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Multiferroic materials and heterostructures / Matériaux et hétérostructures multiferroïques

Domains and domain walls in multiferroics

Domaines et parois de domaines dans les multiferroïques

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ABSTRACT

Multiferroics are gathering solid-state matter in which several types of orders are simultaneously allowed, as ferroelectricity, ferromagnetism (or antiferromagnetism), ferroelasticity, or ferrotoroidicity. Among all, the ferroelectric/ferromagnetic couple is the most intensively studied because of potential applications in novel low-power magnetoelectric devices. Switching of one order thanks to the other necessarily proceeds via the nucleation and growth of coupled domains. This review is an introduction to the basics of ferroelectric/ferromagnetic domain formation and to the recent microscopy techniques devoted to domains imaging, providing new insights into the archetypal multiferroic domain morphologies. Some relevant examples are also given to illustrate some of the unexpected properties of domain walls, as well as the way these domain walls can be manipulated altogether thanks to various types of magnetoelectric coupling.

RÉSUMÉ

Les multiferroïques rassemblent les solides qui permettent la cohabitation de plusieurs ordres ferroïques tels que la ferroélectricité, le ferromagnétisme (ou l'antiferromagnétisme), la ferroélasticité ou la ferrotoroïdicité. Parmi ces ordres, le couple ferroélectrique/ferromagnétique est celui qui permet d'envisager le plus directement la réalisation de dispositifs originaux dans lesquels, par exemple, l'aimantation peut être renversée par l'application d'un champ électrique, par simple application d'une tension et avec un faible coût énergétique. La manipulation d'un ordre par un autre doit opérer par nucléation et croissance de domaines nécessairement couplés. Cette revue introduit les mécanismes gouvernant la formation de ces domaines et les techniques les plus récentes pour leur observation à l'échelle microscopique, donnant ainsi accès aux morphologies les plus typiques de domaines multiferroïques. Quelques exemples parmi les plus significatifs permettront d'illustrer les propriétés spécifiques liées aux parois de ces domaines, ainsi que la façon dont ces parois peuvent être manipulées les unes par l'intermédiaire des autres grâce à différents types de couplage magnétoélectrique.

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The renewed interest in multiferroics and magnetoelectric coupling is largely associated with non-volatile memory applications. The maturity of MRAMs and FeRAMS allows devices to be on the market and their "all-in-one" magneto-electric extensions are envisaged in the same application line. The memory function is on sight and the information is conceived to be encoded thanks to the electric *and* magnetic orders. Expected devices are for example aiming at taking advantages of both ferromagnetic (FM) and ferroelectric (FE) orders without their respective drawbacks, exploiting the easy "write" of FE along with the mastered "read" of FM. The binary operation is here implicit, with complete switching of one order thanks to the other. As the switching is governed by the nucleation and the propagation of domain walls (either FE or FM, eventually coupled one with the other), these objects have promoted a flurry of activity in the scientific community. Their dynamic behavior is directly related to the switching speed and their potential pinning is a serious cause for fatigue, two key issues for applications. Moreover, functionalities can still exist even if the device is not spatially fully saturated, i.e. domains of different orientations coexist with their associated domain walls. Here again, the binary memory function can be on sight. The most archetypal example is the racetrack memory proposed by Parkin [1]: spin polarized electric currents are used to push magnetic domain walls along a permalloy wire; and each magnetic domain corresponds to a bit electrically driven to flow under the read/write elements. A wider playground is provided by FE and/or FM domains in multiferroics, allowing more exotic concepts of devices to emerge. For instance, the revival of analogic components for neuromorphic computing architectures is epitomized in the "memristor". A memristor is a variable resistance that depends on its current history and mimics the synapse function of the brain. Several memristors concepts have been explored and fully electronic examples are given by the spintronic [2] and the ferroelectric memristors [3]. In both cases, the final resistance of the FM or FE media is governed by the relative proportion of domain populations (FM or FE). The ability to tune the device resistance is related to the domain wall kinetic and domain stability. Even more original concepts can be envisaged if the device functionality is no more localized in the domains but in the domain walls. Intrinsic properties of domain walls are indeed different from the ones of the parent domains as the local symmetry at the walls is different from the bulk one. Various interfacial phenomena can arise in these nanoscale objects, leading for example to domain wall conductivity [4], magnetotransport [5], or photovoltaic response [6] (see Section 2). In multiferroic single crystals and thin films, different mechanisms can contribute to the formation of ferroelectric and magnetic domains, and a large variety of domain walls architectures has been observed. These observations are the prerequisite for novel functionalities of electronic devices. This article is focused on recently reported results about domains and domain walls in different types of multiferroic materials, especially the room temperature multiferroic $BiFeO_3$ (BFO) and rare earth hexagonal manganites RE–MnO_3 (RE = Y, Er, Ho).

1. Domain structures in multiferroic materials

Ferroics spontaneously divide into small regions of different polarity, called "domains", while the boundaries between adjacent domains are called "domain walls" or "domain boundaries". In this section, the driving force for domain formation is presented with a review of the different domain topologies reported in multiferroics.

1.1. Formation of domains

In any ferroic materials, the presence and size of domains (and therefore the density of domain walls) depends on their boundary conditions. Self-induced depolarizing fields appear when the polarization has a component perpendicular to the material's surface, and cannot be perfectly screened. In addition, residual stresses caused by epitaxy, shape anisotropy or structural defects induce domains in most multiferroics. The domain size is determined by the competition between the domains energy (increasing with size) and the energy penalty needed to create the domain walls. For instance, in ferroelectrics, smaller domains have smaller depolarizing and elastic energies, but the energy gain by reducing domain size is balanced by the increasing number of domain walls. This leads to the well-known square root dependence of the domain size as a function of the film thickness or Kittel's law [7]. Similarly in ferromagnets, uniform magnetization does not correspond to the energy optimum. The domain structure is the result of the competition between the anisotropy energy due to the spins in the domain wall (which are no longer aligned along the easy axis) and the exchange energy between domains of different magnetization. The similarities are striking, even if ferromagnetic domain sizes are generally larger than ferroelectric ones for a given thickness. The size of the domain wall itself is also different in both kinds of systems: ferroelectric domain walls are only a few nm large (from Landau–Ginzburg–Devonshire theory as well as from transmission electron microscopy observation), while magnetic domain walls are one order of magnitude larger (as determined by the balance between the exchange energy and the magnetic anisotropy). Both kinds of domain walls are nevertheless observed to be coinciding in some multiferroics and one type of domain walls can be manipulated thanks to the other type (see Section 3).

Kittel's law has been extended to ferroelastic thin films epitaxially strained [8], to ferroelectric epitaxial thin films [9–11] and to magnetoelectric multiferroics [12]. The square root dependence of domains width with film thickness appears to be a general property of ferroics and holds over a remarkable range of sizes and shapes. This global trend is observed even if growth and boundary conditions are obviously key issues. For instance, in the most studied multiferroic, namely $BiFeO_3$ with coexistence of ferroelectric and antiferromagnetic orders at room temperature, the ferroelectric domain structure can be "mosaic like" for higher growth rates or "stripe like" for lower growth rates closer to thermodynamic equilibrium (Fig. 1b