



Multiferroic materials and heterostructures / Matériaux et hétérostructures multiferroïques

## Artificial multiferroic heterostructures for an electric control of magnetic properties



*Hétérostructures multiferroïques artificielles pour un contrôle électrique des propriétés magnétiques*

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### ABSTRACT

The control of magnetism by electric fields is an important goal for future low-power spintronics devices. This partly explains the intensified recent interest for magnetoelectric multiferroic materials and heterostructures. The lack of ferro- or ferrimagnetic-ferroelectric materials with large magnetoelectric coupling between the two orders has spurred intensive research on artificial multiferroics combining ferroelectric or piezoelectric materials and ferromagnets. In this paper we review synthetically the potential of thin-film-based heterostructures in which a magnetic film is in contact with a ferroelectric or piezoelectric one to obtain an electric control of magnetic properties. This electric control either results from a strain-induced magnetoelectric coupling, a charge-driven one, or from the modulation of an interfacial exchange-bias interaction.

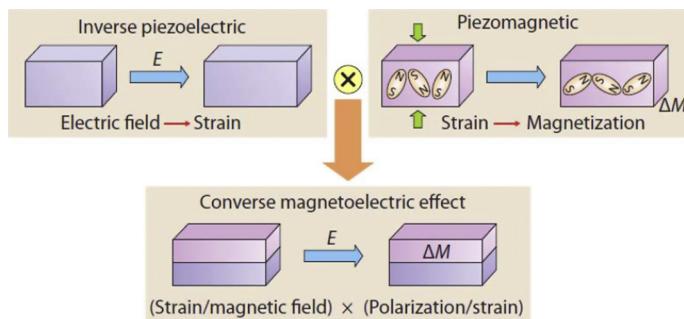
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### RÉSUMÉ

Contrôler électriquement les propriétés magnétiques des matériaux et réaliser ainsi de nouveaux composants faiblement consommateurs en énergie est un des enjeux de la future électronique de spin. Ceci explique l'intérêt considérable porté aux matériaux et architectures multiferroïques. Malgré des recherches intenses, le graal d'un composé à la fois ferroélectrique et ferro- ou ferrimagnétique à température ambiante avec un fort couplage magnétoélectrique entre ces deux propriétés n'a pas encore été trouvé. Pour pallier ce manque, de nombreux travaux ont porté sur les multiferroïques artificiels, hétérostructures combinant un matériau ferroélectrique ou piézoélectrique et un composé ferromagnétique. Cet article constitue une revue succincte du potentiel de ces hétérostructures en géométrie planaire pour obtenir un contrôle électrique des propriétés magnétiques. Un tel contrôle peut être obtenu par un effet magnétoélectrique indirect basé sur un couplage d'origine élastique, de façon directe par une modulation des charges dans le ferromagnétique induite par la ferroélectricité ou encore en exploitant le couplage d'échange à l'interface entre un multiferroïque et un matériau magnétique.

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**Fig. 1.** (Color online.) Schematic illustration of strain mediated converse magnetoelectric coupling. An electric field induces strain on the ferroelectric material which is mechanically transferred to the magnetic material leading changes in the magnetization via the piezomagnetic effect [11].

Multiferroic materials [1], after almost 40 years of hibernation, have been subject of a renewed interest in the early 2000s [2,3]. In particular magnetoelectric multiferroics presenting coupled magnetic and ferroelectric order parameters have attracted a lot of attention. From a fundamental point of view since magnetism and ferroelectricity tends to exclude each other [4], new paths have to be found to stabilize both coupled orders in the same compound. Another attractive trait of these materials is their potential applications for the next generation of electronic devices based on the control of the magnetic state by an electric field and vice versa. Such multifunctional materials could be used in solid-state transformers, magnetic field sensors or actuators. In the particular field of spintronics, they should enable a local control of magnetization to design electrically writable non-volatile magnetic memories with low-power consumption. Despite an intense activity in the field, the grail of a ferromagnetic–ferroelectric multiferroic with large magnetoelectric (ME) coupling at room temperature has not yet been found [3]. This scarcity of room temperature multiferroics with large ME coupling has spurred intensive research on composite multiferroics combining ferroelectric (FE) or piezoelectric materials and ferromagnets (FMs). These two-phase compounds have been elaborated as nanoparticles of one compound embedded in a matrix of the other compound, vertical nanopillar-based heterostructures or multilayers. Good reviews on the subject can be found in references [5–8]. Here, we will focus synthetically on the third geometry, i.e. on thin-film-based heterostructures in which a magnetic film is in contact with a FE or piezoelectric one. We will show how the electric control of magnetic properties can be achieved in such heterostructures—more extensive reviews can be found in references [9] and [10]. This electric control either results from a strain-induced ME coupling, a charge-driven ME effect, or a modulation of the interfacial exchange-bias interaction.

## 1. Strain-mediated magnetoelectric multiferroics

In strain-mediated magnetoelectric multiferroics (Fig. 1), when a magnetic film is in contact with a ferroelectric one, an external electric field yields a change in the size of the FE lattice parameter through the converse piezoelectric effect. The resulting strain induced on the magnetic material in contact, changes its magnetic properties notably its anisotropy by magnetostriction. The corresponding converse ME coupling thus results from the product of the magnetostrictive effect in the magnetic phase and the piezoelectric effect in the FE one. It is defined as [8,12]:

$$\text{Converse ME effect} = \frac{\text{Mechanical}}{\text{Electric}} \times \frac{\text{Magnetic}}{\text{Mechanical}} = \text{piezoelectric} \times \text{magnetostrictive}$$

$$\Delta M = \alpha \Delta E$$

This mechanism allows us to obtain a ME coupling, defined by the  $\alpha$  coefficient, by far larger than in single-phase multiferroics in which it is limited by the square root of the product of the magnetic and electric susceptibilities [2]. This ME coupling has been observed in a large number of systems, unfortunately, it does not generally produce two different magnetic states at electrical remanence. Indeed, the voltage dependence of the magnetic properties reflects that of the strain, as evidenced by the electric field dependence of the strain and magnetization presented in Fig. 2A and B [13].

In heterostructures combining a FE with large piezoelectric coefficients with a soft FM, large magnetostatic energy can be generated by modest voltages which results in a control of its magnetic anisotropy, i.e., its magnetization easy axis. Pertsev [16] used a phenomenological approach to predict an abrupt (progressive) rotation of  $\text{CoFe}_2\text{O}_4$  (Ni) easy-axis magnetization from in-plane to out-of-plane, and vice versa, in heterostructures combining these magnetic materials with  $\text{PbZn}_{1/3}\text{Nb}_{2/3}\text{O}_3$ – $\text{PbTiO}_3$  (PZN–PT) or  $\text{PbMg}_{1/3}\text{Nb}_{2/3}\text{O}_3$ – $\text{PbTiO}_3$  (PMN–PT) ferroelectric relaxors. Hu and Nan [17] confirmed these predictions, extended the study to other combinations of FE ( $\text{BaTiO}_3$  (BTO),  $\text{PbZrTiO}_3$  (PZT)) and FM ( $\text{Fe}$ ,  $\text{Fe}_3\text{O}_4$ ) materials, and discussed the possibility of an in-plane reorientation of the magnetization.

From an experimental point of view, most effort has been devoted to the combination of a FM, from either the transition metal or oxide families with BTO, PMN–PT or PZN–PT materials. For FM materials grown on BTO substrates, several groups reported the observation of jumps or kinks in the temperature dependence of the magnetization (see an example in Fig. 2C)

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