This Project aims to integrate engineered nanoporous materials into novel energy-efficient spintronic applications. Magnetic storage and magneto-electronic devices are conventionally controlled by means of magnetic fields (via electromagnetic induction) or using spin-polarized electric currents (spin-transfer torque). Both principles involve significant energy loss by heat dissipation (Joule effect). The replacement of electric current with electric field to operate these devices would drastically reduce the overall power consumption. Strain-mediated magneto-electric coupling in piezoelectric-magnetostrictive bilayers might appear a proper strategy to achieve this goal. However, this approach is not suitable in spintronics because of the clamping effects with the substrate, need of epitaxial interfaces and risk of fatigue-induced mechanical failure. Possibility to control ferromagnetism of
metals and semiconductors directly with electric field (without strain) has been recently reported, but most significant effects occur below 300 K and only in ultra-thin films or nanoparticles. This Project tackles the development of a new type of nanocomposite material, comprising an electrically conducting or semiconducting nanoporous layer filled with a suitable dielectric material (solid or liquid), where the magnetic properties of the metal/semiconductor are intended to be largely tuned at room temperature (RT) by simply applying a voltage, via electric charge accumulation. The porous layer consists of specific alloys (Cu-Ni or Fe-Cu) or oxide diluted magnetic semiconductors, where surface magnetic properties have been recently reported to be sensitive to electric field at RT. Based on these new materials, three technological applications are envisaged: electrically-assisted magnetic recording, voltage-driven switching of magnetic random-access memories and spin field-effect transistors. The obtained results are likely to open new paradigms in the field of spintronics and could be of high economic transcendence.

Work performed from the beginning of the project to the end of the period covered by the report and main results achieved so far

"The work performed so far since the beginning of the project has focussed in three main key tasks:

i) Synthesis of electrically conducting and semiconducting nanoporous materials: Ni, CuNi, FePt, FeCu and CoPt by electrodeposition (using block-copolymer surfactants) and Cu- and Ni-doped SnO2 layers by dip coating (evaporation-induced self-assembly method) and Ni-doped SnO2 and Co3O4 impregnated with FexCo(3-x)O4 mesoporous powders (nanocasting technique, see Figure 1). The growth of nanoporous CuNi and FeCu by electrodeposition onto colloidal-templated substrates has been also attempted. Nanoporous FeCu films have been obtained by dealloying (selective chemical etching). Finally, the growth of FeRh, FeAl, CuNi and FeCu thin films onto piezoelectric substrates has been also carried out. The obtained materials have been thoroughly characterized from a structural point of view using diffraction and electron microscopy techniques. The mechanical integrity of the grown layers has been assessed and modelled using nanoindentation. The magnetic properties have been studied using vibrating-sample magnetometry and magneto-optic Kerr effect.

ii) Filling of the porous frameworks with a dielectric material. Two approaches are being attempted: filling the pores with dielectric polymers (e.g. propylene carbonate) and coating the inner pore walls using atomic layer deposition (basically with Al2O3 and HfO2).

iii) Fundamental studies of electric field actuation on the surface magnetic properties. Interesting results have been obtained in the CuNi, FeCu, FePt and CoPt systems by immersion of the nanoporous film in suitable electrolytes to generate an electrical double-layer. A reduction of coercivity larger than 30% has been observed by magneto-optic Kerr effect in all these systems under applied voltages of the order of 10 V. In some cases, the coercivity can be also increased by applying suitable voltage values of opposite polarity. In other words, the observed effects are reversible. Some attempts to obtain similar effects using a "dry configuration" in porous samples (not immersed in liquid electrolytes, but using solid dielectric like polymers or ceramic layers) are being performed now. The experimental results are being interpreted with the use of ab-initio calculations, to assess the changes in the effective magnetic anisotropy energy due to applied electric field. The influence of applied voltage on the strain-mediated metamagnetic transition in FeRh has been also investigated. Strain-mediated effects are also being investigated in sputtered FeAl thin films (influence of voltage on the effective magnetic anisotropy). Interesting results have been obtained by electrolyte gating."
Effective magnetic anisotropy. Interesting results have been obtained by electrolyte-gating paramagnetic Co3O4 films, where an ON-OFF magnetic transition (from paramagnetic to ferromagnetic and viceversa) can be induced by DC voltage via magnetoionic effects. The influence of voltage-driven controlled redox reactions in nanoporous CuNi films has been also studied by means of magnetometry and synchrotron radiation facilities.

The results obtained so far have led to around 41 publications in peer reviewed journals (Adv. Funct. Mater., Small, ACS Nano, ACS Appl. Mater. Interf., Sci. Rep., Adv. Science J. Mater. Chem. C, etc.). Also, the results have already been presented in several conferences, sometimes as invited talks (e.g. Thermec or the 2nd IEEE Conference on Advances in Magnetics, MMM-Intermag, ISMANAM, etc.). We have also issued two patents (PCT) related to the synthesis of nanoporous metallic alloy films by surfactant-assisted electrodeposition and the observation of magneto-electric effects in them.

Progress beyond the state of the art and expected potential impact (including the socio-economic impact and the wider societal implications of the project so far)

In this Project we aim to study the effects of electric field on the magnetic properties of nanoporous materials and wish to develop the first prototypes of magnetic memories and spintronic devices that incorporate nanoporous conducting or semiconducting layers in their structures (as a proof-of-concept). To this end, the Project seeks new approaches, beyond the state-of-the-art, to merge the advanced synthetic routes for the synthesis of nanoporous materials with the innovative field of spintronics. As examples, we propose voltage-driven magnetic memory, MRAM or spin-FET as possible spintronic nanoporous architectures. The successful implementation of these designs represents a ground-breaking achievement, which will certainly revolutionize the field of spintronics. The drastic effects of an applied voltage on the magnetic properties of nanoporous frameworks will drastically reduce the energy consumption of miniaturized magnetic memory devices.

So far, much progress (beyond the state-of-the-art) has been achieved in our group in the synthesis of magnetic nanoporous films (both metallic and semiconducting) and their structural and magnetic characterization (using magnetometry and dichroism techniques, at large-scale facilities, like the ALBA or the BESSY synchrotrons). Two patents dealing with (i) the synthesis of nanoporous alloys and (ii) the magneto-electric effects observed in nanoporous alloys have been issued at National and European levels. Hence, the results from this project are likely to have a strong scientific and technological impact and could be of high economic transcendence.
Fig. 1: (a) Transmission electron microscopy (TEM) image of non-infiltrated KIT-6 mesoporous Co3O4; (b) TEM image of a pore in Co3O4 partially filled with the guest FexCo3-xO4 oxide; (c) electron energy loss spectroscopy (EELS) mapping in which Co is imaged in green and Fe in red; (d) EELS relative elemental quantification along the line indicated in (c)[56].

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