presence of

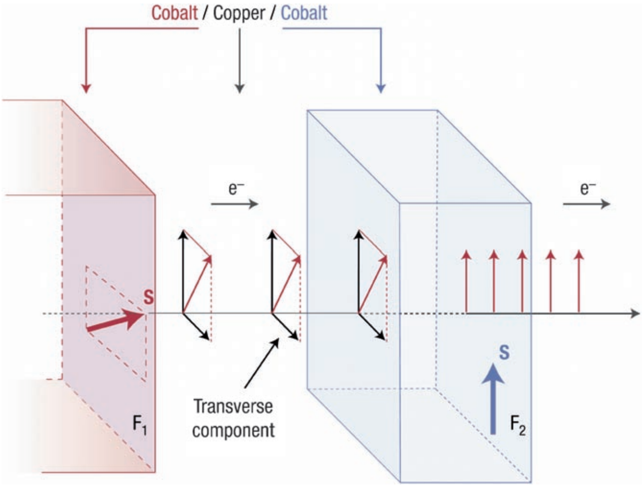

Figure 10. Illustration of the spin-transfer concept introduced by John Slonczewski[45] in 1996. A spin-polarized current is prepared by a first magnetic layer F_1 with an obliquely oriented spin polarization with respect to the magnetization axis of a second layer F_2. When this current goes through F_2, the exchange interaction aligns its spin polarization along the magnetization axis. As the exchange interaction is spin-conserving, the transverse spin polarization lost by the current is transferred to the total spin of F_2, which can also be described by a spin-transfer torque acting on F_2. This can lead to a magnetic switching of the F_2 layer or, depending on the experimental conditions, to magnetic oscillations in the microwave frequency range. Figure from reference [18].

10^5 Amp cm^{-2}, which is smaller than that of the metallic pillar by two orders of magnitude. This is because a smaller number of individual spins is required to switch the smaller total spin momentum of a dilute magnetic material.

In the presence of a large enough magnetic field, the regime of irreversible switching of the magnetization of the "free" magnetic layer in a trilayer is replaced by a regime of steady precessions of this free layer magnetization sustained by the spin-transfer torque.[52] As the angle between the magnetizations of the two magnetic layers varies periodically during the precession, the resistance of the trilayer oscillates as a function of time, which generates voltage oscillations in the microwave frequency range. In other conditions, the spin-transfer torque can also be used to generate an oscillatory motion of a magnetic vortex.

The spin-transfer phenomena raise a series of theoretical problems. The determination of the spin-transfer torque is related to the solution of spin-transport equations,[53–56] while the description of the switching or precession of the magnetization raises problems of nonlinear dynamics.[53] All these problems are interacting, and some of our recent results show that it is possible to obtain very different dynamics by introducing very different spin relaxation times in the two magnetic layers of a trilayer to distort the angular dependence of the torque.[57]

The spin-transfer phenomena will certainly have important applications. Switching by spin transfer will be used in the next generation of MRAM and will bring great advantages in an applied field—precession of the moment in the microwave frequency range.

The first evidence for spin transfer was indicated by experiments of spin injection through point contacts by Tsoi et al.,[47] but a clear understanding came later from measurements[48,49] performed on pillar-shaped metallic trilayers (Figure 11 a). Figure 11 b–c shows examples of our experimental results in the low-field regime of irreversible switching, for a metallic pillar and for tunnel junctions with electrodes of the dilute ferromagnetic semiconductor $Ga_{1-x}Mn_xAs$. For metallic pillars or tunnel junctions with electrodes made of a ferromagnetic transition metal such as Co or Fe, the current density needed for switching is around 10^6–10^7 A cm^{-2}, which is still slightly too high for applications, and an important challenge is the reduction of this current density. The switching time has been measured by other groups and can be as short as 100 ps, which is very attractive for the switching of MRAM. For the tunnel junction in Figure 11 c, the switching current is only about

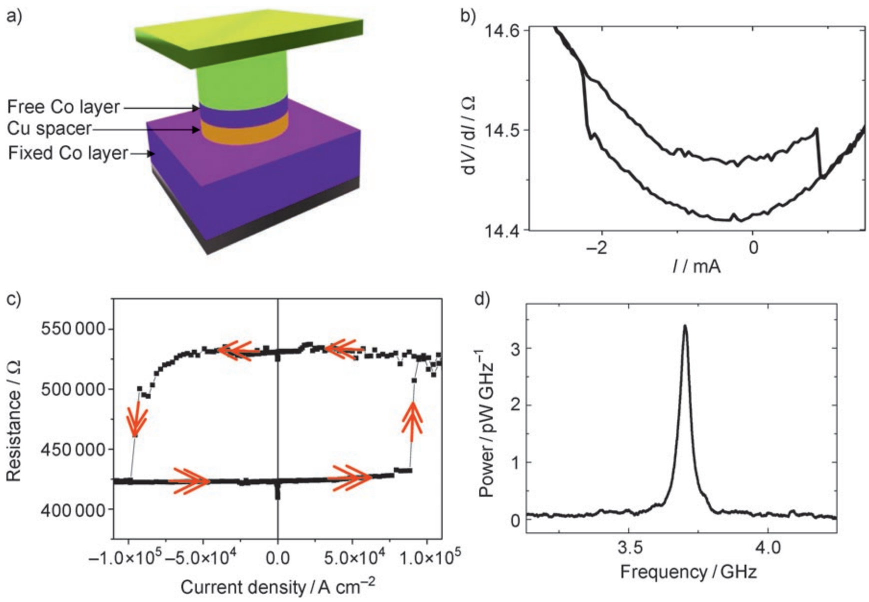

Figure 11. Experiments of magnetic switching and microwave generation induced by spin transfer from an electrical DC current in trilayered magnetic pillars. a) Schematic of a trilayered magnetic pillar. b) Switching by spin transfer between the parallel and antiparallel magnetic configurations of a Co/Cu/Co metallic pillar.[49] The switching between parallel and antiparallel orientations of the magnetizations of the two magnetic layers of the trilayer is detected by irreversible jumps of the resistance at a critical value of the current. The critical current density is of the order of 10^7 A cm^{-2}. c) Switching by spin transfer of a pillar-shaped tunnel junction composed of electrodes of the dilute ferromagnetic semiconductor GaMnAs separated by a tunnel barrier of InGaAs.[50] The critical current is about 100 times smaller than in the Py/Cu/Py pillar. Similar results have been obtained by Hayakawa et al.[51] d) Typical microwave power spectrum of a Co/Cu/Py pillar (Py = permalloy).[57]

terms of precise addressing and low energy consumption. The generation of oscillations in he microwave frequency range will lead to the design of spin transfer oscillators (STOs). One of the main interests of STOs is their agility, that is, the possibility of rapidly changing their frequency by tuning a DC current. They can also have a high quality factor. A disadvantage is the very small microwave power of an individual STO, metallic pillar, or tunnel junction. The solution is certainly the synchronization of a large number of STOs. The possibility of synchronization has already been demonstrated for two nanocontacts inducing spin-transfer excitations in the same magnetic layer.[58,59] In our laboratory we are exploring theoretically and experimentally a concept of self-synchronization of a collection of electrically connected STOs by the RF current components they induce.[60] Our recent experimental results seem to confirm the potential of this concept.

7. Spintronics with Semiconductors and Molecular Spintronics

Spintronics with semiconductors[61,62] is a very attractive concept as it can combine the potential of semiconductors (control of current by gate, coupling with optics, etc.) with the potential of the magnetic materials (control of current by spin manipulation, nonvolatility, etc.). It should be possible, for example, to combine storage, detection, logic, and communication capabilities on a single chip that could replace several components. New concepts of components have also been proposed, for example, the concept of spin field-effect transistors (spin FETs) based on spin transport in semiconductor lateral channels between spin-polarized source and drain electrodes with control of the spin transmission by a field-effect gate.[63] Some nonmagnetic semiconductors have a definite advantage over metals in terms of spin-coherence time and propagation of spin polarization over long distances.[61,62] However, as discussed below, the long-standing problems of the spin FET are still far from being solved.

Spintronics with semiconductors is currently developed along several roads:

a) by working on hybrid structures associating ferromagnetic metals with nonmagnetic semiconductors. As mentioned in the section on spin accumulation, Schmidt et al.[25] have raised the problem of "conductivity mismatch" to inject a spin-polarized current from a magnetic metal into a semiconductor. Solutions have been proposed by the theory[26,27] and one knows today that the injection/extraction of a spin-polarized current into/from a semiconductor can be achieved with a spin-dependent interface resistance, typically a tunnel junction. Spin injection/extraction through a tunnel contact has been now demonstrated in spin LEDs and magneto-optical experiments.[61–62,64]
b) by the fabrication of ferromagnetic semiconductors. The ferromagnetic semiconductor $Ga_{1-x}Mn_xAs$ (x: a few %) has been discovered[65] by the group of Ohno in Sendai in 1996, and, since this time, has revealed very interesting properties, such as the possibility of controlling the ferromagnetic properties with a gate voltage, and also large TMR and TAMR (tunneling anisotropic magnetoresistance) effects. However, its Curie temperature has reached only 170 K, which is well below room temperature and rules out most practical applications. Several room-temperature ferromagnetic semiconductors have been announced but the situation is not clear on this front yet.
c) through spin-polarized currents induced by spin–orbit effects, namely, spin Hall,[66–68] Rashba, or Dresselhaus effects. In the spin Hall effect (SHE), for example, spin–orbit interactions deflect the currents of the spin-up and spin-down channels in opposite transverse directions, thus inducing a transverse spin current, even in a nonmagnetic conductor. This could be used to create spin currents in structures composed of only nonmagnetic semiconductors. Actually, the SHE can be also found in nonmagnetic metals[69,70] and research is also very active in this field. May I mention that, already in the 1970s, I had found very large SHE induced by resonant scattering on spin–orbit split levels of nonmagnetic impurities such as Ir or Au in copper.[71]

Several groups have tried to probe the potential of spintronics with semiconductors by validating experimentally the concept of spin FET[63] described above. Both ferromagnetic metals and ferromagnetic semiconductors have been used for the source and the drain, but the results have been relatively poor. In a recent review article, Jonker and Flatté[61] note that a contrast larger than about 1% (i.e. $[R_{AP}-R_P]/R_P > 1\%$) has never been observed between the resistances of the parallel and antiparallel magnetic orientations of the source and the drain electrode, at least for lateral structures. We have recently proposed[24] that this can be understood in the models[27] I had developed with Henri Jaffrès to describe the spin transport between spin-polarized source and drain electrodes. In both the diffusive and ballistic regimes, a strong contrast between the conductances of the two configurations can be obtained only if the resistances of the interfaces between the semiconductor and the source or drain electrode are not only spin-dependent but also chosen in a relatively narrow window. The resistances must be larger than a first threshold value for spin injection/extraction from/into a metallic source/drain electrode, and smaller than a second threshold value to keep the carrier dwell time shorter than the spin lifetime. For vertical structures with a short distance between source and drain electrodes, the above conditions can be satisfied more easily and relatively large magnetoresistance can be observed, as illustrated in Figure 12. However, the results displayed in Figure 12c show that the magnetoresistance drops rapidly when the interface resistance exceeds some threshold value. This can be explained by the increase of the dwell time above the spin lifetime. Alternatively, the magnetoresistance also drops to zero when an increase of temperature shortens the spin lifetime and increases the ratio of the dwell time to the spin lifetime. For most experiments on lateral structures, it turns out that a part of the difficulties comes from too large interface resistances giving rise to too short dwell times. Min

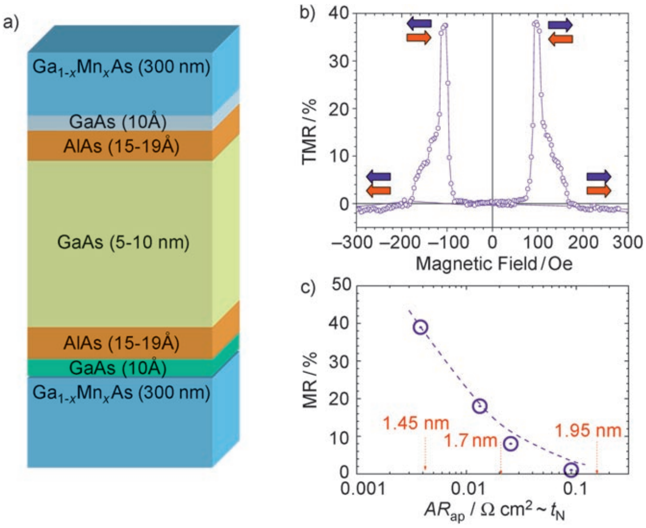

Figure 12. Experimental results[24,72] for spintronics with semiconductors. a) Structure of spintronics material composed of a GaAs layer separated from the GaMnAs source and drain electrodes by tunnel barriers of AlAs. b) MR curve at 4.2 K showing a resistance difference of 40% between the parallel and antiparallel magnetic configurations of the source and the drain electrodes. c) MR ratio as a function of the resistance of the tunnel barriers. AR_{ap} = Specific resistance for antiparallel magnetic configuration.

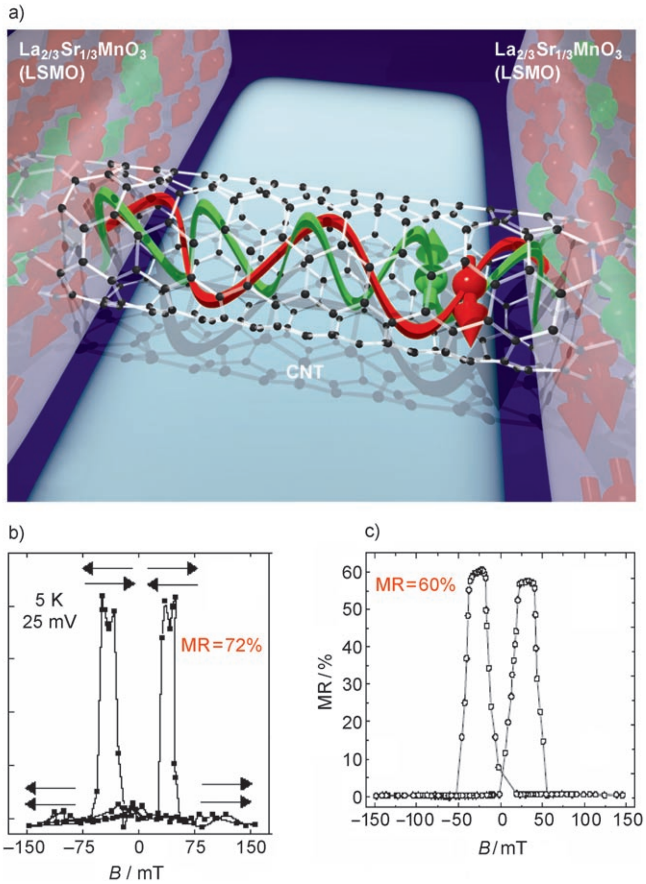

Figure 13. Spintronics with molecules. a) Principle of spin transport through a carbon nanotube (CNT) between magnetic electrodes (illustration courtesy of T. Kontos). b,c) Experimental results[75] at 4.2 K for magnetoresistance of carbon nanotubes between electrodes made of the ferromagnetic metallic oxide $La_{2/3}Sr_{1/3}MnO_3$. A contrast of 72% and 60% is obtained between the resistances for the parallel (high field) and antiparallel (peaks) magnetic configurations of the source and drain electrodes.

et al.[73] arrived at similar conclusions for the particular case of silicon-based structures and propose interesting solutions to lower the interface resistances by tuning the work function of the source and drain electrodes.

A recently emerging direction is spintronics with molecules. Very large GMR- or TMR-like effects are predicted by theory, especially for carbon-based molecules in which a very long spin lifetime is expected from the very small spin–orbit coupling. Promising experimental results have been published during the last years on spin transport in carbon nanotubes.[74,75] In particular, my recent work[75] in collaboration with a group in Cambridge on carbon nanotubes between ferromagnetic source and drain electrodes made of the metallic manganite $L_{1/3}Sr_{1/3}MnO_3$ has shown that the relative difference between the resistances of the parallel and antiparallel configurations can exceed 60–70% (Figure 13), which is well above what can be obtained with semiconductor channels. This can be explained not only by the long spin lifetimes of the electrons in carbon nanotubes but also by their short dwell time related to their high Fermi velocity (a definite advantage on semiconductors). The research is currently very active in this field and, in particular, graphene-based devices are promising.

8. Conclusions

In less than twenty years, we have seen spintronics increasing considerably the capacity of our hard disks, extending the hard disk technology to mobile appliances such as cameras and portable multimedia players, entering the automotive industry and the bio-medical technology, and, with TMR and spin transfer, getting ready to enter the RAM of our computers or the microwave emitters of our cell phones. The researches of today on the spin-transfer phenomena, on multiferroic materials, on spintronics with semiconductors and molecular spintronics, open fascinating new fields and are also very promising of multiple applications. Another perspective, out of the scope of this lecture, should be the exploitation of the truly quantum mechanical nature of spin and the long spin coherence time in confined geometry for quantum computing in an even more revolutionary application. Spintronics should take an important place in the science and technology of our century.

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