Voltage-Control of Magnetism in Nanoscale-Engineered Materials: a New Concept for Energy-Efficient Data Storage

SUMMARY

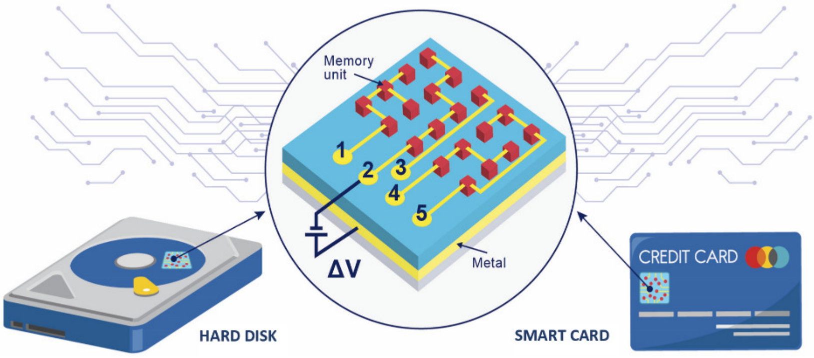

Fig. 1: Schematic drawing of the voltage-actuated multi-element memory chips that we develop to be integrated in energy-efficient hard disk drives or anti-counterfeiting technologies.

Technological progress in industrialized countries has created huge demands for electric energy. Electricity represents almost half of the overall energy spent in Europe. Transmitting electricity from power plants to our homes involves considerable **energy loss in the form of heat dissipation** by Joule effect. About 3-5% of the generated energy is wasted, which is equivalent to thousands of M€ every year. The use of high voltages (and low currents) in power transmission lines is specifically designed to reduce Joule heating losses, since the dissipated power is proportional to i^2, where i denotes the current intensity. **Heating effects are also quite significant in electronic devices**. Magnetic storage in computer hard disks drives (HDDs) and magnetoresistive random-access memories (MRAMs) is conventionally based on the application of localized magnetic fields. The generation of such fields requires the use of rather high electric currents (through miniaturized electromagnets). Other types of non-volatile memories (such as 'flash') integrated, for example, in 'universal series bus' (USB) drives, digital cameras, mobile phones, synthesizers, game consoles, robotics or medical electronics, also require of considerably high currents and are, therefore, not optimized from an energetic point of view.

Our technology brings to light a new concept to **drastically reduce the energy power consumption** (by more than **two orders of magnitude**) and **enhance robustness** and **security** of data storage systems. By analogy to electric power transmission, the main idea here is **to use voltage instead of electric current** to write information in these devices, thus minimizing power dissipation by Joule heating effect. By doing so, the energy efficiency of a myriad of technological applications, such as data servers, cell phones, computers, laptops or many other products related to the Internet of Things can be drastically enhanced. An additional added value of our data storage concept is that the selective application of voltage (instead of magnetic field) to record information on the magnetic memory units improves the robustness and security of the resulting devices, offering unique opportunities, as well, for innovative **anti-counterfeiting** and **anti-hacking technologies** (Fig. 1).

Conceptually, this technology has been built up in **two main phases**. In **phase 1**, which initiated in 2015 (when Prof. Sort was awarded a **Consolidator Grant** from the **European Research Council**), voltage was utilized to **reduce the coercivity** (width of the hysteresis loop) of target materials (mesoporous thin films and patterned structures). As shown in Fig. 2a, the obtained reduction was as high as 88% in some cases. This means that, when the material was subjected to the action of suitable voltages, magnetization reversal (switching between states "1" and "0" in the recorded data) occurred at much lower applied magnetic fields, thus requiring lower electric currents during the writing process. This represents a significant improvement of the energy efficiency since heating effects are minimized. The decrease of coercivity was ascribed to voltage-induced ion motion and changes in the electronic band structure of the investigated materials, which caused a reduction of the magnetocrystalline anisotropy.

However, in this phase 1, electric current was still needed (although being considerably reduced) in order to write the magnetic bits of information. In fact, under normal circumstances, voltage by itself cannot reverse the orientation of the magnetic moment. Nonetheless, in a second phase of the project (**phase 2**), we utilized voltage to directly generate (write) ferromagnetic (FM) signals from initially non-magnetic materials (**OFF-ON magnetic switch**), as shown in Fig. 2b. The effect was first observed in highly nanostructured Co_3O_4 films (with a high density of engineered grain boundaries), prepared by atomic layer deposition, and could be later extrapolated to other materials, such as nanoporous Fe_2O_3 (hematite) patterned dots. Remarkably, we were the first to induce this OFF-ON effect in ordered arrays of patterned structures at room temperature, which is a technological achievement towards device applications. In these oxides, the applied electric fields cause migration of oxygen ions (particularly along grain boundaries) and metallic Co (or Fe) clusters are locally generated ('magneto-ionic' effect). In this case, the information is directly recorded with the applied voltage in the different memory units (as illustrated in Fig. 1) and the energy saving during writing is extreme since no electric current is needed at all for data storage (*i.e.*, no Joule heating effect). Reading heads can then distinguish between zero ("0") remanent magnetization (written with positive voltage) and non-zero ("1") remanent magnetization (written with negative voltage). Additionally, these voltage-actuated films can be integrated into multi-layered stacks and the induced electric field-driven magnetic effects can be directly read via changes in the magnetoresistance. This is the operating principle we are now using to implement the so-called "**magnetoelectric RAM**" (ME-RAM) device. First **prototypes** are available and ready to be exploited in association with *SINGULUS Technologies*.

Some of the building blocks of our technology are oxides with antiferromagnetic (AFM) order. Due to the antiparallel spin configuration of antiferromagnets, these materials are virtually invisible from a magnetic viewpoint (absence of stray fields) and, moreover, they are very rigid to external magnetic fields, meaning that the resulting devices are very robust (*i.e.*, information cannot be deleted with strong magnets, opposite to what happens in currently available credit cards or HDDs). Since the voltage-mediated OFF-ON magnetic switching typically occurs by partially transforming AFM oxides to FM metallic states, our approach also allows to benefit from the coupling between these AFM and FM phases in order to hide/retrieve information between the two counterparts using suitable voltage protocols. This opens a new avenue for our technology, which we are exploiting with the *Fábrica Nacional de Moneda y Timbre*, in the area of **anti-counterfeiting technologies**.

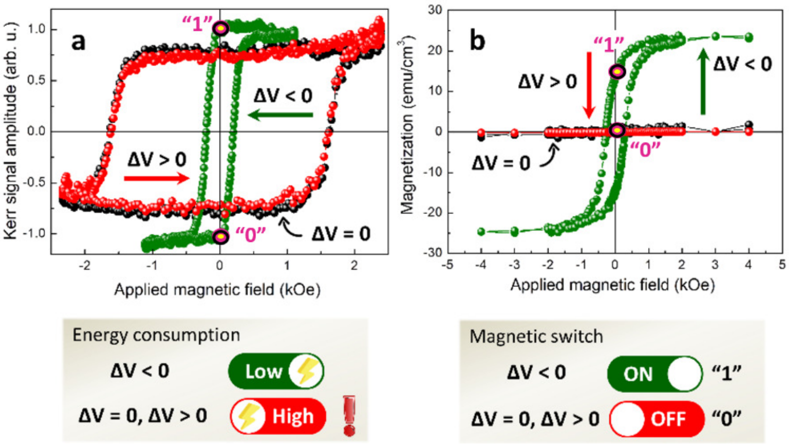

Fig. 2: (a) Reduction of coercivity observed in patterned CoPt/CoO nanoporous disks under the application of a negative voltage (the effect can be reversed and the initial large coercivity can be recovered by subjecting the sample for some time at positive voltage); (b) Generation of ferromagnetic signal from initially non-magnetic Co_3O_4 nanostructured films by application of an external negative voltage (again, the effect is reversible), so that either "1" or "0" bits of information can be selectively written with voltage, with no need of magnetic fields.

To develop the whole idea of this work, we needed to introduce several innovative aspects at all levels of the work-plan, from the synthetic pathways to grow the target materials to the final device architectures.

i) **<u>New materials</u>** have been developed, which consist of metal/semiconductor hybrid thin films and patterned structures with either a mesoporous or a highly nanostructured (*i.e.*, with engineered grain boundaries) microstructure. In metals (typically transition metal + noble metal alloys), magnetoelectric phenomena are related to spin-dependent screening (*i.e.*, electrons with different spin characters respond differently to the applied electric field) and, therefore, only occur within a few nm from the surface. This is why materials with a high surface-area-to-volume ratio are required. In oxide semiconductors, magneto-ionic effects are promoted at grain boundaries, which act as channels with enhanced ion diffusion. To grow these materials, combinations of micelle-assisted electrodeposition, wet chemistry and atomic layer deposition (sometimes reactive sputtering) have been used.

ii) **<u>New protocols</u>** have been implemented to enhance magnetoelectric effects. A greater uniformity and strength of the electric field was obtained using a capacitor design (rather than a transistor-like configuration). First results were obtained utilizing liquid electrolytes (through ultra-narrow electric double layers). Extrapolation to solid electrolytes has been done in the last year, allowing for more compact device designs.

iii) **<u>New device architectures</u>** have been developed to benefit from the observed effects. Two types of substrates are employed: (a) rigid (*e.g.*, Si) and (b) flexible, such as polyimide or P(VDF-TrFE), some of them provided by the *Fábrica Nacional de Moneda y Timbre* to optimize the integration of the voltage-driven components into the production line. The design of the devices is such that it allows inducing ON-OFF magnetic transitions in pre-selected regions of the sample (activated paths), while leaving the rest of dots unaltered. To achieve this, neuromorphic-like layouts (crossbar geometry) are prepared by lithography, establishing different paths of interconnected dots where voltage can be applied selectively. This geometry is convenient for the implementation of the ME-RAM, since it is compatible with the current device architectures of existing MRAMs. Finally, local companies (in particular, *Ingeniería Magnética Aplicada S.L.U.* from Ripollet) are also highly interested in our voltage-driven memory concept and we are working together to develop a prototype of an ultra-secure magnetic key ("*Llavor*" project from the Generalitat de Catalunya).

In summary, our work constitutes a new paradigm to boost energy efficiency in magnetic data storage and codification systems. The concept is applicable to various environments and is being implemented in electric-field-assisted magnetic recording, voltage-driven switching in ME-RAMs and a variety of anti-counterfeiting technologies (smart chips, identification cards, certificates, etc.). This versatility enhances the economic and social impact of our work, both in the areas of **energy efficiency** (where we estimate it could represent savings of several thousand M€/year worldwide in data centers, and hundreds of M€/year in home computers in Spain, with a global CO_2 emissions reduction close to 2%) and **data security** (highly protected magnetoelectric device architectures).