

# THE ORIGIN, DEVELOPMENT AND FUTURE OF SPINTRONICS

Nobel Lecture, December 8, 2007

by

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## 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, magnetic recording for example, are using the spin but only through its macroscopic manifestation, the magnetization of a ferromagnet. This picture started to change in 1988 when the discovery<sup>1-2</sup> of the Giant Magneto-Resistance (GMR) of the magnetic multilayers opened the way to an efficient control of the motion of the electrons by acting on their spin through the orientation of a magnetization. This rapidly triggered the developments of a new field of research and technology, today called spintronics and, like the GMR, exploiting 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<sup>3</sup>, had been experimentally demonstrated and theoretically described in my Ph.D. thesis more than ten years before the discovery of 1988. The GMR was the first step on the road of the exploitation of this influence to control an electrical current. Its application to the read head of hard discs greatly contributed to the fast rise in the density of stored information and led to the extension of the hard disk technology to consumer's 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 axes like the phenomena of spin transfer, spintronics with semiconductors, molecular spintronics or single-electron spintronics.

## FROM SPIN DEPENDENT CONDUCTION IN FERROMAGNETS TO GIANT MAGNETORESISTANCE

GMR and spintronics take their roots in previous researches on the influence of the spin on the electrical conduction in ferromagnetic metals<sup>3-5</sup>. The spin

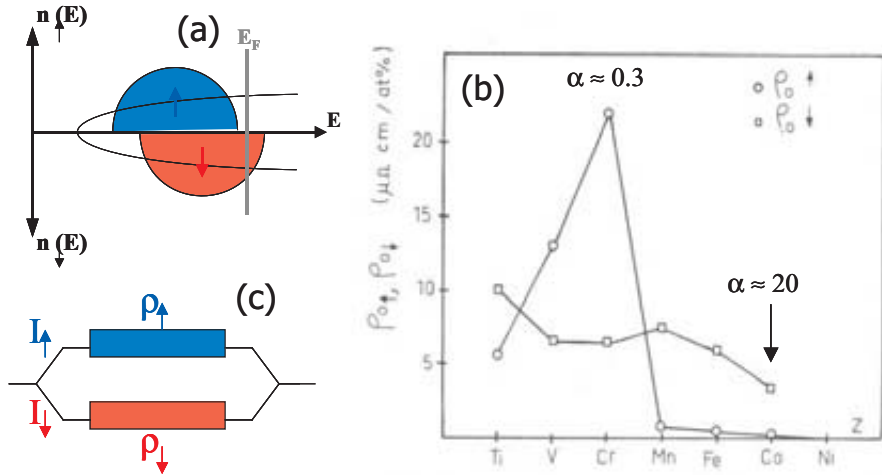


Figure 1. Basics of spintronics. (a) Schematic band structure of a ferromagnetic metal showing the energy band spin-splitting. (b) Resistivities of the spin up and spin down conduction channels for nickel doped with 1% of several types of impurity (measurements at 4.2 K)<sup>4</sup>. The ratio  $\alpha$  between the resistivities  $\rho_{0\downarrow}$  and  $\rho_{0\uparrow}$  of the spin  $\downarrow$  and spin  $\uparrow$  channels can be as large as 20 (Co impurities) or, as well, smaller than one (Cr or V impurities). (c) Schematic for spin dependent conduction through independent spin  $\downarrow$  and spin  $\uparrow$  channels in the limit of negligible spin mixing ( $\rho_{\uparrow\downarrow} = 0$  in the formalism of Ref.[4]).

dependence of the conduction can be understood from the typical band structure of a ferromagnetic metal shown in Fig.1a. The splitting between the energies of the “majority spin” and “minority spin” directions (spin up and spin down in the usual notation) makes 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<sup>3</sup> in 1936 to explain some features of the resistivity of ferromagnetic metals at the Curie temperature. However, in 1966, when I started my Ph.D. 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<sup>4</sup>. In Fig.1b, I show 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  $\alpha$  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, consistently 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, for example, extended to Co-based alloys by Loegel and Gautier<sup>5</sup> in Strasbourg.

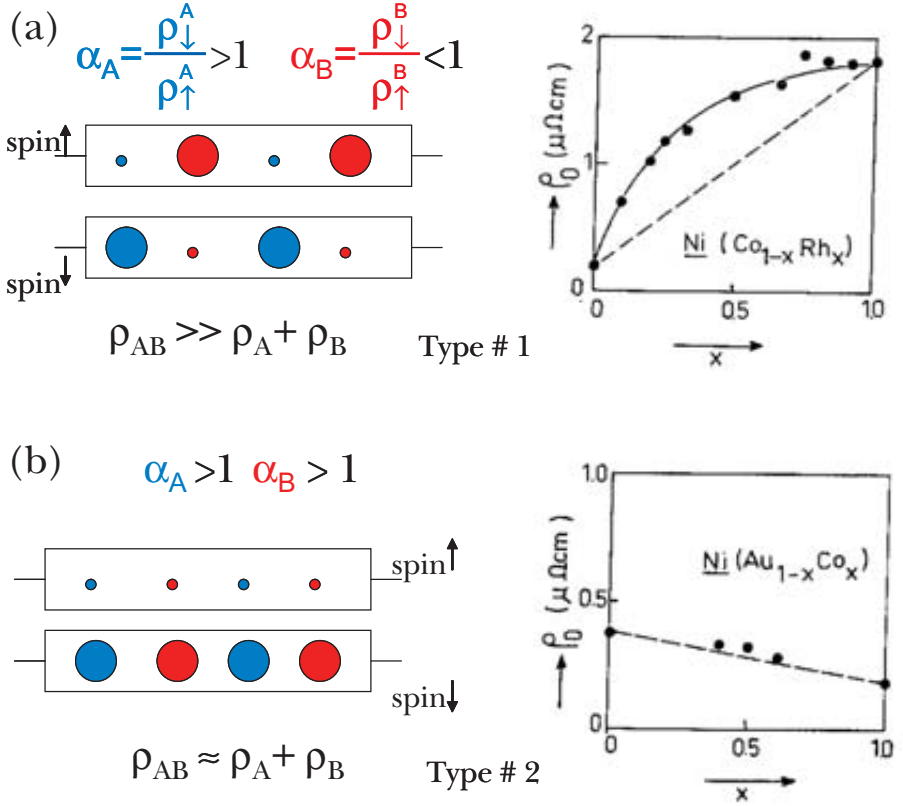


Figure 2. Experiments on ternary alloys based on the same concept as that of GMR<sup>4</sup>. (a) Schematic for the spin dependent conduction in alloys doped with impurities of opposite scattering spin asymmetries ( $\alpha_A = \rho_{A\downarrow}/\rho_{A\uparrow} > 1$ ,  $\alpha_B = \rho_{B\downarrow}/\rho_{B\uparrow} < 1$ , which leads to  $\rho_{AB} \gg \rho_A + \rho_B$ ) and experimental results for Ni(Co<sub>1-x</sub>Rh<sub>x</sub>) alloys. (b) Same for alloys doped with impurities of similar scattering spin asymmetries ( $\alpha_A = \rho_{A\downarrow}/\rho_{A\uparrow} > 1$ ,  $\alpha_B = \rho_{B\downarrow}/\rho_{B\uparrow} > 1$ , which leads to  $\rho_{AB} \approx \rho_A + \rho_B$ ) and experimental results for Ni(Au<sub>1-x</sub>Co<sub>x</sub>) alloys. In GMR the impurities A and B are replaced by multilayers, the situation of a (b) corresponding to the antiparallel (parallel) magnetic configurations of adjacent magnetic layers.

In my thesis, I also worked out the so-called two current model<sup>4</sup> for the conduction in ferromagnetic metals. This model is based on a picture of spin up and spin down currents coupled by spin mixing, i.e. 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 above room temperature in most ferromagnetic metals. The two-current model is the basis of spintronics today, but, surprisingly, the interpretation of the spintronics phenomena is generally based on a simplified version of the model neglecting spin mixing and assuming that the conduction is by two independent channels in parallel, as illustrated by Fig. 1c. 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<sup>6</sup>).

As a matter of fact, some experiments of my thesis with metals doped with two types of impurities<sup>4</sup> were already anticipating the GMR. This is illustrated by Fig. 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), that I call of 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 the impurities A and B of the ternary alloy by layers A and B in a multilayer, the antiparallel magnetic configuration of the layers A and B corresponding to the situation of an alloy of type #1, while the configuration with a parallel configuration corresponds to type #2. This brings 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 nm. Unfortunately, in the seventies, it was not technically possible to make multilayers with layers as thin as a few nm. I put some of my ideas in the fridge and, in my team at the Laboratoire de Physique des Solides of the Université Paris-Sud, from the beginning of the seventies to 1985, I worked on other topics like the extraordinary Hall effect, the spin Hall effect, the magnetism of spin glasses and amorphous materials.

In the mid-eighties, with the development of techniques like the 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 on ternary alloys to multilayers. In addition, in 1986, I saw the beautiful Brillouin scattering experiments of Peter Grünberg and coworkers<sup>7</sup> 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 Ph.D. 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<sup>1</sup> of very large magnetoresistance effects that we called GMR (Fig. 3a). Effects of the same type in Fe/Cr/Fe trilayers were obtained practically at the same time by Peter Grünberg at Jülich<sup>2</sup> (Fig. 3b). The interpretation of the GMR is similar to that described above for the ternary alloys and is illustrated by Fig. 3c. The first classical model of the GMR was published in 1989 by Camley and Barnas<sup>8</sup> and I collaborated with Levy and Zhang for the first quantum model<sup>9</sup> in 1991.

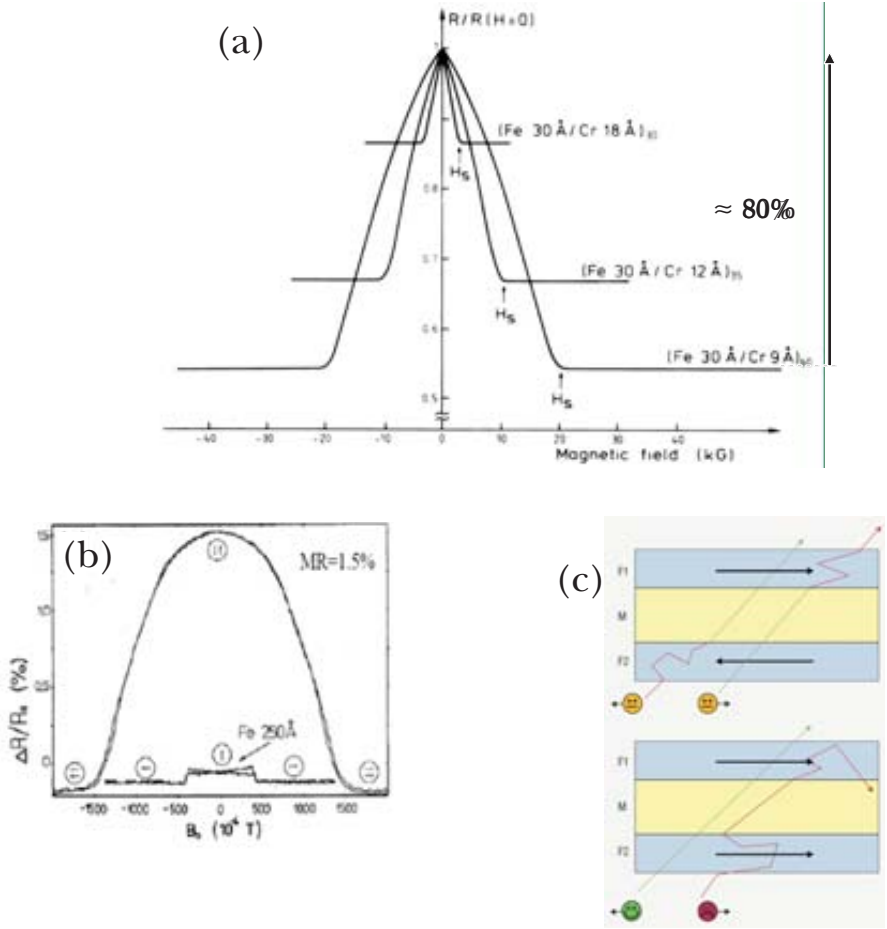


Figure 3. First observations of giant magnetoresistance. (a) Fe/Cr(001) multilayers<sup>1</sup> (with the current definition of the magnetoresistance ratio,  $\text{MR} = 100(R_{\text{AP}} - R_{\text{P}})/R_{\text{P}}$ ,  $\text{MR} = 80\%$  for the (Fe 3nm/Cr 0.9nm) multilayer). (b) Fe/Cr/Fe trilayers<sup>2</sup>. (c) Schematic of the mechanism of the 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 every second magnetic layer and the resistance is high (figure from Ref.[18]).

I am often asked if I was expecting such large MR effects. My answer is yes and no: on 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 scatterings cancelling 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).

## THE GOLDEN AGE OF GMR

Rapidly, our papers reporting the discovery of GMR 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 Agn  s Barth  l  my and Fr  d  ric 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.*<sup>10</sup> 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 behaviour of the GMR due to the oscillations of the interlayer exchange as a function of the thickness of the nonmagnetic layers. Also in 1990 Shinjo and Yamamoto<sup>11</sup>, as well as Dupas *et al.*<sup>12</sup>, demonstrated that GMR effects can be found in multilayers without antiferromagnetic interlayer coupling but composed of magnetic layers of different coercivities. Another important re-

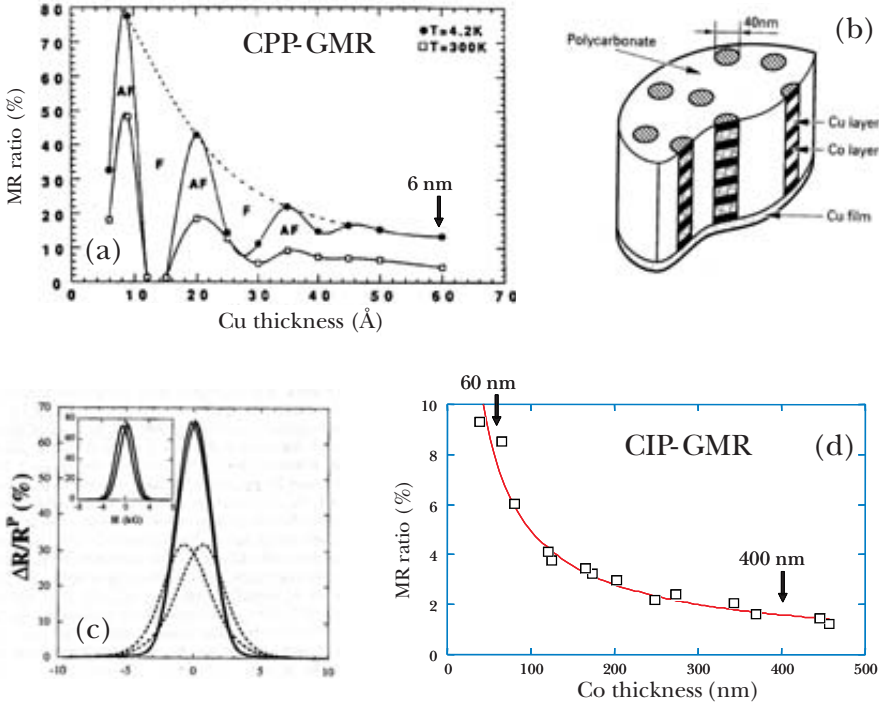


Figure 4. (a) Variation of the GMR ratio of Co/Cu multilayers in the conventional Current In Plane (CIP) geometry as a function of the thickness of the Cu layers<sup>13</sup>. The scaling length of the variation is the mean free path (short). (b) Structure of multilayered nanowires used for CPP-GMR measurements<sup>21</sup>. (c) CPP-GMR curves for (Permalloy 12 nm/Copper 4 nm) multilayered nanowires (solid lines) and (Cobalt 10 nm/Copper 5nm) multilayered nanowires (dotted lines)<sup>21</sup>. (d) ) Variation of the CPP-GMR ratio of Co/Cu multilayered nanowires as a function of the thickness of the Co layers<sup>21</sup>. The scaling length of the variation is the spin diffusion length (long).

sult, in 1991, was the observation of large and oscillatory GMR effects in Co/Cu, which became the archetypical GMR system (Fig. 4a). The first observations<sup>13</sup> were obtained in my group by my Ph. D. student Dante Mosca with multilayers prepared by sputtering at Michigan State University and at about the same time in the group of Stuart Parkin at IBM<sup>14</sup>. Also in 1991, Dieny *et al.*<sup>15</sup> reported the first observation of GMR in spin-valves, i.e. trilayered structures based on a concept of my co-laureate Peter Grünberg<sup>16</sup> 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.

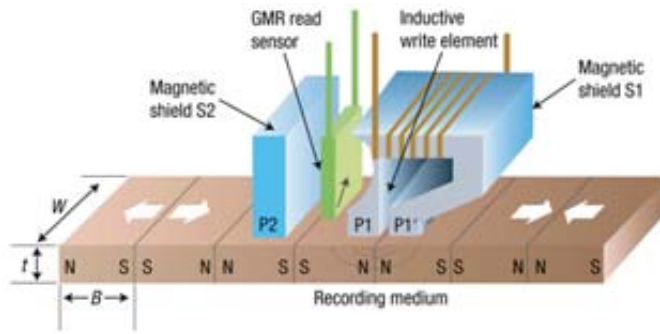


Figure 5. GMR head for hard-disc. Figure from Chappert *et al.*<sup>18</sup>.

Other developments of the research on magnetic multilayers and GMR at the beginning of the seventies are described in the Nobel lecture of my co-laureate Peter Grünberg, with, in particular, a presentation of the various devices based on the GMR of spin valve structures<sup>17-18</sup>. In the read heads (Fig.5) of the Hard Disc Drives (HDDs), the GMR sensors based on spin-valves have replaced the AMR (Anisotropic Magnetoresistance) sensors in 1997. The GMR, by providing a sensitive and scalable read technique, has led to an increase of the areal recording density by more than two orders of magnitude (from  $\approx 1$  to  $\approx 600$  Gbit/in<sup>2</sup> 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 .85-inch disk diameter) for mobile appliances like ultra-light laptops or portable multimedia players. GMR sensors are also used in many other types of application, mainly in automotive industry and bio-medical technology<sup>19</sup>.

## CPP-GMR AND SPIN ACCUMULATION PHYSICS

During the first years of the research on GMR, the experiments were performed only with currents flowing along the layer planes, in the geometry we call CIP (Current In Plane). It is only in 1993 that experiments of CPP-



GMR begun to be performed, that is experiments of GMR with the Current Perpendicular to the layer Planes. This was done first by sandwiching a magnetic multilayer between superconducting electrodes by Bass, Pratt and Shroeder at Michigan State University<sup>20</sup>, and, a couple of years after, in a collaboration of my group with Luc Piraux at the University of Louvain, by electrodepositing the multilayer into the pores of a polycarbonate membrane<sup>21</sup> (Fig. 4b-d). In the CPP-geometry, the GMR is not only definitely higher than in CIP (the CPP-GMR will be probably used in a future generation of read heads for hard discs), but also subsists in multilayers with relatively thick layers, up to the micron range<sup>21</sup>, as it can be seen in Fig. 4c-d. In a theoretical paper with Thierry Valet<sup>22</sup>, 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, the CPP-GMR has revealed the spin accumulation effects which 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 micron 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 Fig. 6. Far from the interface on the magnetic side, the current is larger in one of the spin channels (spin up on the figure), while, far from the interface on the other side, it is equally distributed in the two channels. With the current direction and the spin polarization of the figure, there is accumulation of spin up electrons (and depletion of spin down for charge neutrality) around the interface, or, in other word, a splitting between the Fermi energies (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 which 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, i.e. 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 a spin accumulation in the opposite direction progressively polarizes the current in



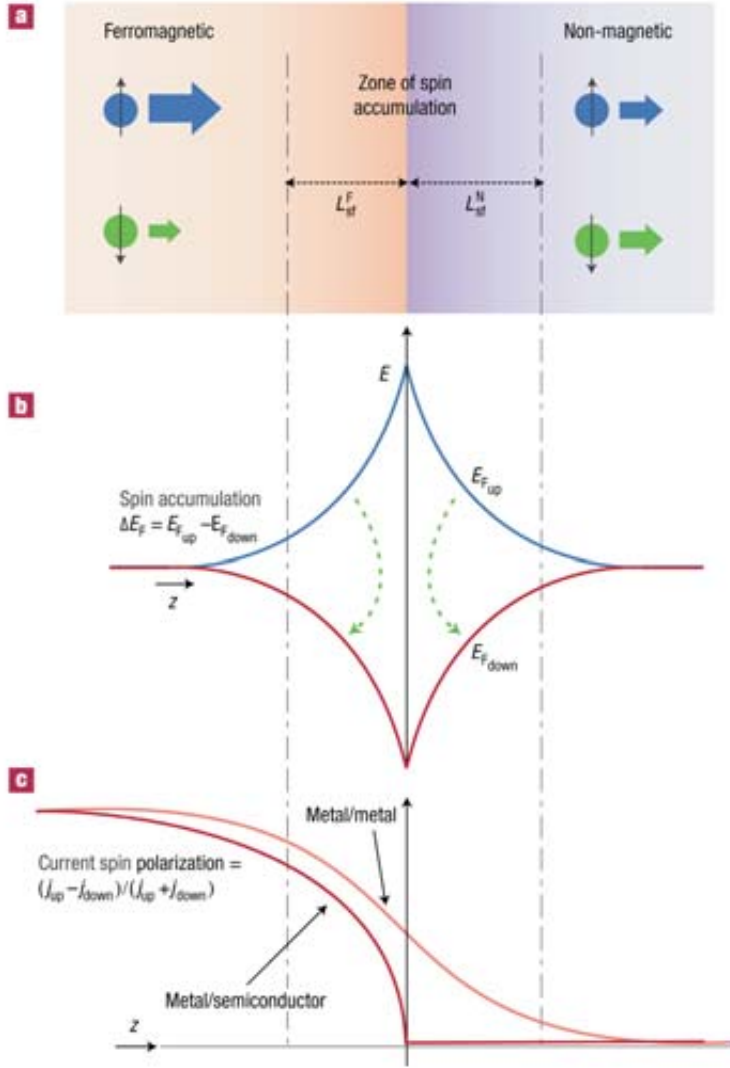


Figure 6. Schematic representation of the spin accumulation at an interface between a ferromagnetic metal and a non magnetic layer. (a) Spin-up and spin-down currents far from an interface between ferromagnetic and nonmagnetic conductors (outside the spin-accumulation zone). (b) Splitting of the chemical 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). Figures from Ref. [18].

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<sup>22</sup> in which the electrical potential is replaced by a spin and position dependent electro-chemical potential. These equations can be applied not only to the simple case of a single interface but 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<sup>23-24</sup>.

The physics of spin accumulation plays 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 DOS in the metal makes that similar spin accumulation splittings on the two sides of the interface, as in Fig. 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 the semiconductor, as shown in Fig. 6c. This problem has been first raised by Schmidt and coworkers<sup>25</sup>. 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)<sup>26-27</sup>. Spin injection through a tunnel barrier has now been achieved successfully in several experiments but the tunnel resistances are generally too large for an efficient transformation of the spin information into an electrical signal<sup>24</sup>.

## MAGNETIC TUNNEL JUNCTIONS AND TUNNELLING MAGNETO-RESISTANCE (TMR)

An important stage in the development of spintronics has been the research on the Tunnelling Magnetoresistance (TMR) of the Magnetic Tunnel Junctions (MTJ). The MTJ 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 been already reported by Jullière<sup>28</sup> in 1975, but they were not easily reproducible and actually could not be really reproduced during 20 years. It is only in 1995 that large ( $\approx 20\%$ ) and reproducible effects were obtained by Moodera's and Miyasaki's groups on MTJ with a tunnel barrier of amorphous alumina<sup>29-30</sup>. From a technological point of view, the interest of the MTJ with respect to the metallic spin valves comes from the vertical direction of the current and from the resulting possibility of a reduction of the lateral size to a submicronic scale by lithographic

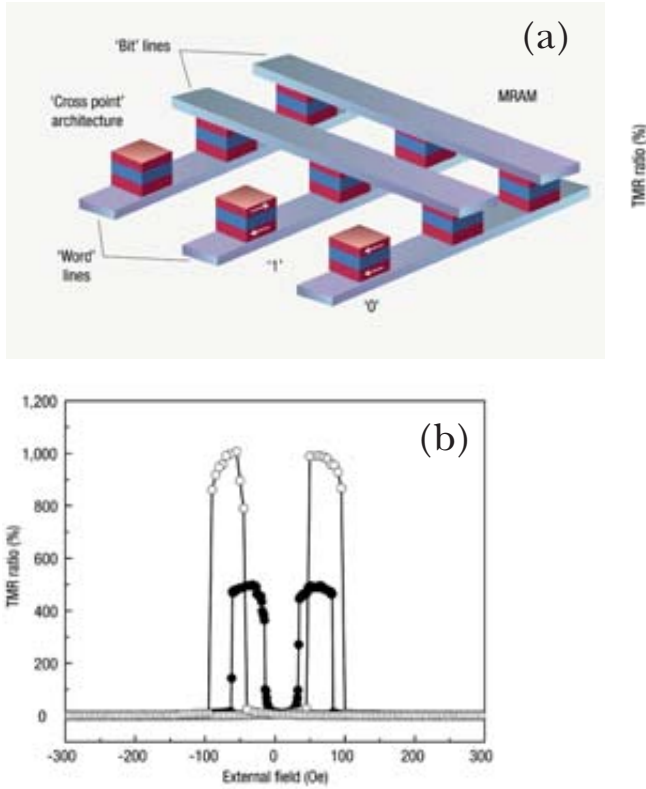


Figure 7. (a) Principle of the magnetic random access memory MRAM in the basic “cross point” architecture. The binary information “0” and “1” is recorded on the two opposite orientations of the magnetization of the free layer of magnetic tunnel junctions (MTJ), which are connected to the crossing points 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 the resulting magnetic field is high enough to orient the magnetization of the free layer. For reading, one measures the resistance between the two lines connecting the addressed cell. Schematic from Ref.[18]. (b) High magnetoresistance,  $TMR = (R_{max} - R_{min}) / R_{min}$ , measured by Lee *et al.*<sup>34</sup> for the magnetic stack:  $(Co_{25}Fe_{75})_{80}B_{20}(4nm)/MgO(2.1nm)/(Co_{25}Fe_{75})_{80}B_{20}(4.3nm)$  annealed at 475°C after growth, measured at room temperature (black circles) and low temperature (open circles).

techniques. The MTJ are at the basis of a new concept of magnetic memory called MRAM (Magnetic Random Access Memory) and schematically represented in Fig. 7a. The MRAMs are expected to combine the short access time of the semiconductor-based RAMs and the non-volatile character of the magnetic memories. In the first MRAMs, put onto 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, see Fig. 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.

The research on the TMR has been very active since 1995 and the most important step was the recent transition from MTJ with amorphous tunnel barrier (alumina) to single crystal MTJ and especially MTJ with MgO barrier. In the CNRS/Thales laboratory we founded in 1995, the research on TMR was one of our main projects and, in collaboration with a Spanish group, we obtained one of the very first TMA results<sup>31</sup> on MTJ with epitaxial MgO. However our TMR was only slightly larger than that found with alumina barriers and similar electrodes. The important breakthrough came in 2004 at Tsukuba<sup>32</sup> and IBM<sup>33</sup> where it was found that very large TMR ratios, up to 200% at room temperature, could be obtained from MgO MTJ of very high structural quality. TMR ratios of about 600% have been now reached<sup>34</sup> (Fig. 7b). In such MTJ, the single crystal barrier filters the symmetry of the wave functions of the tunnelling electrons<sup>35-37</sup>, 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 tunnelling waves with a single crystal barrier is a very good illustration of what is under the word “spin polarization” in a spintronic experiment. In the example of Fig. 8, taken from an article by Zhang and Butler<sup>37</sup>, one sees the density of states of evanescent wave functions of different symmetries,  $\Delta_1$ ,  $\Delta_5$ , 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  $\Delta_1$  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  $\Delta_1$  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  $\Delta_1$  selected by MgO(001). For example, a SrTiO<sub>3</sub> barrier predominantly selects evanescent wave functions of  $\Delta_5$  symmetry, which are connected to minority spin states of cobalt<sup>38</sup>. This explains the negative effective spin-polarization of cobalt we had observed in SrTiO<sub>3</sub>-based MTJ<sup>39</sup>. This finally shows that there 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 Fig. 1b.

There are other promising directions to obtain large TMR 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 which were predicted to be half-metallic, i.e. metallic for one of the spin direction and insulating for the other one, in other words 100% spin-polarized. Very high spin polarization (95%) and record TMR (1800%) have been obtained by our Ph.D. student Martin Bowen with La<sub>2/3</sub>Sr<sub>1/3</sub>MnO<sub>3</sub> electrodes<sup>40</sup> but the Curie temperature of this manganite (around 350K) is

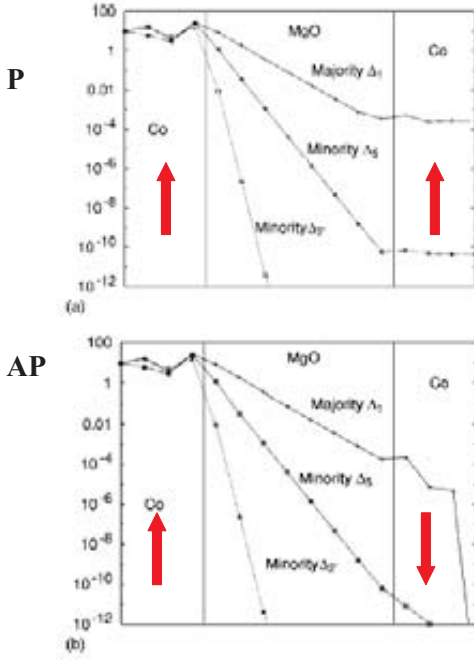


Figure 8. Physics of TMR illustrated by the decay of evanescent electronic waves of different symmetries in a MgO(001) layer between cobalt electrodes calculated by Zhang and Butler<sup>37</sup>. The  $\Delta_1$  symmetry of the slowly decaying tunnelling channel is well represented at the Fermi level 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 tunnelling between the electrodes exists only for the parallel magnetic configuration when a  $\Delta_1$  channel can be connected to both electrodes (above). In the antiparallel configuration (below), both the spin up and spin down  $\Delta_1$  channels are poorly connected on one of the sides. This explains the very high TMR of this type of junction.

too low for applications. It now turns out from recent results in Japan<sup>41</sup> that ferromagnets of the family of the Heusler alloys also present very large TMR ratios with still 90% at room temperature<sup>41</sup>. Another interesting concept that we are exploring is spin filtering by tunnelling through a ferromagnetic insulator layer<sup>42-43</sup>. This can be described as the tunnelling 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<sup>42</sup> at the MIT and at Eindhoven. Promising results with insulating ferromagnets of much higher Curie temperature have been recently obtained, see, for example Ref. [43]. Some of the magnetic barriers we have recently tested in MTJ are also ferroelectric, so that the MTJ 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<sup>44</sup>, as shown in Fig. 9.

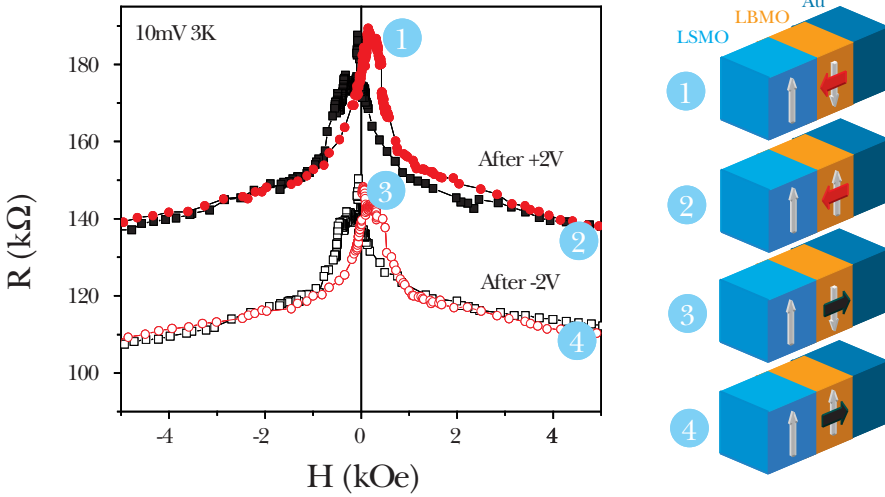


Figure 9. Four state resistance of a tunnel junction composed of a biferroic tunnel barrier ( $\text{La}_{0.1}\text{Bi}_{0.9}\text{MnO}_3$ ) between a ferromagnetic electrode of  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$  and a nonmagnetic electrode of gold. The states 1-4 correspond to the magnetic (white arrows) and electric (black arrows) polarizations represented on the right of the figure. From Gajek *et al.*<sup>44</sup>.

## 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 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 has been introduced by John Slonczewski<sup>45</sup> and appears also in papers of Berger<sup>46</sup>, is illustrated in Fig. 10. As described in the caption of the figure, 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 an irreversible switching of this magnetic moment or, in a second regime, generally in the presence of an applied field, it generates precessions of the moment in the microwave frequency range.

The first evidence that spin transfer can work was indicated by experiments of spin injection through point contacts by Tsoi *et al.*<sup>47</sup> but a clear understanding came later from measurements<sup>48-49</sup> performed on pillar-shaped metallic trilayers (Fig. 11a). In Fig. 11b-c, I present examples of our experimental results in the low field regime of irreversible switching, for a metallic pillar and for a tunnel junctions with electrodes of the dilute ferromagnetic semiconductor  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ . For metallic pillars or tunnel junctions with electrodes made of a ferromagnetic transition metal like Co or Fe, the current density needed for switching is around  $10^6\text{-}10^7\text{ Amp/cm}^2$ , 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 in other groups and

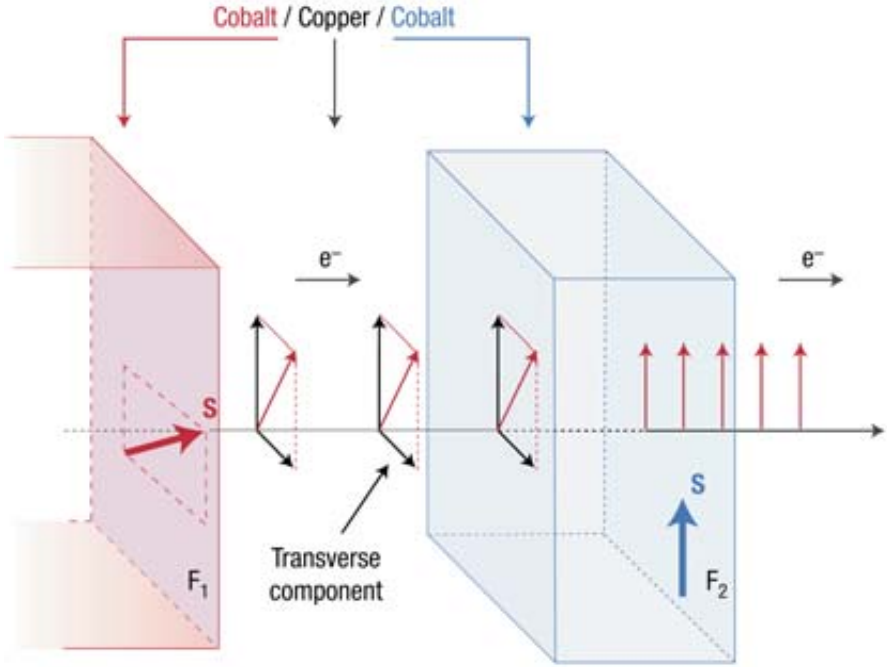


Figure 10. Illustration of the spin transfer concept introduced by John Slonczewski<sup>45</sup> in 1996. A spin-polarized current is prepared by a first magnetic layer  $F_1$  with an obliquely oriented spin-polarization with respect to the magnetization axis of a second layer  $F_2$ . When this current 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 has been 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 Ref.[18].

can be as short as 100 ps, which is very attractive for the switching of MRAM. For the tunnel junction of Fig. 11c, the switching current is only about  $10^5$  Amp./cm<sup>2</sup> and 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<sup>52</sup>. 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 various theoretical problems. The determination of the spin transfer torque is related to the solution of



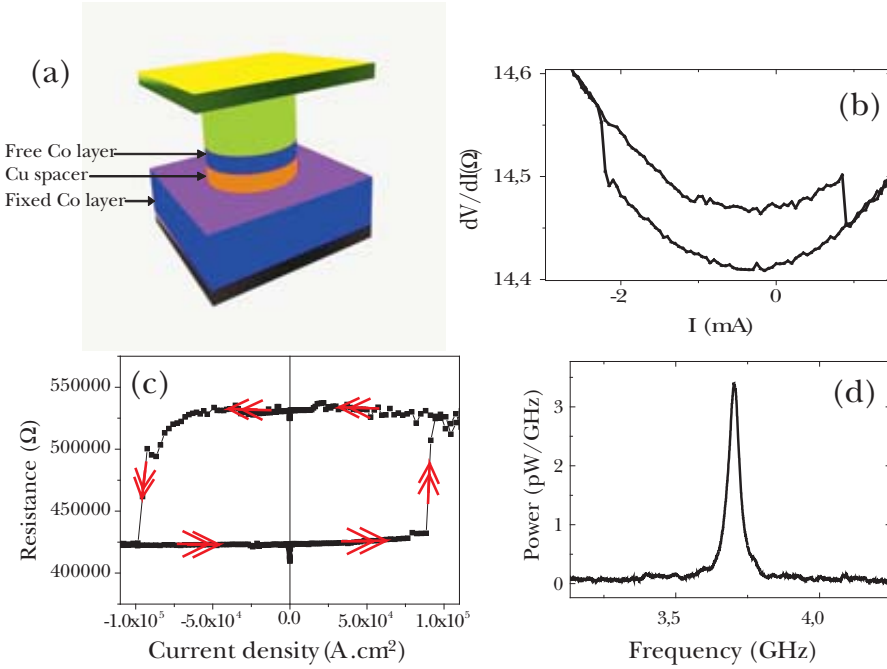


Figure 11. Experiments of magnetic switching and microwave generation induced by spin transfer from an electrical DC current in trilayered magnetic pillars. (a) Schematic of a trilayered magnetic pillar. (b) Switching by spin transfer between the parallel and antiparallel magnetic configurations of a Co/Cu/Co metallic pillar<sup>49</sup>. 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<sup>2</sup>. (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<sup>50</sup>. The critical current is about hundred times smaller than in the Py/Cu/Py pillar. Similar results have been obtained by Hayakawa *et al.*<sup>51</sup>. (d) Typical microwave power spectrum of a Co/Cu/Py pillar (Py =permalloy)<sup>57</sup>.

spin transport equations<sup>53-56</sup>, while the description of the switching or precession of the magnetization raises problems of non-linear dynamics<sup>53</sup>. All these problems are interacting and, for example, some of our recent results show that it is possible to obtain very different dynamics (with, for applications, the interest of oscillations without applied field) by introducing strongly different spin relaxation times in the two magnetic layers of a trilayer to distort the angular dependence of the torque<sup>57</sup>.

The spin transfer phenomena will have certainly important applications. Switching by spin transfer will be used in the next generation of MRAM and will bring great advantages in terms of precise addressing and low energy consumption. The generation of oscillations in the microwave frequency range will lead to the design of Spin Transfer Oscillators (STOs). One of the main interests of the STOs is their agility, that is the possibility of changing rapidly their frequency by tuning a DC current. They can also have a high

quality factor. Their 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 been already demonstrated for two nano-contacts inducing spin transfer excitations in the same magnetic layer<sup>58-59</sup>. 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<sup>60</sup>.

## SPINTRONICS WITH SEMICONDUCTORS AND MOLECULAR SPINTRONICS

Spintronics with semiconductors<sup>61-62</sup> is very attractive 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, non-volatility, etc.). It should be possible, for example, to gather 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 sources and drains with control of the spin transmission by a field effect gate<sup>63</sup>. Some nonmagnetic semiconductors have a definite advantage on metals in terms of spin-coherence time and propagation of spin polarization on long distances<sup>61-62</sup>. However, as it will be discussed below, the long standing problem of the Spin FET is still far from being solved.

Spintronics with semiconductors is currently developed along several roads.

i) The first road is by working on hybrid structures associating ferromagnetic metals with nonmagnetic semiconductors. As this has been mentioned in the Section on spin accumulation, Schmidt *et al.*<sup>25</sup> 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<sup>26-27</sup> 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<sup>61-62,64</sup>.

ii) Another road for spintronics with semiconductors is based on the fabrication of ferromagnetic semiconductors. The ferromagnetic semiconductor  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  ( $x \approx$  a few %) has been discovered<sup>65</sup> by the group of Ohno in Sendai in 1996, and, since this time, has revealed very interesting properties, namely the possibility of controlling the ferromagnetic properties with a gate voltage, and also large TMR and TAMR (Tunnelling Anisotropic Magnetoresistance) effects. However its Curie temperature has reached only 170 K, well below room temperature, which rules out most practical applica-

tions. Several room temperature ferromagnetic semiconductors have been announced but the situation is not clear on this front yet.

iii) The research is now very active on a third road exploiting spin-polarized currents induced by spin-orbit effects, namely the Spin Hall<sup>66-68</sup>, 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 conductors. Actually the SHE can be also found in nonmagnetic metals<sup>69-70</sup> and the research is also very active in this field. May I mention that, already in the seventies, I had found very large SHE induced by resonant scattering on spin-orbit-split levels of non-magnetic impurities in copper<sup>71</sup>.

Several groups have tried to probe the potential of spintronics with semiconductors by validating experimentally the concept of Spin FET<sup>63</sup> 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<sup>61</sup> 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, at least for lateral structures. We have recently proposed<sup>24</sup> this can be understood in the models<sup>27</sup> I had developed with Henri Jaffrès to describe the spin transport between spin-polarized sources and drains. 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 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, 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, the above conditions can be satisfied more easily and relatively large magnetoresistance can be observed, as illustrated by the results I present in Fig. 12. However the results displayed in Fig. 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 *et al.*<sup>73</sup> have 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 the drain.

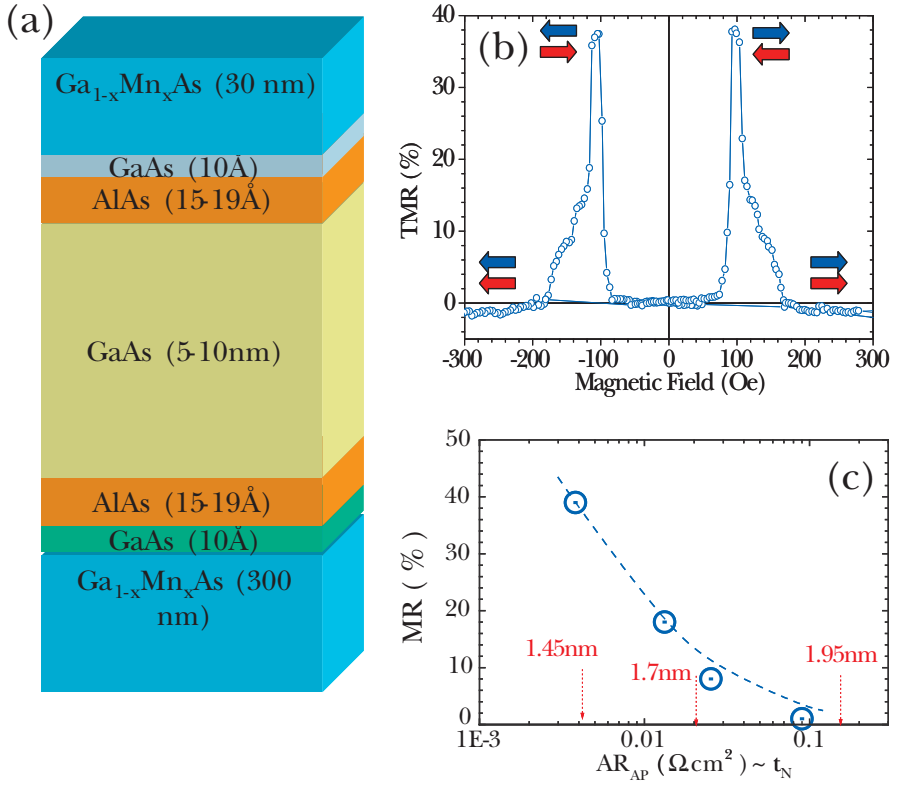


Figure 12. Spintronics with semiconductors illustrated by experimental results<sup>24,72</sup> on the structure represented on the right and composed of a GaAs layer separated from the GaMnAs source and drain by tunnel barriers of AlAs. (a) MR curve at 4.2 K showing a resistance difference of 40 % between the parallel and antiparallel magnetic configurations of the source and the drain. (b) MR ratio as a function of the resistance of the tunnel barriers.

A recently emerging direction is spintronics with molecules. Very large GMR- or TMR-like effects are predicted by the theory, especially for carbon-based molecules in which a very long spin lifetime is expected from the small spin-orbit coupling. Promising experimental results have been published during the last years on spin transport in carbon nanotubes<sup>74-75</sup>. In particular, my recent work<sup>75</sup> in collaboration with a group in Cambridge on carbon nanotubes between ferromagnetic source and drain 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%, 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.

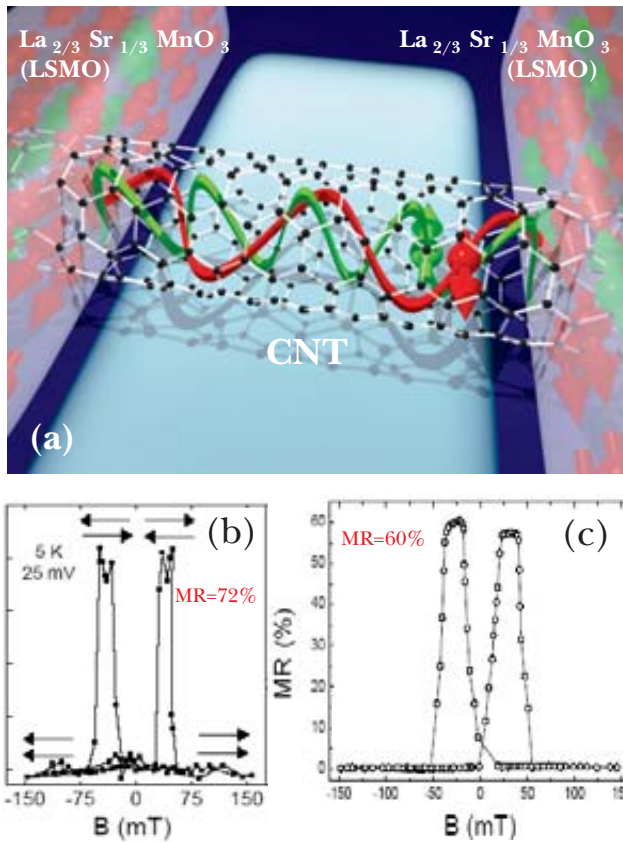


Figure 13. Spintronics with molecules illustrated by, (a): Artistic view of spin transport through a carbon nanotube between magnetic electrodes (courtesy of T. Kontos). (b) and (c): Magnetoresistance experimental results<sup>75</sup> at 4.2 K on carbon nanotubes between electrodes made of the ferromagnetic metallic oxide  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ . 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.

## CONCLUSION

In less than twenty years, we have seen spintronics increasing considerably the capacity of our hard discs, extending the hard disc technology to mobile appliances like cameras or 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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Portrait photo of Albert Fert by photographer Ulla Montan.