Nobel Lecture: Origin, development, and future of spintronics*

Albert Fert

Unité Mixte de Physique CNRS/Thales, 91767, Palaiseau, France and Université Paris-Sud, 91405, Orsay, France

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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 (Baibich *et al.*, 1988; Binash *et al.*, 1989) of the giant magnetoresistance (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 development 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 (1936), had been experimentally demonstrated and theoretically described in my Ph.D. thesis almost 20 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 heads of hard disks 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 expanding 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 from previous research on the influence of the spin on the electrical conduction in ferromagnetic metals (Mott, 1936; Fert and Campbell, 1968, 1971, 1976; Loegel and Gautier, 1971). The spin dependence of the conduction can be understood from the typical band structure of a ferromagnetic metal shown in Fig. 1(a). 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 (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 (Fert and Campbell, 1968, 1971, 1976). In Fig. 1(b), 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 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 1 for Cr or V impurities, consistent with the theoretical models developed by Jacques Friedel for the electronic structures of these impurities. The two-current conduction was rapidly confirmed by other groups and, for example, extended to Co-based alloys by Loegel and Gautier (1971) in Strasbourg.

In my thesis, I also worked out the so-called two-current model (Fert and Campbell, 1968, 1971, 1976) 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 momentum exchange between the two channels by 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 (the degree of equalization depends on the ratio between the "spin mixing resistivity" and the resistivity). 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 by two

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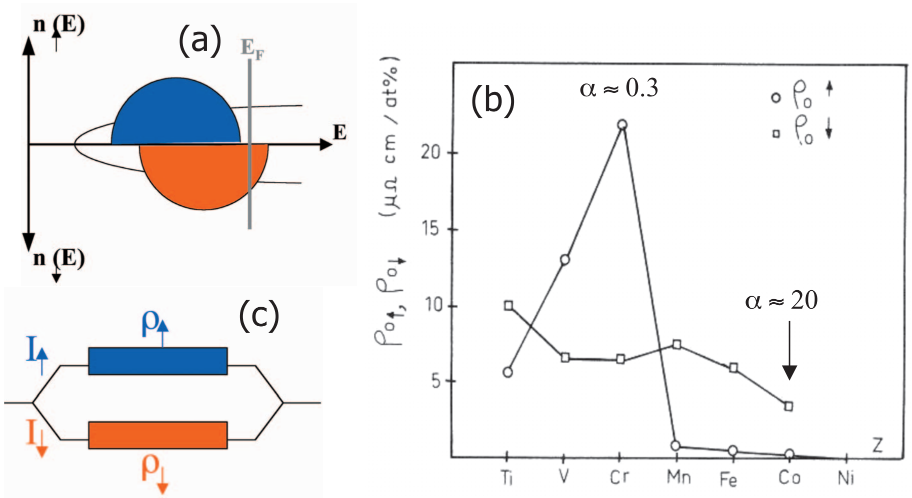

FIG. 1. (Color) Basics of spintronics. (a) Schematic band structure of a ferromagnetic metal showing the energy band spin splitting. (b) Resistivities of the spin up and spin down conduction channels for nickel doped with 1% of several types of impurities (measurements at 4.2 K) (Fert and Campbell, 1968, 1971, 1976). The ratio α between the resistivities $\rho_{0\downarrow}$ and $\rho_{0\uparrow}$ of the spin ↓ and spin ↑ channels can be as large as 20 (Co impurities) or, as well, smaller than 1 (Cr or V impurities). (c) Schematic for spin-dependent conduction through independent spin ↓ and spin ↑ channels in the limit of negligible spin mixing [$\rho_{\uparrow\downarrow}=0$ in the formalism of Fert and Campbell (1968, 1971, 1976)].

independent channels is parallel, as illustrated by Fig. 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 spin mixing, i.e., momentum exchange between the two channels by electron-magnon collisions (electron-electron scattering inside the electron system), should not be confused with the spin-lattice mechanism relaxing the spin accumulation (the relaxation of the spin accumulation to the lattice comes from spin-orbit without direct contribution from electron-magnon scattering).

As a matter of fact, some experiments of my thesis with metals doped with two types of impurities (Fert and Campbell, 1968, 1971, 1976) 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 type No. 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 No. 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 magnetic 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 No. 1, while the configuration with a parallel configuration corresponds to type No. 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,

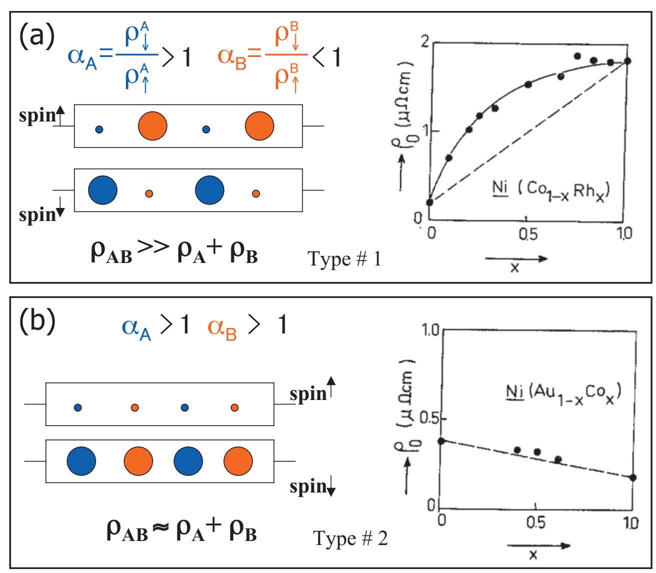

FIG. 2. (Color) Experiments on ternary alloys based on the same concept as that of the GMR (Fert and Campbell, 1968, 1971, 1976). (a) Schematic for the spin-dependent conduction in alloys doped with impurities of opposite scattering spin asymmetries ($\alpha_A=\rho_{A\downarrow}/\rho_{A\uparrow}>1$, $\alpha_B=\rho_{B\downarrow}/\rho_{B\uparrow}<1$, $\rho_{AB}\gg\rho_A+\rho_B$) and experimental results for $Ni(Co_{1-x}Rh_x)$ alloys of total concentration 1%. (b) Same for alloys doped with impurities of similar scattering spin asymmetries ($\alpha_A=\rho_{A\downarrow}/\rho_{A\uparrow}>1$, $\alpha_B=\rho_{B\downarrow}/\rho_{B\uparrow}>1$, $\rho_{AB}\approx\rho_A+\rho_B$) and experimental results for $Ni(Au_{1-x}Co_x)$ alloys of total concentration 1%. In GMR the impurities A and B are replaced by magnetic layers, the situation of (a) [(b)] corresponding to the antiparallel [parallel] magnetic configurations of adjacent magnetic layers.

that is, practically, if they are spaced by only a few nm. Unfortunately, in the 1970s, 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 1970s 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 1980s, with the development of techniques like 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 Grünberg *et al.* (1986) 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 (Baibich *et al.*, 1988) of very large magnetoresistance effects that we called GMR [Fig. 3(a)]. Effects of the same type in Fe/Cr/Fe trilayers were obtained practically at the same time by Grünberg at Jülich (Binash *et al.*, 1989) [Fig. 3(b)]. The interpretation of the GMR is similar to that described above for the ternary alloys and is illustrated by Fig. 3(c). The first classical model of the GMR was published by Camley and Barnas (1989) and I collaborated with Levy and Zhang for the first quantum model (Levy *et al.*, 1990) 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 scatterings canceling 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 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.* (1990) 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 due to the oscillations of the interlayer exchange as a function of the thickness of the nonmagnetic layers. Also in 1990 Shinjo and Yamamoto (1990), as well as Dupas *et al.* (1990), 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 [Fig. 4(a)]. The first observations (Mosca *et al.*, 1991) 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 (Parkin *et al.*, 1991). Also in 1991, Dieny *et al.* (1991) reported the first observation of GMR in spin valves, i.e., trilayered structures 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 based on the GMR of spin-valve structures (Parkin, 2002; Chappert *et al.*, 2007). In the read heads (Fig. 5) of the hard disk drives (HDDs), the GMR sensors based on spin valves have replaced the anisotropic magnetoresistance (AMR) 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 ≈ 1 to ≈ 600 Gbit/in.^{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-in. disk diameter) for mobile appliances like ultralight laptops or portable multimedia players. GMR sensors are also used in many other types of applications, mainly in the automotive industry and biomedical technology (Freitas *et al.*, 2003).

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 one calls CIP (current in plane). It is only in 1993 that experiments of CPP-GMR began to be performed, that is experiments of GMR with the current perpendicular to the layer

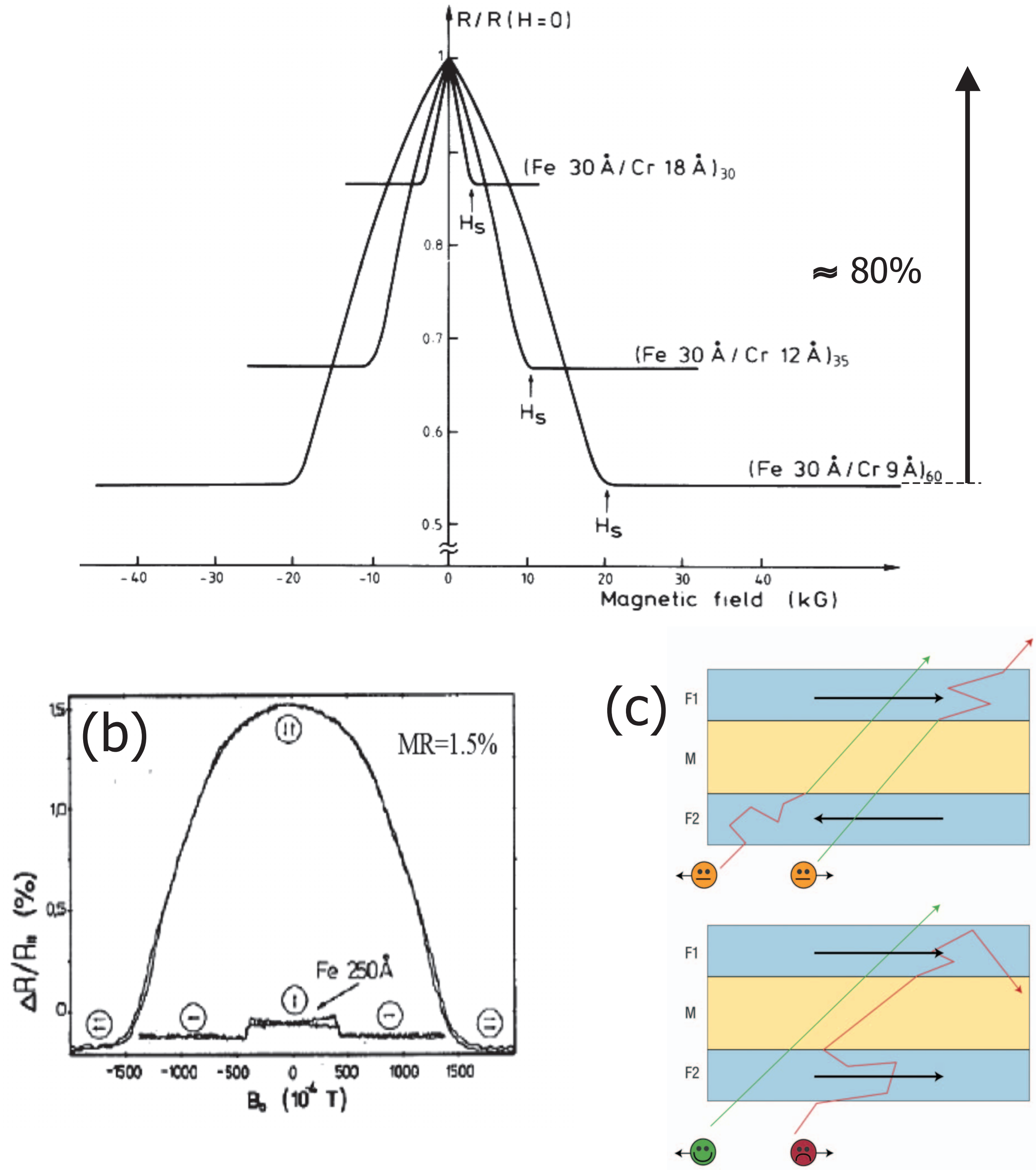

FIG. 3. (Color) First observations of giant magnetoresistance. (a) Fe/Cr(001) multilayers (Baibich *et al.*, 1988) [with the current definition of the magnetoresistance ratio $\mathrm{MR}=100(R_{\mathrm{AP}}-R_{\mathrm{P}})/R_{\mathrm{p}}$, MR=85% for the (Fe 3 nm/Cr 0.9 nm) multilayer]. (b) Fe/Cr/Fe trilayers (Binash *et al.*, 1989). (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 leads 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. From Chappert *et al.*, 2007.

planes. This was done first by sandwiching a magnetic multilayer between superconducting electrodes by Bass, Pratt, and Shroeder at Michigan State University (Pratt *et al.*, 1991; Bass and Pratt, 1999), 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 (Piraux *et al.*, 1994; Fert and Piraux, 1999) [Figs. 4(b)–4(d)]. In the CPP geometry, the GMR is not only definitely higher than in CIP (the CPP-GMR will probably be used in a future generation of read heads for hard disks), but also subsists in multilayers with relatively thick layers, up to the micron range (Piraux *et al.*, 1994; Fert and Piraux, 1999), as it can be seen in Figs. 4(c) and 4(d). In a theoretical paper with Thierry Valet (Valet and Fert, 1993), I showed that, owing to spin accumulation effects occurring in the CPP geometry, the length scale of the spin transport becomes the long spin

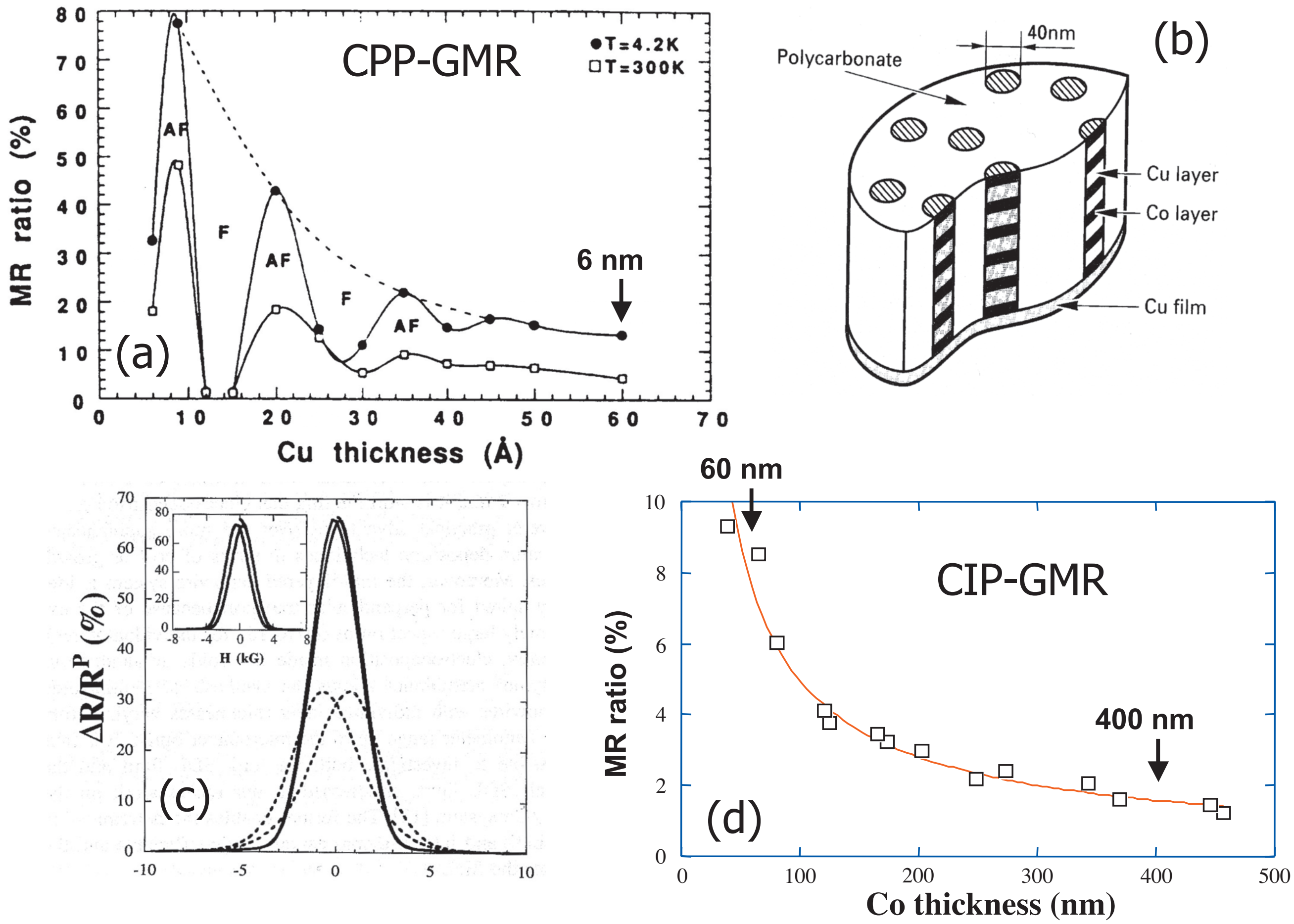

FIG. 4. (Color) (a) Variation of the GMR ratio of Co/Cu multilayers in the conventional current in plane (CIP) geometry as a function of the thickness of the Cu layers (Mosca *et al.*, 1991). The scaling length of the variation is the mean-free path (short). (b) Structure of multilayered nanowires used for CPP-GMR measurements. (c) CPP-GMR curves at 77 K for (Permalloy 12 nm/copper 4 nm) multilayered nanowires (solid lines) and (cobalt 10 nm/copper 5 nm) multilayered nanowires (dotted lines) (Piraux *et al.*, 1994; Fert and Piraux, 1999). (d) Variation of the CPP-GMR ratio of Co/Cu multilayered nanowires as a function of the thickness of the Co layers (Piraux *et al.*, 1994; Fert and Piraux, 1999). The scaling length of the variation is the spin diffusion length (long). The inset shows the curves for Permally/copper at 4.2 K.

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 plays 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 in 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 words, 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

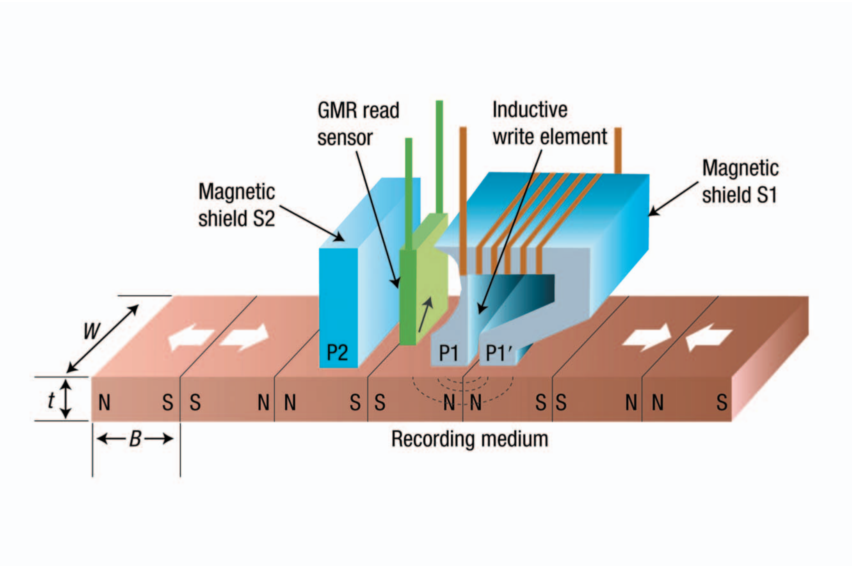

FIG. 5. (Color) GMR head for hard disk. From Chappert *et al.*, 2007.

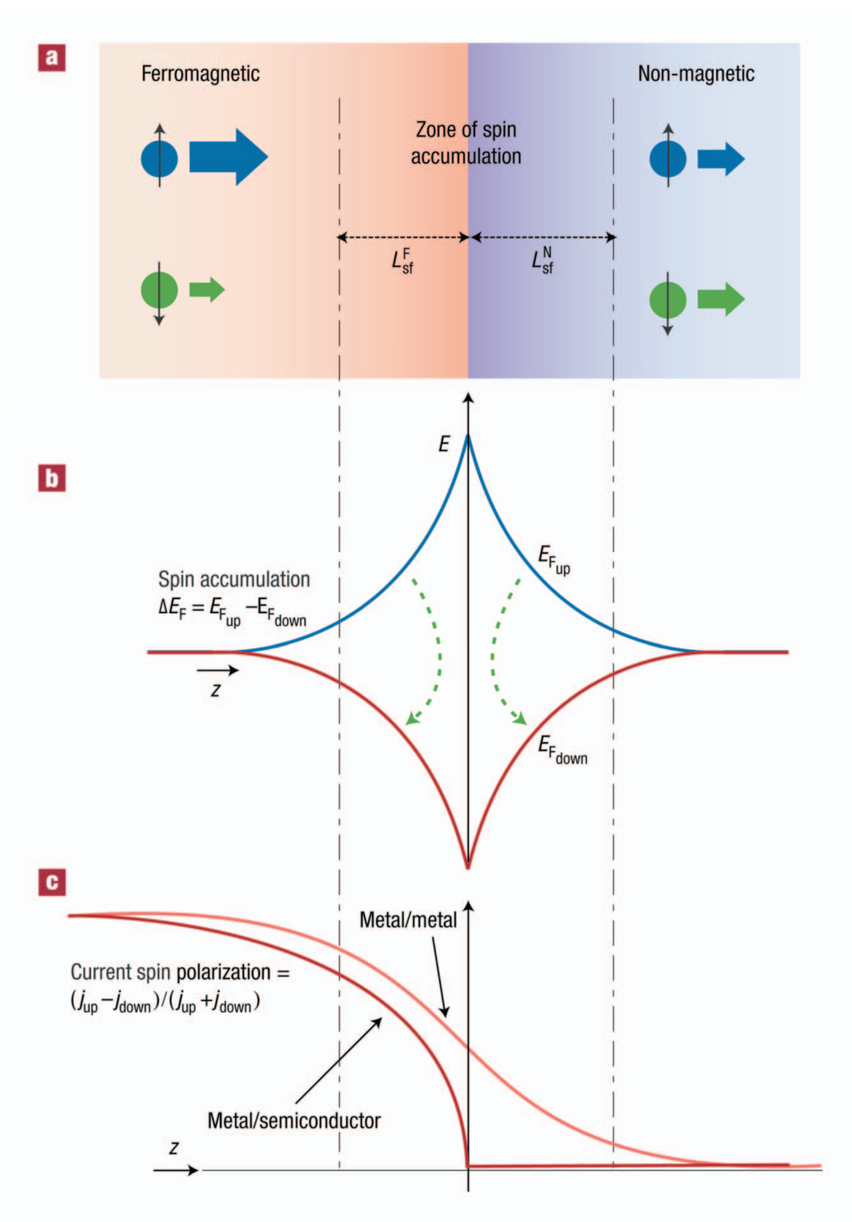

FIG. 6. (Color) Schematic representation of the spin accumulation at an interface between a ferromagnetic metal and a nonmagnetic layer. (a) Spin up and spin down currents far from an interface between ferromagnetic and nonmagnetic conductors (outside the spin-accumulation zone). (b) Splitting of the 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). From Chappert *et al.*, 2007.

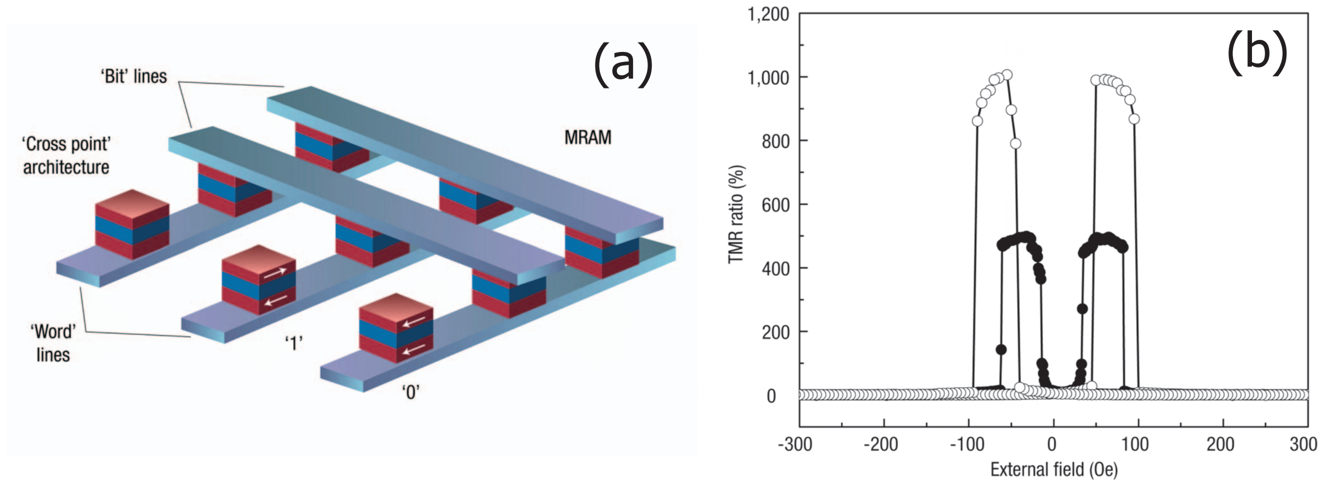

FIG. 7. (Color) (a) Principle of the magnetic random access memory (MRAM) in the basic "cross point" architecture. The binary information "0" and "1" is recorded on the two opposite orientations of the magnetization of the free layer of magnetic tunnel junctions (MTJs), which are connected to the crossing 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 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 Chappert *et al.*, 2007. (b) High magnetoresistance, $\mathrm{TMR}=(R_{\max}-R_{\min})/R_{\min}$, measured by Lee *et al.* (2007) 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 (closed circles) and low temperature (open circles).

(spin extraction), the situation is similar except that a 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 equations (Valet and Fert, 1993) 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 to a multi-interface system 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 (Yu and Flatté, 2002; Fert *et al.*, 2007).

The physics of spin accumulation play an important role in many fields of spintronics, for example, in one of the most active fields 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 makes that similar spin accumulation splittings on the two sides of the interface, as shown 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. 6(c). This problem was first raised by Schmidt *et al.* (2000). 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 by Rashba) (Rashba, 2000; Fert and Jaffrès, 2001). 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 (Fert *et al.*, 2007).

MAGNETIC TUNNEL JUNCTIONS AND TUNNELING MAGNETORESISTANCE (TMR)

An important stage in the development of spintronics has been the research on the tunneling magnetoresistance (TMR) of the magnetic tunnel junctions (MTJ). The 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 been already reported by Jullière (1975), but they were not easily reproducible and actually could not be really reproduced for 20 years. It is only in 1995 that large (≈20%) and reproducible effects were obtained by Moodera and Miyasaki's groups on MTJ with a tunnel barrier of amorphous alumina (Miyazaki and Tezuka, 1995; Moodera *et al.*, 1995). 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 techniques. The MTJs are at the basis of a new concept of magnetic memory called MRAM (magnetic random access memory) and are schematically represented in Fig. 7(a). The MRAMs are expected to combine the short access time of the semiconductor-based RAMs and the nonvolatile 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. 7(a). The next generation of MRAM, called ST-RAM, 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 an amorphous tunnel barrier (alumina) to single crystal MTJ and especially MTJ with a 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 results (Bowen *et al.*, 2001) 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 (Yuasa *et al.*, 2004) and IBM (Parkin *et al.*, 2004) 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 (Lee *et al.*, 2007) [Fig. 7(b)]. In such MTJ, the single crystal barrier filters the symmetry of the wave functions of the tunneling electrons (Mathon and Umerski, 1999; Mavropoulos *et al.*, 2000; Zhang and Butler, 2004), 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 under the word "spin polarization" in a spintronic experiment. In the example of Fig. 8, taken from Zhang and Butler (2004), one sees the density of states of evanescent waves 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 subband 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 predominantly selects evanescent wave functions of Δ_5 symmetry, which are connected to minority spin states of cobalt (Velev *et al.*, 2005; Bowen *et al.*, 2006). This explains the negative effective spin polarization of cobalt we had observed in $SrTiO_3$-based MTJ (De Teresa *et al.*, 1999). 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 Fig. 1(b).

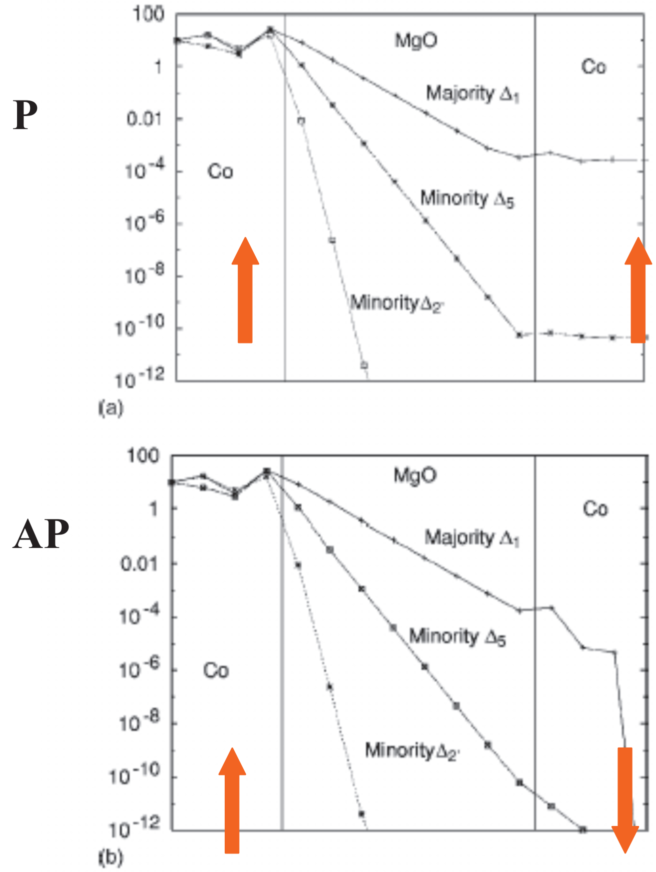

FIG. 8. (Color) Physics of TMR illustrated by the decay of evanescent electronic waves of different symmetries in a MgO(001) layer between cobalt electrodes calculated by Zhang and Butler (2004). The Δ_1 symmetry of the slowly decaying tunneling channel is well represented at the Fermi level of the spin 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. This explains the very high TMR of this type of junction.

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 directions 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 $La_{2/3}Sr_{1/3}MnO_3$ electrodes (Bowen *et al.*, 2003) but

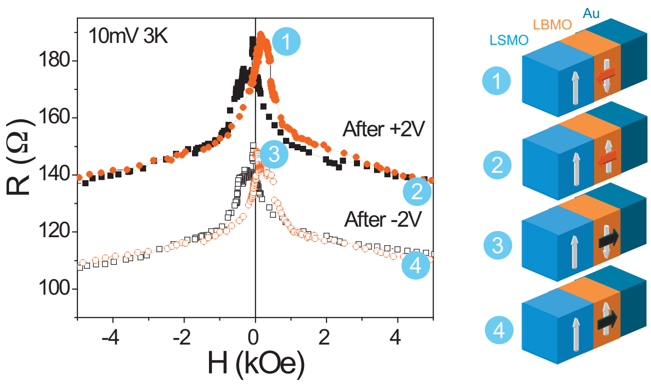

FIG. 9. (Color) Four state resistance of a tunnel junction composed of a biferroic tunnel barrier ($La_{0.1}Bi_{0.9}MnO_3$) between a ferromagnetic electrode of $La_{2/3}Sr_{1/3}MnO_3$ and a nonmagnetic electrode of gold. The states 1–4 correspond to the magnetic (white arrows) and electric (black arrows) polarizations represented on the right side of the figure. From Gajek *et al.*, 2007.

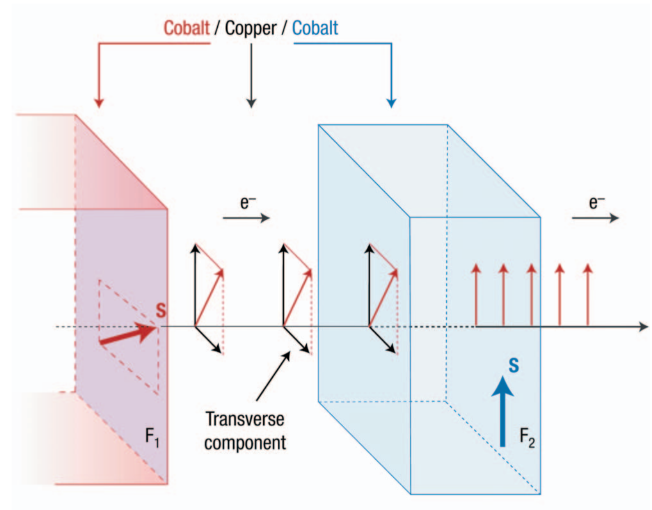

FIG. 10. (Color) Illustration of the spin-transfer concept introduced by Slonczewski (1996). A spin-polarized current is prepared by a first magnetic layer F with an obliquely oriented spin polarization with respect to the magnetization axis of a second layer F_2. When this current goes through F_2, 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. From Chappert *et al.*, 2007.

the Curie temperature of this manganite (around 350 K) is too low for applications. It now turns out from recent results in Japan (Ishikawa *et al.*, 2006) that ferromagnets of the family of the Heusler alloys also present very large TMR ratios with still 90% at room temperature (Ishikawa *et al.*, 2006). Another interesting concept that we are exploring is spin filtering by tunneling through a ferromagnetic insulator layer (Leclair *et al.*, 2002; Ramos *et al.*, 2007). This 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 (Leclair *et al.*, 2002) at MIT and at Eindhoven. Promising results with insulating ferromagnets of much higher Curie temperature have been recently obtained, see, for example, Ramos *et al.* (2007). Some of the magnetic barriers we have recently tested in MTJ 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 (Gajek *et al.*, 2007), as shown in Fig. 9.

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 Slonczewski (1996) and appears also in papers by Berger (1996), 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.* (1998) but a clear understanding came later from measurements (Katine *et al.*, 2000; Grollier *et al.*, 2001) performed on pillar-shaped metallic trilayers [Fig. 11(a)]. In Figs. 11(b) and 11(c), I present examples of our experimental results in the low field regime of irreversible switching, for a metallic pillar and for a tunnel junction with electrodes of the ferromagnetic semiconductor $Ga_{1-x}Mn_xAs$. For metallic pillars or tunnel junctions with electrodes made of a dilute ferromagnetic transition metal like 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 in other groups and can be as short as 100 ps, which is very attractive for the switching of MRAM. For the tunnel junction of Fig. 11(c), the switching current is only about 10^5 A/cm^2 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

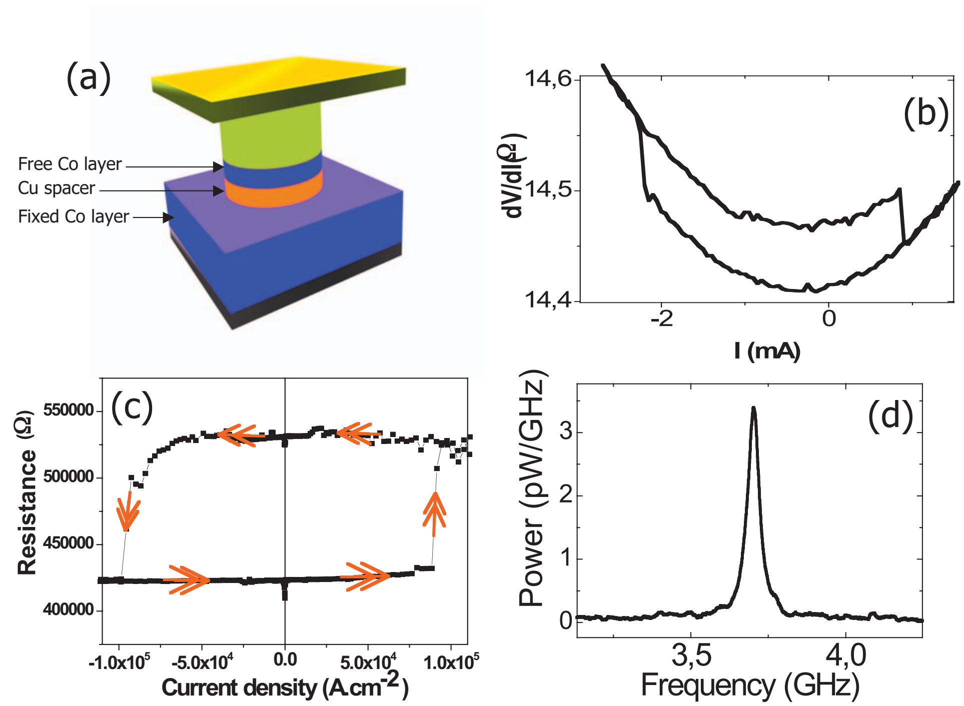

FIG. 11. (Color) Experiments of magnetic switching and microwave generation induced by spin transfer from an electrical dc current in trilayered magnetic pillars. (a) Schematic of a trilayered magnetic pillar. (b) Switching by spin transfer between the parallel and antiparallel magnetic configurations of a Co/Cu/Co metallic pillar (Grollier *et al.*, 2001; Grollier, 2003). 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 (Elsen *et al.*, 2006). The critical current is about 100 times smaller than in the Py/Cu/Py pillar. Similar results have been obtained by Hayakawa *et al.* (2005). (d) Typical microwave power spectrum of a Co/Cu/Py pillar (Py=Permalloy) (Boulle, 2006; Boulle *et al.*, 2007).

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 (Rippart *et al.*, 2004). 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 spin-transport equations (Kovalev *et al.*, 2002; Slonczewski, 2002; Barnas *et al.*, 2005; Stiles and Miltat, 2006), while the description of the switching or precession of the magnetization raises problems of nonlinear dynamics (Stiles and Miltat, 2006). All these problems interact 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 (Boulle, 2006; Boulle *et al.*, 2007).

The spin-transfer phenomena will have certainly important applications. Switching by spin transfer will be used in the next generation of MRAM (ST-RAM) 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 nanocontacts inducing spin-transfer excitations in the same magnetic layer (Kaka *et al.*, 2005; Mancoff *et al.*, 2005). 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 (Grollier *et al.*, 2006).

SPINTRONICS WITH SEMICONDUCTORS AND MOLECULAR SPINTRONICS

Spintronics with semiconductors (Jonker and Flatté, 2006; Awschalom and Flatté, 2007) 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, nonvolatility, 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 (Datta and Das, 1990). Some nonmagnetic semiconductors have a definite advantage on metals in terms of spin-coherence time and propagation of spin polarization on long distances (Jonker and Flatté, 2006; Awschalom and Flatté, 2007). 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.* (2000) 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 (Rashba, 2000; Fert and Jaffrès, 2001) 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 and extraction through a tunnel contact has been now demonstrated in spin LEDs and magneto-optical experiments (Stephens *et al.*, 2004; Jonker and Flatté, 2006; Awschalom and Flatté, 2007).

(ii) Another road for spintronics with semiconductors is based on the fabrication of ferromagnetic semiconductors. The ferromagnetic semiconductor $Ga_{1-x}Mn_xAs$ ($x \approx$ a few percent) has been discovered (Ohno *et al.*, 1996) 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 (tunneling anisotropic magnetoresistance) effects. However, its Curie temperature has reached only 170 K, well below room temperature, which rules out most practical applications. 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 (Zhang, 2000; Kato *et al.*, 2004; Koenig *et al.*, 2007), 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 (Vila *et al.*, 2007; Seki *et al.*, 2008) and the research is also very active in this field. 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 in copper (Fert *et al.*, 1981).

Several groups have tried to probe the potential of spintronics with semiconductors by validating experimentally the concept of spin FET (Datta and Das, 1990) 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 (2006) 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 (Fert *et al.*, 2007) this can be understood in the models (Fert and Jaffrès, 2001) 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) 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. 12(c) 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.* (2006) 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.

A recently emerging direction is spintronics with molecules (see Fig. 13). 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 very small spin-orbit coupling. Promising experimental results have been published during the last years on spin transport in carbon nanotubes

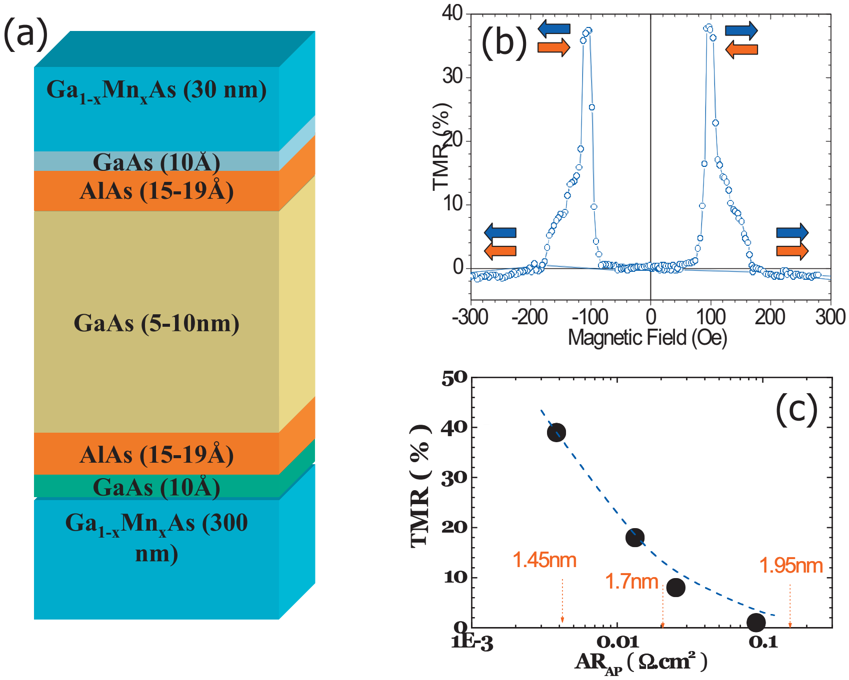

FIG. 12. (Color) Spintronics with semiconductors illustrated by experimental results (Mattana *et al.*, 2003; Fert *et al.*, 2007) on the structure represented on the right side and composed of a GaAs layer separated from the GaMnAs source and drain by tunnel barriers of AlAs. (a) MR curve at 4.2 K showing 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.

(Cottet *et al.*, 2006; Hueso *et al.*, 2007). In particular, my recent work (Hueso *et al.*, 2007) in collaboration with a group in Cambridge on carbon nanotubes between a 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.

CONCLUSION

In less than 20 years, we have seen spintronics increasing considerably the capacity of our hard disks, extending the hard disk technology to mobile appliances like cameras or portable multimedia players, entering the automotive industry and biomedical 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 research of today on the spin-transfer phenomena, on multiferroic materials, on spintronics with semiconductors, and molecular spintronics, opens fascinating new fields and is also very promising of multiple applications. Another perspective, out of the

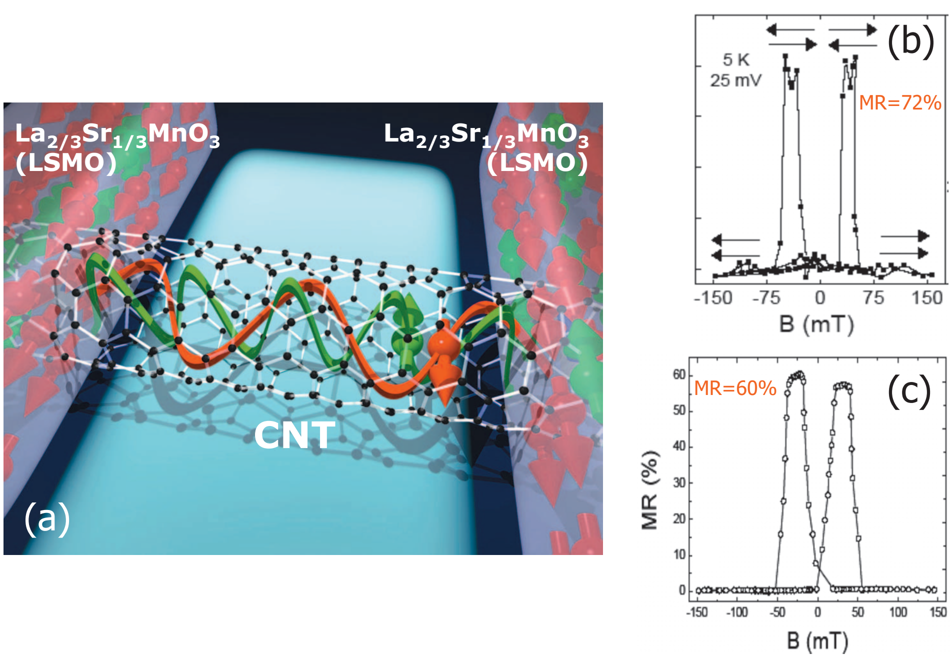

FIG. 13. (Color) Spintronics with molecules as illustrated. Left: Artistic view of spin transport through a carbon nanotube between magnetic electrodes (courtesy of T. Kontos). Right: Magnetoresistance experimental results (Hueso *et al.*, 2007) at 4.2 K on carbon nanotubes between electrodes made of 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. The voltage difference at constant current can reach 60 mV.

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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