Characterization and Electro-mechanical Control of Charged Domain Walls

Caractérisation et Contrôle électro-mécanique des parois de domaines chargées

Ferroelectric (FE) materials have important applications for non-volatile memories because of their fast switching and long retention. Insulating by nature, the recent discovery of FE domain wall (DW) conduction [Seidel2009] has triggered a new era: DWs exhibit very different electronic properties than the parent materials and can be controlled (written or erased) by application of electric fields. DWs are intrinsically nano-sized objects with a thickness of few unit cells, and therefore highly scalable. Their variable conductivity opens the door to numerous applications where the electronic properties can be tuned both through the DW density and conductivity [Guyonnet2011]. The conceptual breach is based on the wall itself becoming the active element of the device.

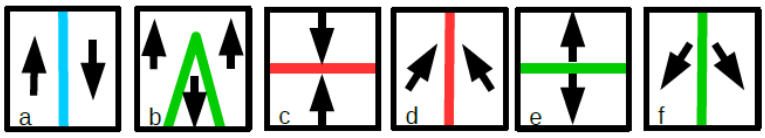

Figure 1 *(a) Neutral and (b) to (f) charged domain walls in some possible configurations. Red stands for positive charge, green for negative. Configuration (b) appears during polarization reversal. (c) and (d) Head to head (H-H) and (e) and (f) Tail to Tail (T-T) CDWs. After [Bednyakov2015]*

A comprehensive picture can be achieved by control of the DW electrostatic (H-H, T-T, H-T), ferro-elastic (strain), and chemical nature (e.g., oxygen vacancies trapped at the DW). The understanding and the technological mastering of DW properties are necessary to realize the potential for novel low power applications in electronics.

Neutral domain walls (NDW) are by far the most common DW in FEs since they easily minimize the electrostatic energy, whereas the free charge density required to screen a charged domain wall (CDW) is of the order of 10^{20} cm^{-3}, much greater than typical values in FE oxides. For this reason, CDWs are rarely observed naturally, nevertheless, they appear as a true, new paradigm for post-CMOS electronics precisely because they can be understood as nanometric metallic conductors separated by highly insulating dielectric regions. They support currents two orders of magnitude greater than NDWs [Crassous2015] and nine orders of magnitude greater than in the intervening domains [Sluka2013]. Two types of CDW can be considered: head-to-head (H-H) with large bound positive charge or tail-to-tail (T-T) which are negatively charged.

Reversibly and rapidly displacing CDWs by external stimuli would represent an important step forward for viable electronic architectures. A first experimental demonstration of control of a CDW reported in *Science Advances* [Sharma2017] showed that resistance of a multistate FE memory depends on the CDW length. The thesis work aims to demonstrate reversible electrical or mechanical control of the DW position. The work will be carried out within the framework of a collaboration between the CEA, the UMPhys CNRS/Thalès and the ICMMO laboratory (Université Paris Sud).

Both thin films and single crystals will be studied, providing results relevant to a wide range of applications. The work will be carried out in three stages:

- **Controlled growth** of single-crystals and thin films with high-density arrays of DWs and stabilization of CDWs by trailing field PFM and by frustrated poling.
- **Static characterization** of the chemistry and electronic structure of FE DWs.
- **Operando analysis of DW motion** under electrical/mechanical stress using state-of-the-art microscopy techniques.

UMPhy will prepare the epitaxial growth by pulsed laser deposition of the $BiFeO_3$ thin films and engineer CDWs using trailing field conducting tip atomic force microscopy (C-AFM) as well as complementary piezo-response analysis. ICMMO will provide high quality single crystal growth under electric field, necessary for the frustrated poling, as well as structural characterization techniques. The analysis of the electronic structure and surface chemistry, bringing expertise in operando analysis under electric field or mechanical stress will be carried out at the CEA where PhD student will be based.

Low Energy Electron Microscopy (LEEM) will measure the electrostatic surface potential across the DW. Photoelectron Emission Microscopy (PEEM) will map the local chemistry and electronic structure [Barrett2013].

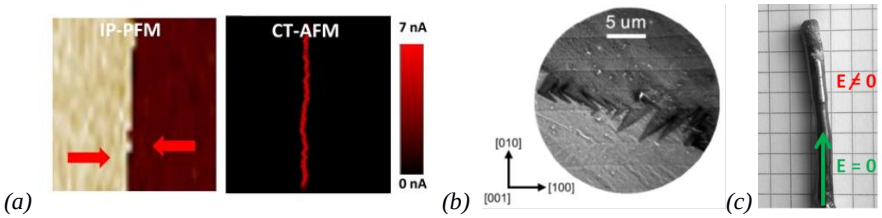

Figure 2 *(a) In plane phase image of two domains separated by a H-H 71° CDW prepared by trailing field AFM and current map showing enhanced conductivity with a maximum level of about 5 nA @ V_{SRO}=2 (UMPhys). (b) LEEM image of H-H DWs during in-plane polarization switching in $BaTiO_3$ (CEA) [Rault2014] and (c) BTO single crystal obtained by frustrated poling (ICMMO)*

Mapping of FE domains will be performed by Piezoresponse Force Microscopy (PFM) and C-AFM will measure the local electrical properties of DWs.

Different DWs configurations (H-T, H-H, T-T), environments (ambient or UHV), chemical doping levels (oxygen vacancies) or static strain levels (epitaxial thin films on different substrates) will be assessed.

In-situ near (PFM, C-AFM) and full (LEEM, PEEM) field microscopy [Barrett2016] will be carried out to image the switching process under mechanical stress (Fig. 3(a)).
Using clean room facilities, specific control electrodes will apply an electric field across the sample to stimulate CDW migration whilst read electrodes will determine the resistance state of the primitive device as shown in Fig. 3(b).

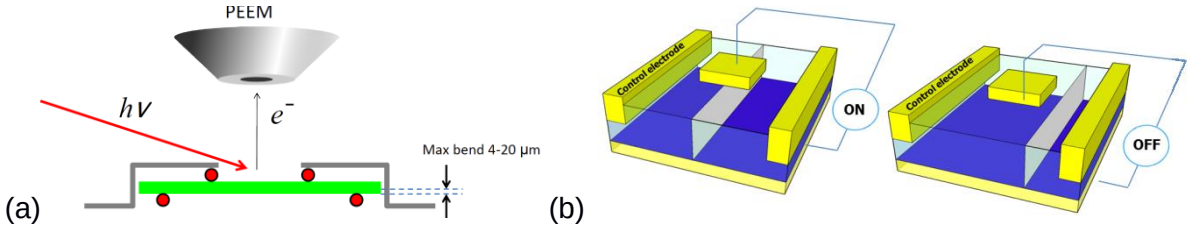

Figure 3 *(a) 4-point piezo-driven jig for mechanical control of DW (b) electrical control of CDW movement.*

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