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MULTILAYERS OF MAGNETIC MATERIALS AND THEIR APPLICATION IN SPINTRONICS

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An overview is presented on a state of art in a rapidly developing field of spin electronics. Major attention is attributed to a new class of ferromagnetic materials exhibiting spin-polarized carriers (half-metallic ferromagnets). Various systems based on ferromagnetic metals, metal oxides, and semiconductors as well as related technological problems are discussed. Several important applications based on spin-polarized electronic transport in the multilayered systems are illustrated and recent developments in fabrication of novel devices are described.

Keywords: ferromagnetic materials, thin films, heterostructures, spintronics

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1. Introduction

During almost all last century, researchers were concentrating mainly on the understanding of magnetization and micromagnetic phenomena of ferromagnetic materials rather than on a deeper study of their specific electronic properties. Precise control of electronic charge rather than a spin of carriers was the major task of electronic industry. However, now we are confronting with a new rapidly expanding research area of magnetism – spin electronics, or simply spintronics, exploiting spin-dependent electrical transport in various multilayered structures of ferromagnetic materials. During the last few years, increasing attention was attributed to a new class of ferromagnetic materials known as half-metallic ferromagnets (HMF). Manganites, chromium dioxide (CrO_2), magnetite (Fe_3O_4), and others are examples of such materials. In contrast to the well-known ferromagnets such as Co, Ni, and Fe they exhibit almost completely spin-polarized carriers which are of key importance for most applications in spintronics.

The first steps towards the utilization of spin-dependent transport started in 1988 with the discovery of the Giant Magnetoresistance (GMR) effect in metallic multilayers (the so-called spin valves) [1]. Now GMR spin valves are widely used in modern computers for read heads of hard disks. New devices based on tunnelling magnetoresistance (TMR) in magnetic tunnel junctions seem to be the next generation of devices that will have soon important applications [2]. Exploitation of spin in addition to electrical charge in semiconductors provides a number of new promising possibilities. In future, semiconductor-based spintronics could combine storage, detection, logic, and communication capabilities on a single chip.

Magnetic multilayers composed of various magnetic and nonmagnetic materials are of key importance for spintronics. Depending on a kind of basic ferromagnetic materials and related applications, this wide and multidisciplinary research area can be divided into three major fields of activity (see Table 1):

1. Metallic multilayers, exhibiting giant magnetoresistance effect, and related applications.
2. Spintronics based on ferromagnetic oxides and related heterostructures.
3. Semiconductor multilayers and their application in semiconductor-based spintronics.

To ensure progress in all these fields, there is increasing need in advanced ferromagnetic materials offering spin-polarized carriers at room temperature. Attention needs to be focussed on technology of thin

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Table 1. Basic spintronics materials, their properties, and applications.

Materials used in spintronics	Useful properties	Application. Devices
1. FM metals and alloys Fe, Co, Mn, NiMnSb, Mn_2VAl **2. FM oxides** $La_{1-x}A_xMnO_3$, $LaSrMoO_4$, CrO_2, Fe_3O_4 **3. Magnetic semiconductors** (Ga,Mn)As, (Be)ZnMnSe **4. Nanostructured materials** Magnetic quantum dots	**1. Intrinsic properties** • Anisotropic magnetoresistance (AMR) • Colossal magnetoresistance (CMR) • Kerr effect • Hall effect • Spin filtering effect **2. Extrinsic properties** • Giant magnetoresistance (GMR) in FM-M multilayers • Tunnelling magneto-resistance (TMR) • Domain wall magneto-resistance (DWMR) • Magnetization reversal by spin transfer • Shift of domain wall by spin transfer	• Read heads • Magentic sensors • Magnetic switches • Information storage devices • Magnetic memory (MRAM) • Spin-dependent field effect transistor (SFET) • Spin-dependent light emiting diode (SLED) • Multifunctional electronics • Computer logics • Magnetooptical elements

films and multilayers and deeper knowledge of spin-dependent transport in thin films, interfaces, and various device structures.

Hereafter we will overview the most important ferromagnetic materials, their heterostructures, new effects, and promising applications.

2. Half-metallic ferromagnets

Significant contribution of d-electrons on formation of electronic band structure in a vicinity of Fermi level of ferromagnetic materials results in specific electronic properties. The most interesting feature is the magnetization-induced splitting of the conduction band into two subbands. Thus, certain preferential orientation of carriers may be expected for a ferromagnet cooled below the Curie temperature. The degree of spin polarization of carriers, P, is defined by the relationship [3]:

$$P = \frac{N\uparrow(E_\mathrm{F}) - N\downarrow(E_\mathrm{F})}{N\uparrow(E_\mathrm{F}) + N\downarrow(E_\mathrm{F})}, \qquad (1)$$

where $N\uparrow$ and $N\downarrow$ are the density of states for spin-up and spin-down carriers at the Fermi level. The spin polarization of a material may be either positive or negative. $P > 0$ is measured if the majority spin at the Fermi level is parallel to the bulk magnetization and $P < 0$ if the minority spin at the Fermi level is aligned parallel to the magnetization vector.

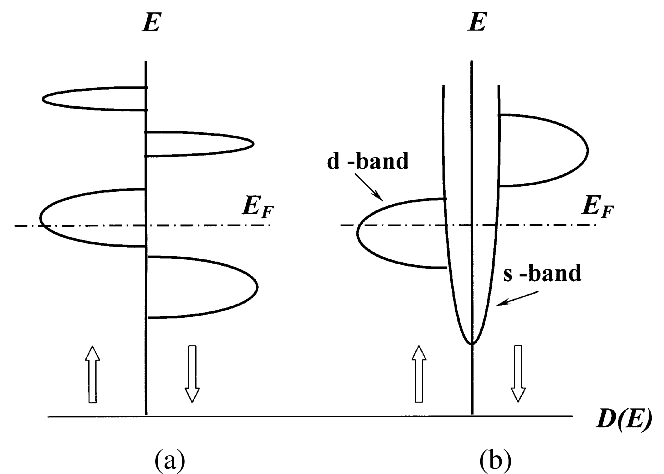

Fig. 1. (a) Schematic drawing of band structure of half-metallic ferromagnet and (b) that of elemental ferromagnets (Co, Fe, and Ni).

The concept of a half-metal has been introduced first by de Groot et al. in 1983 [4]. A half-metallic feromagnet (HMF) has been defined as a material demonstrating a metallic density of states in one of the spin channels and a gap in the density of states in the other one (see Fig. 1(a)). Perfect spin polarization ($P = 100\%$) is of key importance for spintronics: half-metallic electrodes can act as sources of spin-polarized electrons as well as a magnetically controllable spin filter [5, 6].

Various experimental techniques such as spin-resolved photoelectron emission spectroscopy, positron annihilation, spin-dependent transport measured in point contacts and tunnel junctions, the ferromagnet–

Table 2. The most important half-metallic ferromagnets and their properties: Curie temperature, electronic configuration of spin-up and spin-down states at E_F, magnetic moment, resistivity at 300 K, and polarization of carriers [4–6].

Half-metallic ferromagnet	T_C, K	Spin-up state	Spin-down state	Magnetic moment, μB	ρ(300 K), $\mu\Omega$·cm	P, %
$La_{2/3}Sr_{1/3}MnO_3$	390	$3d$-e_g (Mn)	$3d$-t_{2g} (Mn)	3.7	~50	80–100
Sr_2FeMoO_6	421	–	$5d$-t_{2g} (Mo)	4		
CrO_2	396	$3d$-t_{2g} (Cr)	–	2	~5	80–90
Fe_3O_4	860	–	$3d$-t_{2g} (Fe)	4	~10000	80–90
NiMnSb	730	–	e_{2g} (Ni)	4		30–60

superconductor tunnel junction technique as well as tunnelling of carriers between ferromagnetic electrodes separated by an ultrathin nanometric insulator layer were employed to probe half-metallicity of most ferromagnets known up to now [5, 6].

The well-known ferromagnetic metals such as Fe, Ni, and Co demonstrate high Curie temperature values of 1043 K, 631 K, and 1403 K, respectively. Unfortunately, spins of carriers in all these metals are only partially polarized ($P \leq 30\%$) [5]. The presence of two kinds of carriers in metallic Co, Fe, and Ni may be easily understood taking into account that the Fermi level in all these metals crosses spin-polarized d- and unpolarized s-bands (see Fig. 1(b)).

Among the materials exhibiting the highest polarization values are oxides $La_{1-x}Sr_xMnO_3$, Sr_2FeMoO_6, CrO_2, Fe_3O_4, intermetallic compounds (Heusler alloys), as NiMnSb [3, 5, 6] (see Table 2). Half-metallic oxides with the highest Curie temperature values, namely Fe_3O_4 (T_C = 860 K), Sr_2FeMoO_6 (T_C = 421 K), and CrO_2 (T_C = 390 K) are the most promising for room temperature applications [5, 6]. Also it is worth noting diluted magnetic semiconductors such as (Ga, Mn)As exhibiting almost completely spin-polarized carriers. However, use of these semiconducting materials for spintronics is limited by rather low Curie temperature values (T_C < 150 K) [7].

We are turning now to characterize the most important ferromagnetic materials and their properties in more detail.

3. Advanced half-metallic ferromagnets and their properties

Colossal magnetoresistance manganites referred to as $R_{1-x}M_xMnO_3$ (here R is a trivalent rare earth ion such as La, Nd, Pr etc., and M represents a divalent (Ca, Ba, and Sr) or a tetravalent Ce ion) crystallize in a simple cubic perovskite structure. Hopping of carriers between neighbouring Mn^{3+} and Mn^{4+} ions provides p-type electrical conductivity in the compounds doped by divalent ions, while a mixed Mn^{2+}–Mn^{3+} valence results in electronic conductivity of Ce-doped manganite. The highest electrical conductivity in most of the manganites is indicated at $x = 0.3-0.4$. The characteristic phase transition from a paramagnetic insulator (PI) to a ferromagnetic metal (FM) is observed in both hole- and electron-doped manganites. The corresponding Curie temperature, T_C, depending either on composition or oxygen content may vary in a wide range (from about 100 K to 350 K). The highest T_C value of about 350 K has been measured for Sr-doped $La_{0.67}Sr_{0.33}MnO_3$, meanwhile significantly lower T_C values (<300 K) were reported up to now for Ca-, Ba-, and Ce-doped manganites [8].

A number of authors reported an enormous decrease in resistance (by several orders of magnitude) for high-quality manganite thin films just below their PI–FM transition temperature under applied magnetic field of 3–5 T [5]. This unusual magnetoresistance effect known as colossal magnetoresistance (CMR) seems very attractive for several applications. However, for spintronics, there is greater need in low field magnetoresistance (LFMR), i. e. in a reproducible resistance change by applying magnetic field in the mT range [5, 6].

Search for a possible low-field magnetoresistance (LFMR) effect and increased operation temperature of the manganites has been the goal of researchers worldwide. Significant progress in reducing the field scale has been recently achieved by employing spin-polarized carriers in polycrystalline material and various artifical device structures. High P values for the manganites have been proved recently by spin-polarized photoemission [9] and scanning tunnelling spectroscopy [10] measurements. Significant low field tunnelling magnetoresistance (at $B \sim 1$ mT) has been reported by measured tunnelling of spin-polarized electrons in the FM/I/FM trilayer structures based on manganite thin films [5].

High-quality manganite films and related multilayered structures now can be prepared relatively easy by applying magnetron sputtering, pulsed laser deposition, and other techniques. Thus, most of the manganites seem very promising for fabrication of tunnelling heterostructures. High tunnelling magnetoresistance of the heterostructures could be employed for magnetic field sensors, read heads, and various memory applications. Nevertheless, enhanced T_C values of the manganites would be highly appreciated for devices operating at room temperature.

Magnetite (Fe_3O_4) has an inverse cubic spinel structure ($a = 0.8396$ nm). Fe cations in this structure occupy interstices of a face-centred-cubic frame of oxygen ions. Fe^{3+} occupies the eight tetrahedral (A) sites, while Fe^{3+} and Fe^{2+} equally share the 16 octahedral (B) sites [11, 12].

Magnetite is a half-metallic ferromagnet exhibiting the highest Curie temperature ($T_C \cong 858$ K) among other known ferromagnetic (FM) oxides such as $La_{1-x}Sr_xMnO_3$, Sr_2FeMoO_6, and CrO_2. Electrical conductivity of the magnetite is due to hopping of spin-polarized electrons between ferrimagnetically ordered Fe^{2+} and Fe^{3+} states. Resistance of the compound increases with cooling down to the so-called Verway transition point at $T = T_V = 120-110$ K where a structural transition from a ferromagnetic to a high resistance charge ordered state is indicated. In contrast to CMR manganites, relatively small magnetoresistance was measured for the oxide in the whole temperature range [14].

Thin magnetite films and related heterostructures are of great importance for novel applications although preparation of high-quality Fe_3O_4 films is complicated due to the presence of other Fe oxides, namely Fe_2O_3 (hematite) and FeO (wuestite) in the Fe–O phase diagram [11]. There is great need in suitable lattice-matched substrates to grow high-quality magnetite films and multilayers. Epitaxial growth of Fe_3O_4 thin films has been demonstrated recently on lattice-matched MgO(100) ($a_{MgO} = 0.418$ nm $\cong a_{Fe_3O_4}/2$) [13–15] and perovskite ($SrTiO_3$) [14] substrates. However, for such applications as sensors and magnetic memory devices, growth of various hybrid device structures composed of various ferromagnetic, antiferromagnetic, highly conducting films, and isolating barrier layers will be highly appreciated.

Chromium dioxide (CrO_2) is a metallic oxide exhibiting ferromagnetic properties below 390 K. The oxide has a rutile structure with a tetragonal unit cell ($a = b = 0.4419$ nm and $c = 0.212$ nm) consisting of two formula units. Chromium ions in this structure are in the Cr^{4+} state with the electronic configuration [Ar]$3d^2$ and a magnetic moment of 2 μB per ion. Nearly complete spin polarization has been certified for the compound by spin-polarized photoemission and vacuum tunnelling experiments [16, 17]. Metallic conductivity of CrO_2 is governed by the majority carriers, meanwhile, there is a gap of about 1.5 eV in the minority density of states. CrO_2 is characterized by a relatively small magnetoresistance with unusual field dependence. However, use of CrO_2 as a source of spin-polarized electrons in thin film devices seems rather complicated due to difficulties in preparation of thin films and multilayers and thermal decomposition of the compound above 450 °C [18].

Double perovskite (Sr_2FeMoO_6) has a layered structure with alternating perovskite-like $SrFeO_3$ and $SrMoO_3$ layers. The compound is a half-metallic ferromagnet with Curie temperature of 410–450 K depending on stoichiometry and lattice defects. Metallic conductivity of Sr_2FeMoO_6 is governed by spin-down Mo electrons. The up-states of the Fe ion are filled, but the down-states are empty so that the Mo electrons may hop to them. Thin films of the layered compound could be used as a source of spin-polarized electrons in various device structures. It must be noted, however, that preparation of high-quality thin films and multilayered structures of this layered compound is more complicated in comparison to those of perovskite manganites [5, 19].

Diluted magnetic semiconductors such as (Ga, Mn)As and (Hg, Mn)Se form another class of half-metals. These ferromagnetic semiconductors have two different band gaps for each spin direction. When a small concentration of electrons or holes is doped (or, in more recent experiments, injected) into the semiconductors, the carriers will only conduct current if their spins are completely polarized. However, recent theoretical calculations predict possible T_C values just above room temperature for several doped wide-gap semiconductors such as GaP, ZnO and others [7].

4. Metallic multilayers. Giant magnetoresistance devices (spin valves)

Spin valve is a device composed of a nonmagnetic conducting layer of several nanometres (3–5 nm) in thickness sandwiched between two magnetic layers [20, 21]. The GMR effect of such multilayer systems

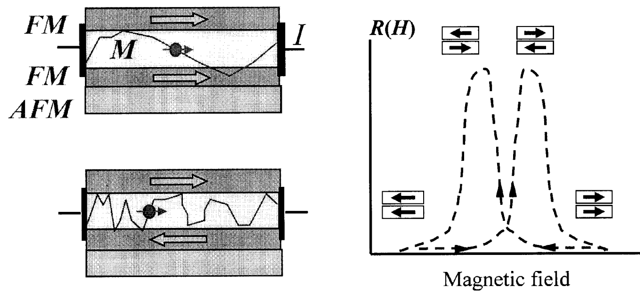

Fig. 2. Schematic drawing of spin valve consisting of thin layer of normal metal sandwiched between two adjacent ferromagnetic layers demonstrating giant magnetoresistance under applied magnetic field.

is a dramatic variation of the electrical resistance with applied magnetic field. Resistance of the multilayered structure is high in the absence of an external field. However, it drops with applied magnetic field forcing the initially antiparallel magnetization of the coupled FM layers into a parallel alignment.

An example of the spin valve (GMR-based sensor) is shown in Fig. 2. The magnetoresistance ratio for the device is defined as

$$\mathrm{GMR} = \frac{\Delta R}{R} = \frac{(R_{\max} - R_{\min}) \cdot 100(\%)}{R_{\min}}, \quad (2)$$

where $R_{\max}$ and $R_{\min}$ are the maximal and minimal resistance values corresponding to parallel and antiparallel orientation of magnetic moments of the adjacent FM layers. Most electronic applications need multilayer systems such as NiFe/Cu/Co/Cu with low switching field values of about 1 mT. The maximum GMR effect reported up to date is about 80% reduction in resistivity for multilayers and a 20% reduction for trilayers at ambient temperature. Spin-dependent scattering is the main but not the only possible explanation of the GMR effect. Early studies on thin film structures revealed the phenomenon of exchange bias through which an antiferromagnetic layer can cause an adjacent ferromagnetic layer to develop a preferred direction of magnetization [22, 23].

About ten years after the discovery of the basic phenomena, the first GMR-based devices started to enter the market as sensors and read heads in magnetic recording systems and, in particular, in computer hard-disk drives. However, to date new magnetoelectronic devices based on metallic multilayers such as spin transistors and spin valve based random access memories are under development. The increasing technological interest stimulates further research of the magnetic multilayers.

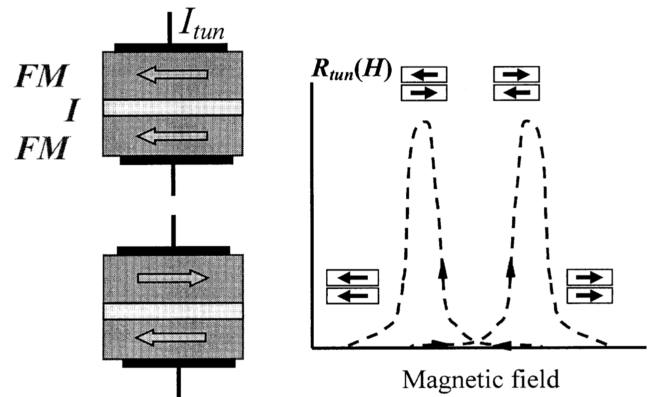

Fig. 3. Schematic drawing of tunnelling heterostructure composed of two ferromagnetic layers separated by a nanometric barrier layer. The heterostructure demonstrates tunnelling magnetoresistance (TMR) effect in a low magnetic field.

5. Magnetic tunnel junctions and MRAM

Magnetic tunnel junction (MTJ), shown schematically in Fig. 3, is a multilayer structure containing very thin insulating layer stacked between adjacent FM layers [5]. In MTJ, a voltage applied between ferromagnetic films causes a tunnelling current to flow between the FM electrodes across the interlayer. Probability of tunnelling for spin-polarized carriers from a ferromagnetic electrode depends on magnetization direction of electrodes. The resultant resistance of the MTJ is different for the parallel and antiparallel orientation of the magnetic moments of the electrodes.

The tunnelling magnetoresistance effect (TMR) was firstly reported by Julliere in 1975 [24]. However, strong interest appeared only in the early 1990s, when increased values of TMR were reported. The magnitude of TMR depends on the relative orientation of the magnetizations on both sides of the interlayer. In this case the measured resistance is higher for antialignment of the adjacent ferromagnetic electrodes.

In some cases, TMR appears to perform better than GMR: for double layers, record values for TMR of around 50% have been reported at room temperature compared to 20% for GMR in the same kind of double-layer system [5]. The MTJ with size of elements below the micron range can be fabricated by applying lithography techniques. Small size MTJ is of great importance for computer memory, MRAM (Magnetic Random Access Memory). The MRAM presently in development are expected to reach similar densities and access times as the current semiconductor-based dynamic random access memory (DRAM). Reduced write time of MRAM and the fact that MRAM retains data af-

ter the power is turned off are the main advantages of MRAM over DRAM [6].

Most of the perovskite manganites exhibiting fully spin-polarized carriers demonstrate high chemical and thermal stability. Thus, one can expect formation of sharp interfaces in the heterostructures containing different metal oxides. Most of the oxides with a simple perovskite or perovskite related crystalline structure are characterized by similar lattice parameters related to unit cell dimensions of their basic perovskite unit $a_{\rm per}$ ($a_{\rm per} \approx 3.8-3.9$ Å, $\sqrt{2}a_{\rm per} \approx 5.3-5.5$ Å). This provides a promising possibility to grow different oxides heteroepitaxially in a form of lattice-matched bilayer films and multilayer structures.

6. Ferromagnetic semiconductor heterostructures and their application

Semiconductor spintronics offers several important advantages such as control of doping and fabrication of various structures, signal amplification, electronically tunable spin–orbit coupling, optical manipulation, and simple integration using available semiconductor technology.

The heterostructures composed of ferromagnetic semiconductors combine useful properties of both ferromagnetic and semiconductor material systems. Semiconductor-based spintronics could combine storage, detection, logic, and communication capabilities on a single chip to produce a multifunctional device. Optical properties of the semiconductors may be employed to transform a magnetic information into an optical signal [7].

It is also possible to inject spin-polarized carriers into semiconductor heterostructures from ferromagnetic metal in order to transform spin information into an optical or electrical signal. However, there is lack of knowledge about spin-polarized transport and spin dynamics in inhomogeneous semiconductors and their heterostructures. Several complications such as band bending at the interface and conductivity mismatch effects can arise for ferromagnetic metals in contact with semiconductors.

7. Other device structures. Important applications

Thin films of ferromagnetic materials can be combined with other materials exhibiting different magnetic, electrical, and optical properties. During the last few years there were a number of reports of various heteroepitaxial bilayer films composed of FM, high temperature superconductor (HTS), ferroelectric (FE), and other oxide materials as well as the corresponding multilayer heterostructures. The multilayers composed of CMR manganites and high temperature superconductors such as $YBa_2Cu_3O_7$ (YBCO) demonstrate interesting combination of properties. Electrical properties of the interfaces between CMR manganites and HTS compounds have been reported recently [25–30]. It was found that injection of spin-polarized carriers from CMR material into a HTS through an ultrathin $SrTiO_3$ barrier leads to a suppression of superconducting properties such as critical temperature and critical current. This suggests that spin-polarized transport can be utilized as a tool for the investigation of spin-dependent electronic properties and opens the possibility of a new class of superconducting devices.

8. Unresolved technological problems and trends for further research

For creation of possible hybrid circuit devices based either on metallic or metal oxide multilayers, there is a great need in sufficient crystalline quality of individual layers in order to obtain their optimal properties. There still remain many problems that must be overcome before a viable hybrid comercial devices can be produced. Selection of compatible materials and substrates suitable for heteroepitaxial growth of bilayer or multilayer films will be of crucial importance for further technological progress. Chemical stability of the materials, matching of their lattices in various heterostructures as well as thermal expansion match are among the most important requirements. One needs to develop the most suitable technologies and to optimize growth conditions in order to obtain necessary properties and sufficient surface quality of individual layers. Special attention could be paid on strain-dependent structure of ferromagnetic domains at the interfaces, interdiffusion of elements, and especially oxygen exchange at the interface between different materials.

Summarizing one can conclude that concerted efforts on physics, technology, and material science are highly appreciated to solve the problems mentioned above.

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DAUGIASLUOKSNIAI FEROMAGNETIKŲ DARINIAI IR JŲ PANAUDOJIMAS SPINTRONIKOJE

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Santrauka

Apžvalgoje nušviečiama nauja mokslo, o kartu ir praktinės bei gamybinės veiklos sritis – sukininė elektronika, arba spintronika. Pastarosios tikslas – informacijos saugojimui, apdorojimui ir atgaminimui panaudoti ne tik elektrono krūvį, bet ir sukinį. Norima siekti magnetiniu lauku keisti krūvininkų magnetinių sukinių kryptį ir kartu valdyti tekančią elektros srovę. Šiems tikslams įgyvendinti reikalingos naujos feromagnetinės (FM) medžiagos – orientuotų elektronų laidininkai (OEL), kurių krūvininkų sukiniai būtų orientuoti tam tikra kryptimi (dažniausiai išilgai vidinio magnetinio lauko vektoriaus).

Daugelio FM metalų (Fe, Ni, Co) krūvininkai yra tik dalinai orientuoti, ir jų sukinių poliarizacijos laipsnis $P \leq 30\%$, tuo tarpu šiuo metu žinomų magnetinių puslaidininkių (pvz., GaMnAs) Curie temperatūros vertės yra gerokai mažesnės už 300 K. Todėl prietaisams gaminti labiausiai tinka kai kurie feromagnetiniai metališkieji lydiniai, intermetaliniai junginiai (BiMn), o ypatingai – laidieji FM oksidai: $La_{1-x}Sr_xMnO_3$, Sr_2FeMoO_6, CrO_2 ir Fe_3O_4, kurių sukinių poliarizacijos P vertės yra artimos 100%, o Curie temperatūra T_C viršija 300 K.

Daugiausia dėmesio skiriama daugiasluoksniams tuneliniams OEL dariniams, kuriuose elektronai gali tuneliuoti tarp dviejų FM sluoksnių, FM ir normalaus (N) ar superlaidaus (S) laidininko. Tokių heterodarinių didelės tunelinės magnetovaržos (TMV) vertės yra išmatuojamos, esant palyginti silpniems magnetiniams laukams ($B \sim 10$ mT), todėl jie yra labiausiai pageidautini magnetinio lauko jutikliams, taip pat operatyviosios magnetinės atminties elementams kurti. Plačiau aptartos kai kurios technologinės tokių heterodarinių gaminimo problemos, tunelinių sandūrų bei tarpfazinių ribų savybės, paminėti naujausi sukininės elektronikos prietaisai.