# Ferroelectric Domain Walls

Tel Aviv, 13-15 Nov 2017

cecam
Centre Européen de Calcul Atomique et Moléculaire

קרן לאומית למדע
ISRAEL SCIENCE FOUNDATION

TEL AVIV UNIVERSITY

# Ferroelectric Domain Walls

## Tel Aviv, 13-15 Nov 2017

# Program and Scientific Abstracts

MONDAY
NOVEMBER
13TH

| | |
|---|---|
| *08:30–09:00* | *Registration and Light Breakfast* |
| *09:00–09:40* | **David Vanderbilt:** *Ferroelectric reversal and domain walls in corundum derivatives* |
| *09:40–10:20* | **Andrew Rappe:** *Intrinsic ferroelectric domain wall motion revealed by first-principles based molecular dynamics* |
| *10:20–10:50* | *Coffee Break* |
| *10:50–11:30* | **Mael Guennou:** *Imaging Domain walls by Raman scattering and low energy electron microscopy* |
| *11:30–12:10* | **Pavlo Zubko:** *Imaging ferroelectric domain structures with X-ray nanodiffraction* |
| *12:10–12:50* | **Yachin Ivry:** *Unscreening screening: from disappearing barium atoms to blooming nanodomains* |
| *12:50–14:00* | *Lunch* |
| *14:00–14:40* | **Maxim Mostovoy:** *Ferroelectric properties of magnetic skyrmions and merons* |
| *14:40–15:20* | **Sergey Prosandeev:** *Unusual phenomena induced by time-dependent toroidal moments* |
| *15:20–16:00* | **Sang-Wook Cheong:** *Topological Vortex Domains in Hybrid Improper Ferroelectricity* |
| *16:00–16:30* | *Coffee Break* |
| *16:30–17:10* | **Javier Junquera:** *Second-principles simulations of counter-rotating vortices pairs in $PbTiO_3/SrTiO_3$ superlattices* |
| *17:10–17:50* | **Ramamoorthy Ramesh:** *Emergent chirality and phase coexistence in polar vortices formed in oxide superlattices (videoconference)* |
| evening | Banquet |

| | | |
|---|---|---|
| TUESDAY NOVEMBER 14TH | 08:30–09:00 | *Light Breakfast* |
| | 09:00–09:40 | **Marin Alexe:**<br>*Photoelectric processes in $BiFeO_3$ domain walls* |
| | 09:40–10:20 | **Jirka Hlinka:** *Terahertz-range polar response of 71 degree wall of bismuth ferrite* |
| | 10:20–10:50 | *Coffee Break* |
| | 10:50–11:30 | **Marty Gregg:** *Mapping Carrier Response in Conducting Ferroelectric Domain Walls* |
| | 11:30–12:10 | **Semën Gorfman:** *Time-resolved X-ray diffraction studies of extrinsic and intrinsic mechanisms of piezoelectricity in ferroelectrics* |
| | 12:10–12:50 | **Dennis Meier:** *Emulating the functionality of electronic components using domain walls* |
| | 12:50–14:00 | *Lunch* |
| | 14:00–14:40 | **Beena Kalisky:** *Probing $SrTiO_3$ domain walls with scanning SQUID microscopy* |
| | 14:40–15:20 | **Massimiliano Stengel:** *Macroscopic polarization from antiferrodistortive cycloids in ferroelastic $SrTiO_3$* |
| | 15:20–16:00 | **Yoram Dagan:** *Turning superconductivity and spin-orbit interaction across the phase diagram of (111) $LaAlO_3/SrTiO_3$* |
| | 16:00–16:30 | *Coffee Break* |
| | 16:30–17:10 | **Sergey Artyukhin:** *Low-energy structural dynamics of ferroelectric domain walls in hexagonal rare-earth manganites* |
| | 17:10–17:50 | **Andrés Cano:** *Formation and inner structure of topological defects in hexagonal manganites* |

| | | |
|---|---|---|
| WEDNESDAY NOVEMBER 15TH | 08:30–09:00 | *Light Breakfast* |
| | 09:00–09:40 | **Igor Luk'yanchuk:**<br>*Ferroelectric multibit cells* |
| | 09:40–10:20 | **Jan Seidel:**<br>*Domain walls and phase boundaries for memory applications* |
| | 10:20–10:50 | *Coffee Break* |
| | 10:50–11:30 | **Jorge Iniguez:**<br>*Electric skyrmions* |
| | 11:30–12:10 | **Patricjya Paruch:** *Novel functional properties at ferroelectric domain walls: insights from scanning probe microscopy* |
| | 12:10–12:50 | **Nava Setter:** *Insight into structure, properties, and mobility of ferroelectric domain walls* |
| | 12:50–14:00 | *Lunch* |

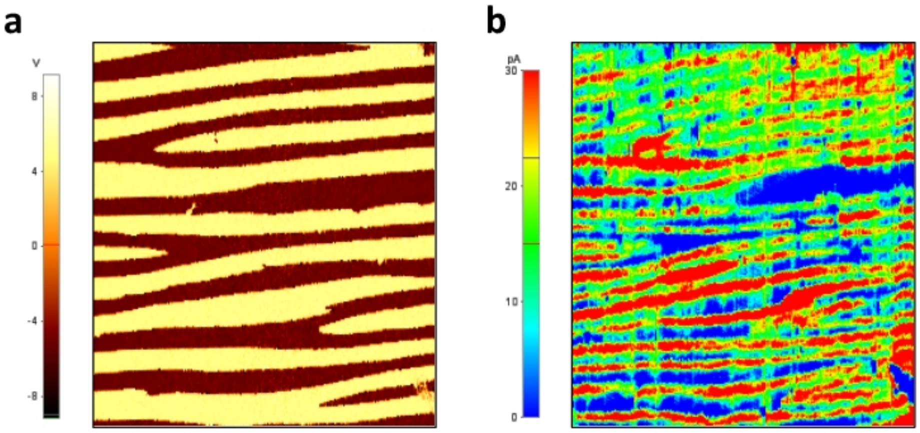


***Figure**:*
*Spatially resolved imaging of (a) 71° domain configuration and (b) photoconductive current measured with a 5 V bias under illumination.*

**Marin Alexe**

*Department of Physics,*
*University of Warwick,*
*Coventry, CV4 7AL (UK)*

# Photoelectric Processes in BiFeO$_3$ Domain Walls

In the recent past, the field of photo-ferroelectrics has been revitalized by the reports of photovoltaic (PV) effect in BiFeO$_3$ (BFO)[1]. Until now, the electronic properties of DWs under illumination has been studied mostly by macroscopic characterisation techniques and a direct correlation remain elusive so far. Here, we present local investigations of photoelectric properties using atomic force microscopy system which is able to characterize local photoelectric and photovoltaic effects with a nanometre resolution (Ph-AFM). We show detailed analysis of photoconductivity of DWs in BFO and their roles in the APV effect in ferroelectric semiconductors[2]. Furthermore, a novel time-resolved spectroscopic scanning probe method, i.e. photo-induced transient spectroscopy scanning probe microscopy (PITS-SPM)[3], was used to map important photoelectric process, such as quantum yield and lifetime of non-equilibrium carriers, in nanoscale photoactive entities. Based on the PITS-SPM system, we mapped with nanoscale resolution the quantum yield and carrier lifetime in domain walls, providing a deep insight into the origin of their enhanced photoconductivity.

[1] S. Y. Yang, et. al., Nat. Nanotechnol. 5, 143 (2010).
[2] M.M. Yang, A. Bhatnagar, Z.D. Luo, M. Alexe, Scientific Reports 7, 43070 (2017).
[3] M. Alexe, Nano Letters, 12, 2193 (2012).

***Collaborators:***
*Ming-Ming Yang*

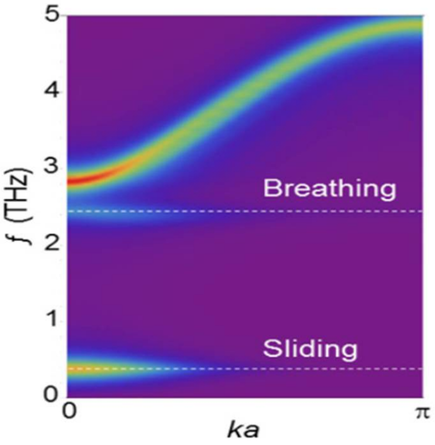


***Figure***: *A phonon spectral function A(w,k) for the wavevector in the direction, perpendicular to the plane of the domain wall, computed within a simplified 1D model. In addition to a dispersive phonon mode (top), two domain wall-localized modes are seen. The lowest energy mode corresponds to oscillations of DW plane, while the mode at the bottom of the phonon band is a breathing mode, corresponding to oscillations of DW width.*

**Sergey Artyukhin**

*Quantum Materials Theory,*
*Istituto Italiano di*
*Tecnologia, Genova (Italy)*

# Low-energy structural dynamics of ferroelectric domain walls in hexagonal rare-earth manganites

Domain walls (DW) in ferroic materials are 2D systems whose vibrational properties and electronic structure may be very different from that of the bulk. We report on the electric field - driven dynamics of ferroelectric domain walls in hexagonal manganites, where ferroelectric polarization is produced due to condensation of a trimerization mode[1,2]. Recent broadband (106 to 1010 Hz) scanning impedance microscopy measurements reveal that the electrical response of the DWs in hexagonal rare-earth manganites (h-$RMnO_3$) is dominated by the bound-charge oscillation rather than free-carrier conduction at the DWs[3]. As a measure of the rate of energy dissipation, the effective conductivity of DWs on the (001) surfaces of h-$RMnO_3$ at gigahertz frequencies is drastically higher than that at dc, whereas the effect is absent on surfaces with in-plane polarized domains. Using model Hamiltonian with parameters extracted from first-principles simulations, we compute vibrational modes of a supercell containing domain walls. In addition to bulk phonons, we find domain wall-localized modes, the lowest of which at around 30 GHz corresponds to oscillations of the DW plane around its equilibrium position. The domain wall plane effectively behaves as a particle in a washboard-like potential, whose curvature determines the frequency of the vibration. This mode has no dispersion perpendicular to the DW, as shown in the Figure, but disperses in the plane of the domain wall, so that DW is acting as a waveguide for that mode. The results agree with experimental observations, suggesting that the lowest DW-localized mode contributes to GHz loss in ferroelectrics. The frequencies of higher-energy localized modes found in simulations are located in the THz range at the bottom of the corresponding bulk phonon bands. Calculations suggest that the frequency of the lowest DW-localized mode decreases exponentially with the increase of domain wall width, and becomes zero for domain walls much wider than the lattice constant, thus recovering the gapless domain wall translation mode in the continuum limit. Since the electric polarization in proper ferroelectrics is much larger than in improper ferroelectric hexagonal manganites, the piezoelectric effect is expected to be important there, giving rise to a strong coupling of the DW-localized mode with surface acoustic waves. These observations advance the understanding of loss and structural dynamics in ferroelectrics and break the ground for novel devices.

[1]M. Mostovoy, Nature Materials 9, 188 (2010).

[2] S. Artyukhin, K.T. Delaney, N.A. Spaldin, and M. Mostovoy, Nature Materials 13, 42 (2014).

[3] X. Wu, U. Petralanda, L. Zheng, Y. Ren, R. Hu, S.-W. Cheong, S. Artyukhin, K. Lai, Science Advances 3, e1602371 (2017).

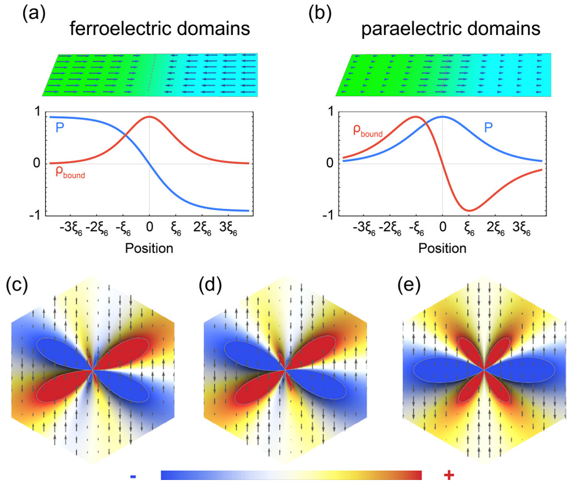


***Figure**: Emergent electrostatics at trimerization domain walls and vortices. (a,b) Trimerization domain wall separating ferroelectric (a) and nonferroelectric (b) domains. The arrows in the top panels represent the electric polarization resulting from the trimerization. In both cases, we observe the emergence of a nonzero distribution of bound charges localized at the wall due to the longitudinal variation of the polarization across the wall (bottom panels). (c–e) Density plot of the bound-charge distribution emerging at the core of the trimerization vortices.*

**Andrés Cano**
*CNRS, Univ. Bordeaux, ICMCB, Bordeaux (France) and Department of Materials, ETH Zurich, 8093 Zürich (Switzerland)*

# Formation and inner structure of topological defects in hexagonal manganites

Ferroelectric domain walls in the hexagonal manganites $RMnO_3$ are the by-product of "trimerization" domain boundaries and vortices. These topological defects display intriguing functional features related to their local transport properties and can be regarded as analog systems for the study of the formation of cosmic strings in the early universe. I will discuss a global Kibble-Zurek mechanism that can explain the formation of these defects[1] and their inner atomic structure as revealed by means of STEM experiments combined with a field-theory description[2]. Our theory reveals novel features such as the emergence of a continuous U(1) symmetry at the vortex cores and a multipolar charge (re-)distribution that become universal in these regions. These inner features are expected to bring forth an additional degree of functionality to these topological defects.

[1] Global formation of topological defects in the multiferroic hexagonal manganites, Q. N. Meier, M. Lilienblum, S. M. Griffin, K. Conder, E. Pomjakushina, Z. Yan, E. Bourret, D. Meier, F. Lichtenberg, E. K. H. Salje, N. A. Spaldin, M. Fiebig, and A. Cano, Phys. Rev. X (2017); arXiv:1703.08321

[2] Topological defects in hexagonal manganites - Inner structure and emergent electrostatics, M.E. Holtz, K. Shapovalov, J.A. Mundy, C.S. Chang, Z. Yan, E. Bourret, D.A. Muller, D. Meier, and A. Cano, Nano Letters (2017).

**Sang-Wook Cheong**

*Rutgers University (USA)*

# Topological Vortex Domains in Hybrid Improper Ferroelectricity

Hybrid improper ferroelectricity (HIF), which describes a state with the polarization induced by a hybridization of two non-polar lattice instabilities, holds great promise toward the realization of room-temperature multiferroelectricity. The key idea is to design new materials in which ferroelectricity and (anti)ferromagnetism can be coupled by the same lattice instability, therefore providing an indirect but strong coupling between polarization and magnetism. Exemplary compounds with HIF include the double-layered Ruddlesden-Popper (RP) perovskites with the chemical formula of $A_3B_2O_7$ ($A^{2+}$ = alkali metal; $B^{4+}$ = transition metal), $PbTiO_3/SrTiO_3$ superlattice and $AA'B_2O_6$ double perovskites. In addition, the presence of related polar space groups has been reported in Dion-Jacobson compounds $ABiNb_2O_7$ (A = Rb, Cs), RP-phase ferrite composites and $Ca_3Mn_2O_7$.

We have, for the first time, grown single crystals of $(Ca,Sr)_3Ti(Mn)_2O_7$ and $Sr_3Sn_2O_7$, and experimentally confirmed the existence of hybrid improper ferroelectricity in the crystals. Furthermore, we found that charged ferroelectric domain walls, some of which are highly conducting, are mysteriously abundant in $(Ca,Sr)_3Ti_2O_7$ crystals. Our careful examination of the domains and domain walls of $(Ca,Sr)_3Ti_2O_7$ has led to unprecedented discoveries: a good fraction of domain walls are Neel-type, and large-range configurations of domains and domain walls are, in fact, consistent with topological $Z_4$ vortex domains. The abundance of charged walls in $(Ca,Sr)_3Ti_2O_7$ results from the $Z_4$ vortex domain configurations. Our results on a number of HIF materials pave a new avenue to design new ferroelectrics with enhanced functionalities and also novel multiferroics with room-temperature magnetoelectricty.

**Yoram Dagan**

*School of Physics and Astronomy, Tel Aviv University, 69978, Tel Aviv (Israel)*

# Tuning superconductivity and spin-orbit interaction across the phase diagram of (111) $LaAlO_3/SrTiO_3$ interface

The two dimensional electron liquid formed at the (111) interface between $SrTiO_3$ and $LaAlO_3$ is a laboratory for studying electronic properties in tunable correlated hexagonal systems. Symmetry changes imposed by the interface and by various structural transitions in the bulk can affect the electronic properties at the interface. In addition, this system can be smoothly tuned from the superconductor to the insulator regime.

The normal state properties of the (111) $LaAlO_3/SrTiO_3$ interface are indicative of contributions from electron-type and hole-type charge carriers. The latter are also consistent with the polar structure of this interface. Upon applying gate voltage to add electrons, a band with a higher spin state gets populated, resulting in a six-fold anisotropic magnetoresistance[1]. Superconductivity is observed in a dome-shaped region in the carrier density – temperature phase diagram. The upper critical field is strongly anisotropic and exceeds the Clogston-Chandrasekhar limit. This suggests strong spin-orbit interaction $\mathbf{e}_{so}$. Surprisingly, $\mathbf{e}_{so}$ is also nonmonotonic as a function of gate voltage as found both from analysis of the superconducting properties and of the weak antilocalization measurements[2].

Finally, the nature of the transition from superconductor to insulator will be discussed in the framework of a quantum phase transitions in an electronically disordered two-dimensional superconducting system[3].

[1] P.K. Rout, I. Agireen, E. Maniv, M. Goldstein, Y. Dagan Physical Review B 95 (24), 241107.

[2] P.K. Rout, E. Maniv, Y. Dagan, arXiv preprint arXiv:1706.01717.

[3] M. Mograbi et al., to be published.

.

**Semën Gorfman**

*Department of Materials Science and Engineering, Tel Aviv University (Israel)*

# Time-resolved X-ray diffraction studies of extrinsic and intrinsic mechanisms of piezoelectricity in ferroelectrics

Such technologically important properties of ferroelectrics as giant piezoelectricity and enhanced dielectricity are mediated by at least two different mechanisms. One is purely “intrinsic”: it accounts for any changes of long-range atomic structure (shift of atomic positions in the unit cell and lattice parameters) in response to external perturbation (i.e. external electric field). Another is “extrinsic”: it described a motion of domain walls, separating differently oriented strain domains and changing the volumetric ratios between them. The comparative significance of “intrinsic” and “extrinsic” mechanisms as well as their dependence on the chemical composition, average and local symmetry remains debated and poorly understood because of the lack of appropriate measurement techniques.

Here, we introduce high-resolution and time-resolved X-ray diffraction to assess “intrinsic” and “extrinsic” contributions to the electromechanical coupling in ferroelectrics. We developed stroboscopic X-ray data-acquisition system to collect diffraction intensity as a function of time and dynamically applied electric field (see e.g. [1] for more details). We measure and analyse in-situ X-ray diffraction signal (in the form of rocking curves, reciprocal space maps and powder diffraction profiles) simultaneously with the macroscopic hysteresis loop as demonstrated in the example (Figure 1). The angular positions of diffraction peaks are used to calculate the field dependence of the bulk lattice parameters, the profile shapes and the exchange of intensities between different Bragg peaks components are used to measure change of a microstructure. We will discuss the origin of piezoelectricity in single crystals of $Sr_{0.5}Ba_{0.5}Nb_2O_6$ (SBN) and $Na_{0.5}Bi_{0.5}TiO_3$ (NBT). Owing to the fundamental differences of their structures and symmetries, these materials have completely different type and topology of domains patterns. The symmetry of SBN rules out the formation of ferroelastic, keeping 180° domain only. However, despite the absence of ferroelastic domains we observed the strong “domains”-related mechanisms of piezoelectricity. The monoclinic symmetry of NBT promotes the formation of extremely complex and flexible domains with rotatable domain walls directions. However, despite expected presence of strongly flexible domain pattern, we observe the strong “intrinsic” contribution to the electromechanical response. This type of response will be discussed in terms of the model of polarization rotation.

[1] Gorfman, S. Sub-microsecond X-ray crystallography: techniques, challenges and applications for materials science. Crystallogr. Rev. 20, 210–232 (2014).

[2]Gorfman, S. et al. Time-Resolved X-Ray Diffraction Reveals the Hidden Mechanism of High Piezoelectric Activity in a Uniaxial Ferroelectric. Phys. Rev. Lett. 114, 97601 (2015).

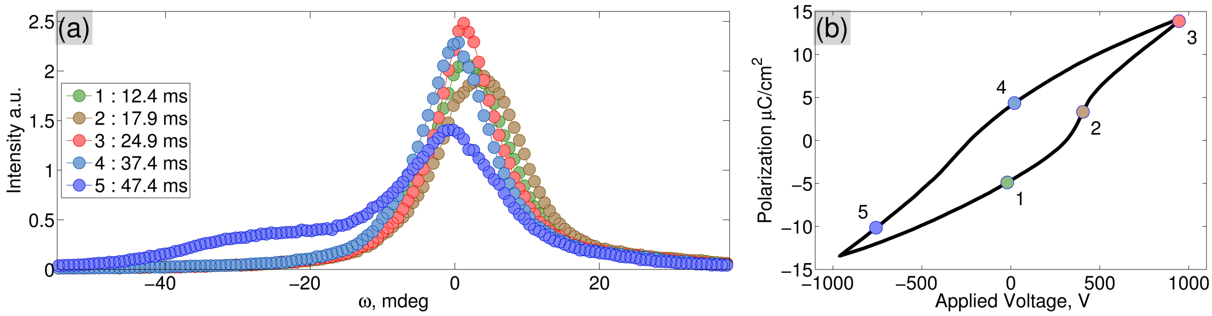


***Figure**: The example of stroboscopic X-ray diffraction study of ferroelectric single crystal of $Sr_{0.5}Ba_{0.5}Nb_2O_6$ under AC (20 Hz) cyclic electric field. (a) Bragg rocking curves of 0 0 7 reflection, collected at the different points on the P-E hysteresis loop. (b) P-E field hysteresis loop where the states, corresponding to the diffraction curves in (a) are pinpointed. The cyclic change of the position, width and line shape of the Bragg profile reflect the changes of the domain microstructure.*

**Mael Guennou**

*Materials Research and Technology department, Luxembourg Institute of Science and Technology, 41 rue du Brill, 4422 Belvaux (Luxembourg)*

# Imaging Domain walls by Raman scattering and low energy electron microscopy

As a sensitive structural probe, Raman spectroscopy has played a pivotal role in the study and understanding of ferroic phase transitions, especially through the measurements of associated soft modes. With the widespread use of micro-Raman spectroscopy, it has also become a well established technique for characterization of domain structures down to the micron scale. Its use for investigation of ferroic domains walls is therefore natural, but remains challenging because of the small size of domain walls which usually lies well below the diffraction limit.

In spite of this fundamental limitation, domains walls usually show up clearly in Raman maps, due to minute changes in the Raman spectrum in the vicinity of the wall. Here, we show how the analysis benefits from the use of Principal component analysis (PCA) in order to extract the domain wall signature against the signals of the ferroelectric, ferroelastic or anti-phase domains[1]. It is shown that PCA allows to quickly and reliably identify small Raman peak variations at ferroelectric DWs. The method proves particularly fruitful in the study of ferroelectric domain walls of uniaxial $LiNbO_3$. By analysing shifts in Raman modes in a series of Mg-doped crystals with several concentrations, we show how defects are stabilized at domain walls and contribute to the Raman contrast in combination with strain effects[2].

Low-energy electron microscopy (LEEM), on the other hand, is a complementary technique sensitive to the surface potential of a sample. Here, we use it to study the polar character of ferroelastic domain walls in the prototypic $CaTiO_3$[3]. The Figure shows images acquired in the MEM and LEEM regimes, and MEM-LEEM transition curves in domains and at domain walls. As expected, adjacent domains present the same surface potential because the material is non-polar. However, the twin walls show clear surface potential contrast with respect to the domains. This contrast is not correlated to surface topography, and therefore has to include an electric response of the wall. This provides direct in-situ evidence of the polar nature of ferroelastic twin walls in $CaTiO_3$.

We also study the manipulation of the surface charge at twin walls upon electron injection, realized by increasing the energy of the

***Collaborators:***

*G. F. Nataf*
*J. Kreisel*
*P. Hicher*
*R. Haumont*
*A. Oktay*
*E.K.H. Salje*
*L. Tortech*
*C. Mathieu*
*D. Martinotti*
*N. Barrett*

***Funding:***

*Luxembourg National Research Fund (FNR) under project CO-FERMAT FNR/P12/4853155 /Kreisel.*

incoming electron beam of the LEEM. We show that the contrast between domain and domain walls is modified upon electron irradiation with a characteristic time of 10-15 minutes and in some cases even vanishes. It is possible to recover the initial state, i.e. before charge injection, by annealing above 250°C. The ability to observe polarity at the nanoscale and to tune the surface charge at twin walls gives perspectives toward functionalization of polar twin walls in $CaTiO_3$.

[1] G. Nataf et al., submitted 2017.
[2] G. Nataf, M. Guennou, A. Haußmann, N. Barrett, and J. Kreisel, Phys. Status Solidi RRL 10, 222 (2016).
[3] G. Nataf et al., submitted 2017.

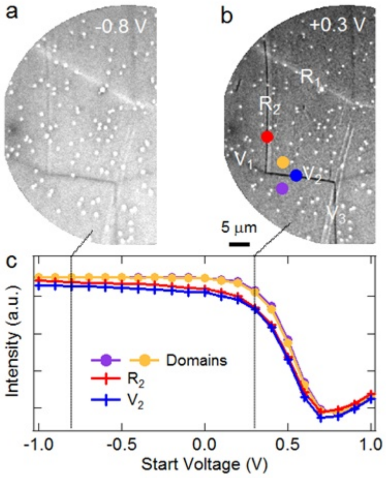


***Figure**: (a) and (b) LEEM images of a $CaTiO_3$ single crystal showing contrasts attributed to the polarity of the domain walls. (c) MEM-LEEM transition curves revealing the differences in surface potential in the domains and at domain walls.*

**Jiri Hlinka**
*Institute of Physics, The Czech Academy of Sciences, Na Slovance 2, 182 21 Prague 8, Czech Republic (Czech Republic)*

# Terahertz-range polar response of 71 degree wall of bismuth ferrite

Our recent theoretical investigation of dielectric permittivity and properties of electrically active lattice resonances in nanotwinned bismuth ferrite crystals using an earlier established interatomic potential[1] will be displayed in detail. The results suggest that an array of 71 degree domain walls with about 2-5 nm spacing enhances the static permittivity of bismuth ferrite by more than an order of magnitude[2]. This somewhat accidentally encountered enhancement is associated with an electrically active excitation, corresponding to a collective vibration of pinned domain walls at a remarkably high frequency of about 0.3 THz[2]. The origin of the excitation is discussed in terms of domain wall dynamics. The role of all possible lattice degrees of freedom of the domain wall is also elucidated systematically, (see Figure, referring to the Cartesian coordinates of the symmetry-adapted simulations supercell). This finding opens several questions, for example about the relation to the phenomenology developed for incommensurate ferroelectrics[3-5] or to the theoretically predicted Bloch-to-Ising phase transition in some ferroelectric domain walls[6-9]. We shall try to cover these subjects as well as to discuss the possible experimental issues.

***Collaborators:***
*Marek Pasciak*
*Sabine Koerbel*
*Pavel Marton*

[1] M. Graf, M. Sepliarsky, S. Tinte, and M. G. Stachiotti, Phys. Rev. B 90, 184108 (2014).
[2] J. Hlinka, M. Pasciak, S. Körbel, P. Marton, Phys. Rev. Lett. 119, 057604 (2017).
[3] R. Currat and T. Jansen, Solid State Physics 41 (Academic Press, New York, 1988).
[4] J. Hlinka, M. Quilichini, R. Currat, and J.F. Legrand, J. Phys. Condens. Matter 9, 1461 (1997).
[5] J. Hlinka, J. Petzelt, B. Brezina, and R. Currat, Phys. Rev. B 66, 132302 (2002).
[6] P. Marton, I. Rychetsky, and J. Hlinka, Phys. Rev. B 81, 144125 (2010).
[7] V. Stepkova, P. Marton, and J. Hlinka, J. Phys. Condens. Matter 24, 212201 (2012).
[8] P. V. Yudin, A. K. Tagantsev, E. A. Eliseev, A. N. Morozovska, and N. Setter, Phys. Rev. B 86, 134102 (2012).
[9] P. Marton, V. Stepkova, and J. Hlinka, Phase Transitions 86, 103 (2013).

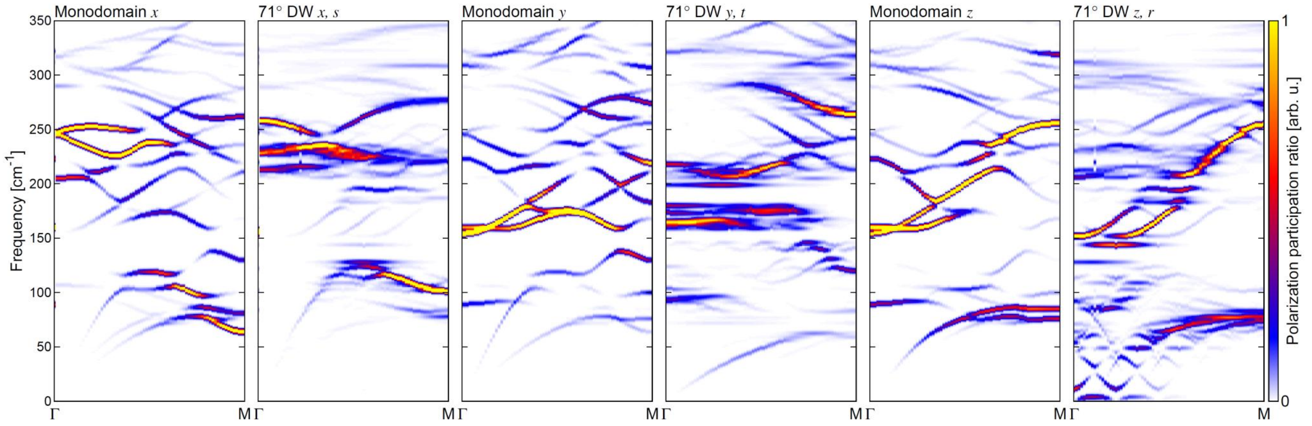


***Figure**:*
*Contribution of the selected phonon modes of the paraelectric reference cubic phase into (from left to right):*
*z- polarized modes of the single domain rhombohedral state;*
*the x- polarized modes of the supercell with 71 degree domain walls,*
*the y- polarized modes of the single domain rhombohedral state;*
*the y- polarized modes of the supercell with 71 degree domain walls,*
*the z- polarized modes of the single domain rhombohedral state;*
*the z- polarized modes of the supercell with 71 degree domain walls.*
*Only the utmost right panel includes the particularly strongly IR active mode in the sub-THz frequency range..*

**Jorge Íñiguez**
*Materials Research and Technology Department, LIST, 5 avenue des Hauts-Fourneaux, L-4362 Esch/Alzette (Luxembourg)*

# Electric Skyrmions

**Collaborators:**
*M.A.P. Gonçalves*
*C. Escorihuela-Sayalero*
*J.C. Wojdel*
*J. Junquera*
*P. García-Fernández*

***Acknowledgements:***
*Work at LIST funded by the Luxembourg National Reserach Fund through the CORE (Grant C15/MS/10458889 NEWALLS) and AFR (Grant No. 9934186) programs.*

I will present our latest theoretical predictions on how to control the properties of functional oxides, and even induce completely new behaviors, by appropriately engineering their nano-structure, domains or domain walls. In paricular, I will focus on our recent efforts to stabilize dipole arrangements that have the topological features of a skyrmion. We manage to do this by taking advantage of the Bloch character of ferroelectric domain walls in prototype compound $PbTiO_3$. The resulting skyrmions display novel features that are not present in their magnetic counterparts..

**Yachim Ivry**
*Department of Materials Science and Technology & Solid State Institute, Technion – Israel Institute of Technology, Haifa, 3200003 (Israel)*

# Unscreening screening: from disappearing barium atoms to blooming nano domains

***Collaborators:***
*Asaf Hershkovitz*
*Maya Barzilay*
*Cecile Saguy*
*Itamar Holzman*
*Alon Hendler-Avidor*
*Florian Johann*

***Acknowledgements:***
*We thank the Horev Fellowship for Leadership in Science and Technology, supported by the Taub Foundation, the Zuckerman STEM Leadership Program as well as the Technion's Russel Berrie Nanoscience Institute and Grand Energy Program for supporting our work.*

How does the surface affect ferroic domain walls? Which types of domain walls dominate- ferroelectric of ferroelastic? To answer these questions, we examined carefully the interplay between elastic and electric domains in the classic materials $BaTiO_3$ and PZT at a scale ranging from atomic-scale dipoles to nano domains under variable environmental conditions. Our observations reveal that the surface of a ferroelectric plays a significant role in stabilizing the ferroelectric and ferroelastic domain walls, hence paving the way for a novel methodology for domain-wall engineering.

**Javier Junquera**
*Departamento de Ciencias de la Tierra y Física de la Materia Condensada, Universidad de Cantabria, Cantabria Campus Internacional, Avenida de los Castros s/n, E-39005, Santander (Spain)*

# Second-principles simulations of counter-rotating vortices pairs in $PbTiO_3/SrTiO_3$ superlattices

When ultrathin ferroelectric layers of $PbTiO_3$ are embedded in superlattices with a paraelectric material, such as $SrTiO_3$, the interplay between elastic, electrostatic, and gradient energies produces complex patterns of the electrical polarization. In particular, nanometer scale of clock- and counterclock-wise rotating vortices arrays have been recently detected[1] and exotic properties such as the emergence of a negative capacitance have been measured[2]. In this work we carry out atomistic simulations to determine the properties of these emergent structures.

Performing predictive simulations in these systems is difficult due to the long spatial scales involved in the formation of counter-rotating vortices pairs, the strong competition between a large number of phases and the sensitivity of the results to external perturbations like strain, periodicity, temperature or electric fields. In order to overcome these problems we employ a recently developed second-principles method [3] that can cope with all the degrees of freedom associated to a large number of atoms retaining high accuracy. Our simulations predict the existence of several quasi-degenerate phases at low energies each displaying different properties including net polarization, non-null topological constants and chirality. The later prediction supports the findings of optical activity in x-ray circular dichroism experiments. Moreover, depending on the periodicity of the superlattice these chiral vortex phases coexist with ferroelectric phases and reversible phase transitions can be induced by external electric fields[4].

[1] A. K. Yadav et al., Nature 530, 198 (2016).
[2] P. Zubko et al., Nature 534, 524 (2016).
[3] J. Wojdel et al. J. Phys.: Condens. Matter. 25, 305401 (2013).
[4] A. R. Damodaran et al., Nature Materials (2017), https://www.nature.com/nmat/journal/vaop/ncurrent/full/nmat4951.html .

***Collaborators:***
*Pablo Aguado-Puente*
*Jorge Íñiguez*
*Pablo García-Fernández*

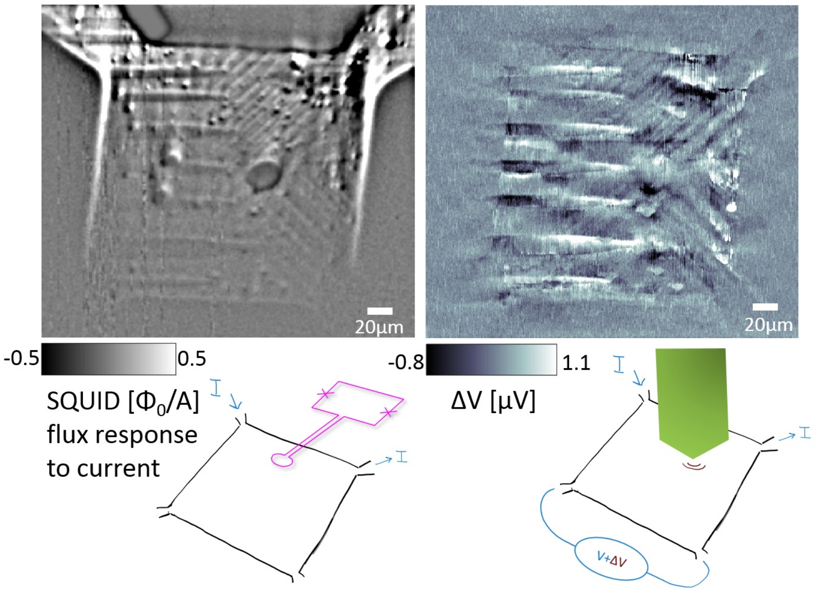
***Funding:***
*Financial support from the Spanish Ministry of Economy and Competitiveness through grant number FIS2015-64886-C5-2-P.*

**Beena Kalisky**

*Department of Physics and Institute of Nanotechnology and Advanced Materials, Bar-Ilan University (Israel)*

# Probing $SrTiO_3$ domain walls with scanning SQUID microscopy

The interface formed by growing $LaAlO_3$ on $SrTiO_3$, both non magnetic insulators, exhibits conductivity, superconductivity and even magnetism. It is not surprising that the symmetry of the $SrTiO_3$ substrate is a dominant player in the plethora of physical phenomena found at the interface. We first encountered the interplay between the $SrTiO_3$ ferroelastic domain walls and the interface while imaging the magnetic flux generated from the interfacial current flow. We found that a big part of the current can be modulated over the $SrTiO_3$ domain walls and that macroscopic transport measurements are strongly affected. We then turned to investigate the origin of the modulations. In order to do so we applied local stress to the sample and imaged the change in resistivity. Surprisingly, we found that the resistivity changed mainly along the domain walls which are highly sensitive to pressure [Nat. Mat. 2017 doi:10.1038/nmat4966]. Our study shows that the Scanning SQUID is very useful for the investigation of buried domain walls and their effect on nearby layers.



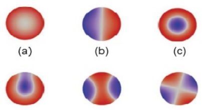
**Igor A. Luk'yanchuk**
*University of Picardie (France)*

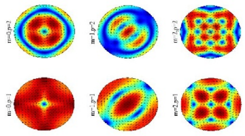
# Ferroelectric multibit cells

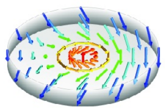
Confinement that imposes additional symmetries on the system can stabilize exotic topological states[1,2] that bring novel functionalities, which do not exist in bulk materials. Confined ferroelectrics that support domain walls[3], vortices[4], skyrmions (compact topologically protected coreless chiral structures)[5-7], are of special interest for applications because topological excitations in ferroelectrics can be relatively easily controlled and manipulated by electric fields. Ferroelectric topological excitations can be reduced in size to atomic scales, and can be manipulated and switched by electric fields with a critical threshold field, in addition to being tunable through lattice strain. The depolarizing charge associated with the topological excitation permits coupling with the incident electromagnetic field and mutual electrostatic cross-talk at the speed of light between excitations. Here we propose ferroelectric multibit cells utilizing the ability of multiaxial ferroelectric materials to pin the polarization at a sequence of the multistable states and, as a consequence, demonstrate the hysteresis loops with multiple branches[8]. Employing the catastrophe theory principles we show that these states are symmetry-protected against the information loss and thus realize novel topologically-controlled access memory (TAM). Figure shows the topological classes of polarization excitations that are realized in ferroelectric oxide nanopillars and nanodots, depending on the anisotropy of the system induced by

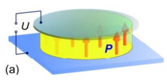
***Collaborators:***
*L. Baudry*
*A. Sené*
*J. Pasoli*
*L. Lachoche*
*V. Vinokour*

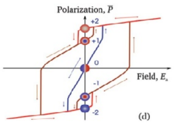
***Funding:***
*H2020 ITN action*
*“NOTEDEV”*

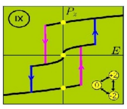


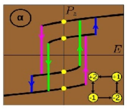












strain. For the uniaxial easy-axis polarization, the different configurations of “up” and “down” oriented domains form[9], whereas for the easy-plane anisotropy one- and multi- in-plane vortex configurations can arise[4]. If anisotropy is weak, one expects formation of switchable chiral skyrmions with “up”- and “down” oriented cores[5-7]. Variety of possible topological textures leads to the symmetry-protected multi-branch hysteresis loops, implementing the memory cells with 5, 3 and 4-level logic correspondingly. Our findings enable developing a platform for the emergent many-valued non-Boolean information technology and target challenges posed by needs of quantum and neuromorphic computing.

[1] N. Mermin, *The topological theory of defects in ordered media*, Rev. Mod. Phys., 51, 591 (1979)

[2] J. Seidel (ed.). *Topological structures in ferroic materials: domain walls*, vortices and skyrmions. Springer, 2016.

[3] G. Catalan, et al., *Domain wall nanoelectronics*, Rev. Mod. Phys. 84 119 (2012)

[4] G. Pascoli L. Lahoche, I. Luk'yanchuk, Integrated Ferroelectrics 99, 60 (2008)

[5] A. Sené, L. Baudry, I. Luk'yanchuk, et al. Superlattices and Microstructures 49, 314 (2011)

[6] L. Baudry, A. Sene, I. Luk’yanchuk, L. Lahoche, Thin Solid Films 519, 5808 (2011)

[7] Y. Nahas, S. Prokhorenko, L. Louis et al., L. Nat. Comm. 6, 1 (2015)

[8] L. Baudry, I. Lukyanchuk and V. Vinokur, V. M. Sci. Rep. 7: 42196 (2017).

[9] P.-W. Martelli, S. M. Mefire, and I. Luk'yanchuk, Europhys. Lett. 111, 50001 (2015).

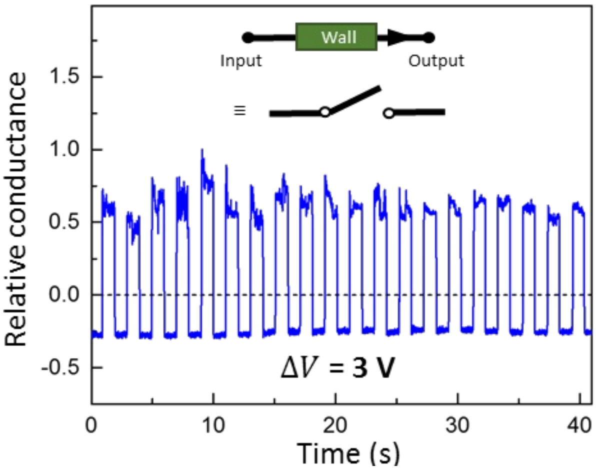


***Figure*:**
*The application of a moderate gate voltage ΔV allows for controlling the domain wall output at positively charged domain walls in hexagonal manganites. The graph presents the normalized domain-wall current, $I_{wall}$, measured as a function of time over 20 switching cycles between resistive ($I_{wall} < I_{bulk}$) and conducting ($I_{wall} > I_{bulk}$) behaviour. Reversible control can be reliably realized for 20 switching cycles, which is the largest number of cycles we investigated.*

**Dennis Meier**

*Department of Materials Science and Engineering, Norwegian University of Science and Technology, 7491 Trondheim (Norway)*

# Emulating the functionality of electronic components using domain walls

***Collaborators:***
*A. Cano*
*J. Schaab*
*J. A. Mundy*
*M. Stengel*
*Y. Kumagai*
*M. Fiebig*
*N. A. Spaldin*
*R. Ramesh*

***Funding:***
*The Alexander von Humboldt Foundation, the Swiss National Science Foundation (SNF), and the Norwegian University of Science and Technology (NTNU) through the Onsager Fellowship Program and the Outstanding Academic Fellows Program.*

Oxide materials exhibit a broad range of tunable phenomena, including magnetism, multiferroicity, and superconductivity. Oxide interfaces are particularly intriguing. The low local symmetry combined with the sensitivity to electrostatics and strain leads to unusual physical properties beyond the bulk properties[1]. Recently, ferroelectric domain walls have attracted attention as a novel type of oxide interface. Most exotic are the so-called charged walls where a polarity mismatch causes local, diverging electrostatic potentials requiring charge compensation and hence a change in the electronic structure. Aside from their unusual electric properties, such walls are spatially mobile and can be created, moved, and erased on demand[2,3]. This additional degree of flexibility enables domain walls to take an active role in future devices and hold a great potential as multifunctional 2D systems for nanoelectronics[4].

In order to make use of the domain wall properties and ultimately design domain-wall-based devices and circuitry for nanotechnology, however, additional functionality beyond just conduction is required that allows the behaviour of classical electrical components to be emulated at the nanoscale.

In my talk I will present unique features that occur at ferroelectric domain walls in multiferroic oxides and discuss how they may be used to eventually emulate electronic components. In the first part, I will address geometrically driven charged domain walls in hexagonal manganites. For our studies we choose the narrow-bandgap, p-type semiconductor $ErMnO_3$ as it naturally develops all fundamental types of ferroelectric domain wall at room temperature, including neutral (side-by-side) as well as negatively (tail-to-tail) and positively charged (head-to-head) wall configurations[5]. The walls are explicitly robust and hence represent an ideal template onto which the desired electronic behavior can be imposed[6,7]. I will show how the electronic properties can be optimized and controlled[8], and discuss the possibility to use such walls for designing 2D digital switches and half-wave rectifiers[9]. In the second part, I will consider domain walls in spin-spiral multiferroics with strong magnetoelectric couplings[10] and additional functionality that arises from the interplay of charge and spin degrees of freedom[3]. Because of the coupling, it is possible to reversibly control the configuration at ferroelectric domain walls in by magnetic fields, switching between nominally charged state and neutral domain wall states[11,12]. Even though the results obtained on low-temperature multiferroics, such as $TbMnO_3$ and $Mn_{0.95}Co_{0.05}WO_4$, are conceptual rather than device oriented, the continuing and successful search for spin-spiral multiferroics with higher ordering temperature turns the design of related domain-wall-based devices into a realistic goal.

The goal of my talk is to push the emergent discussion about conceptually new domain-wall applications that go beyond spatially mobile, conducting two-dimensional channels. The opportunity to emulate electrical components based on domain walls brings us an important step closer to the realization of reconfigurable all-domain-wall circuits for next-generation nanotechnology.

[1] H. Y. Hwang, et al., Nature Mater. 11, 103–113 (2012).
[2] G. Catalan, J. Seidel, R. Ramesh, and J. Scott, Rev. Mod. Phys. 84, 119–156 (2012).
[3] D. Meier, J. Phys.: Condens. Matter 27, 463003 (2015).
[4] E. K. H. Salje, Chem. Phys. Chem. 11, 940–950 (2010).
[5] D. Meier, et al., Nature Mater. 11, 284–288 (2012).
[6] E. Hassanpour, et al., New J. Phys. 18, 43015 (2016).
[7] E. K. H. Salje, New J. Phys. 18, 051001 (2016).
[8] J. Schaab, et al., Adv. Electron. Mater. 2, 1500195 (2016).
[9] J. Mundy, et al., Nature Mater. 16, 622–629 (2017).
[10] M. Fiebig, Th. Lottermoser, D. Meier, and M. Trassin, Nature Reviews Materials 1, 16046 (2016).
[11] N. Leo, et al., Nature Commun. 6, 6661 (2015).
[12] M. Matsubara, et al., Science 348, 1112–1115 (2015).
[13] S. Manz, et al., Nature Photonics 10, 653–656 (2016).

**Maxim Mostovoy**
*Zernike Institute for Advanced Materials, University of Groningen, Nijenborgh 4, 9747 AG, Groningen (The Netherlands)*

# Ferroelectric properties of magnetic skyrmions and merons

I will discuss ferroelectric properties of magnetic topological defects, such as skyrmions and merons. Their non-trivial topology is a source of rich and interesting physics.
Magnetic skyrmions have been recently observed in chiral magnets, such as the ferromagnetic metal, MnSi, and ferrimagnetic insulator, $Cu_2OSeO_3$. Berry phase acquired by electrons and magnons propagating through skyrmions gives rise to complex coupled dynamics of charges and spins mediated by effective gauge fields. Low critical currents needed to manipulate skyrmions opened a new active field of research – skyrmionics, which has a goal of developing skyrmion-based magnetic memory and data processing devices.
It was shown theoretically that isolated skyrmions, skyrmion crystals, merons and other unusual states can exist in magnets with conventional centrosymmetric lattices where they are stabilized by competing exchange interactions[1-4]. Non-collinear magnetic orders in frustrated magnets spontaneously break inversion symmetry and induce an electric polarization. In my talk I will discuss the possibility to control topological magnetic defects in Mott insulators with an applied electric field.

[1] T. Okubo, S. Chung and H. Kawamura, Phys. Rev. Lett. 108, 017206 (2012).
[2] A. O. Leonov and M. Mostovoy, Nature Communications 6, 8275 (2015); 8, 14394 (2017).
[3] S. Hayami, S.-Z. Lin, and C. D. Batista, Phys. Rev. B 93, 184413 (2016).
[4] Y. A. Kharkov, O. P. Sushkov and M. Mostovoy, arXiv:1703.09173.

# Novel functional properties at ferroelectric domain walls: insights from scanning probe microscopy

**Patricja Paruch**
*DQMP, University of Geneva,*
*24 Quai Ernest Ansermet,*
*Geneva (Switzerland)*

In ferroelectric materials, domain walls separate regions with different polarisation orientatio.
Fundamentally, they provide an excellent model system of the rich physics of pinned elastic interfaces, whose behaviour is key for controlling domain size and stability in technological applications. In addition, domain walls can present physical properties and a local internal structure quite different from those of the parent phase. The extreme localisation of such properties at these intrinsically nanoscale features makes them potentially useful as active components in future miniaturised electronic devices. Particularly exciting has been the discovery of domain-wall-specific electrical conduction in many ferroelectric families. Here, I will present our scanned probe microscopy observations of such conduction at 180° domain walls in otherwise insulating Pb(Zr,Ti)$O_3$ (PZT) thin films, highlighting the key role of oxygen vacancies and surface adsorbates, whose distribution can be modulated to reversibly control domain wall transport[1]. At present, we are investigating in more details this interaction between the polarisation, defects, and surface adsorbates, both in terms of the domain wall conductivity, and for its effects on the geometric properties and dynamic behaviour of the domain walls.
In the same ferroelectric samples, we also observe an unusual electromechanical response, forbidden by symmetry in the parent phase but permitted at domain walls as a result of the local symmetry breaking, and the possible emergence of a localised domain-wall-specific polarisation[2]. This enhanced shear response could be technologically important for ferroelectric based surface acoustic wave devices.
Most recently, using nonlinear optical microscopy, we show that indeed there exists a planar polarisation within the domain walls of both PZT and lithium tantalate (LTO), giving rise to pronounced second-harmonic signals. Local polarimetry analysis of these signals, combined with numerical modelling, reveals the presence of Néel-like and Bloch-like configurations in PZT and LTO, respectively, with moreover signatures of domain wall chirality reversal at line defects crossing the LTO crystals[3].

[1] Guyonnet et al., Adv. Mat. 23, 5377 (2011); Gaponenko et al., APL 106, 162902 (2015)
[2] Guyonnet et al. APL 95, 132902 (2009); Guyonnet et al., JAP 108, 042002 (2010)
[3] Cherifi-Hertel et al., Nat. Comm. 8, 15768 (2017)).

***Collaborators:***
*Iaroslav Gaponenko*
*Philippe Tückmantel*
*Christian Weymann*
*Jill Guyonnet*
*Benedikt Ziegler*
*Hélène Béa*
*Cédric Blaser*
*Thierry Giamarchi*
*Lane W. Martin*
*Salia Cherifi-Hertel*
*Riccardo Hertel*
*Katia Gallosh*

***Funding:***
*The Swiss National Science Foundation grants 200021_153174, 206021_150777*

**Sergey Prosandeev**
*Physics Department and Institute for Nanoscience and Engineering,University of Arkansas, Fayetteville, Arkansas 72701 (USA)*

# Unusual phenomena induced by time-dependent toroidal moments

Simulations we conducted about 8 years ago suggested that pulses of electric field can be triggered by the time-evolution of magnetic vortices[1]. Conversely, we more recently proposed that a dynamical magnetization can arise from a temporal change of electrical vortices[2]. One consequence of such latter finding is the occurrence of optical activity when the electric toroidal moment is time-dependent, which is in line with the evidence pointed out by Pauster[3] that chiral molecules or crystals can be optically active. Another consequence of this finding is that vibrating ferroelectric domain walls can generate a magnetization in non-magnetic systems[4], which was, in fact, experimentally found (but then overlooked, see [5] and references therein) in the past. All these unusual phenomena will be reported and discussed in this talk.

[1] S. Prosandeev and L. Bellaiche. Electric Field Induced by Dynamical Change of Dipolar Configurations in Ferromagnets, Phys. Rev. Lett. 102, 097205 (2009).
[2] S. Prosandeev, A. Malashevich, Z. Gui, L. Louis, R. Walter, I. Souza, and L. Bellaiche, Natural optical activity and its control by electric field in electrotoroidic systems, Phys. Rev. B 87, 195111 (2013).
[3] L. Pasteur Researches on the Molecular Asymmetry of Natural Organic Products (Edinburg: Alembic 1897).
[4] S. Prosandeev, A. Malashevich, I. P. Raevski, and L. Bellaiche. Dynamical magnetoelectric effects associated with ferroelectric domain walls, Phys. Rev. B 91, 100101 (2015).
[5] S. A. Flerova and I. E. Chupis, Bull. Russ. Acad. Sci.: Phys. 57, 409 (1993).

***Collaborators:***
*L. Bellaiche*

***Acknowledgements:***
*This work is supported by ONR Grant N00014-12-1-1034.*

**Ramamoorty Ramesh**

*Department of Materials Science and Engineering & Department of Physics, UC Berkeley, and Materials Sciences Division, LBNL (USA)*

# Emergent chirality and phase coexistence in polar vortices formed in oxide superlattices

The complex interplay of spin, charge, orbital, and lattice degrees of freedom has provided for a plethora of exotic phase and physical phenomena. Among these, in recent years, topological states of matter and spin textures have emerged as fascinating consequences of the electronic band structure and the interplay between spin and spin-orbit coupling in materials. In this work, we leverage the competition between charge, orbital, and lattice degrees of freedom in superlattices of $PbTiO_3/SrTiO_3$ to produce complex, vortex-antivortex pairs (that exhibit smoothly varying ferroelectric polarization with a 10 nm periodicity) that are reminiscent of topological features such as skyrmions and merons. Using a combination of advanced layer-by-layer growth techniques, atomic-resolution mapping of structure and local polar distortions using scanning-transmission electron microscopy, and phase-field modeling approaches we present a comprehensive picture of the nature of the varying polarization profile in such vortex states. The continuous rotation of the polar state into the vortex structures is thought to occur from an interplay of polar discontinuities at the $PbTiO_3/SrTiO_3$ interface, the phase transformation strain and gradient energy in the $PbTiO_3$ layer, and the strain imposed by the substrate. Finally, the implications of these observations are discussed as they pertain to producing new states of matter and emergent phenomena (such as chirality) in such superlattices.

**Andrew Rappe**
*University of Pennsylvania,*
*Department of Chemistry,*
*231 S.34th St,*
*Philadelphia, PA 19104-6323 (USA)*

# Intrinsic ferroelectric domain wall motion revealed by first-principles based molecular dynamics

***Collaborators:***
*Shi Liu*
*Ilya Grinberg*

The existence of domain walls, which separate regions of different polarization, can influence the dielectric, piezoelectric, pyroelectric, and electronic properties of ferroelectric materials. In particular, domain-wall motion is crucial for polarization switching, which is characterized by the hysteresis loop, a signature feature of ferroelectric materials. Experimentally, the observed dynamics of polarization switching and domain-wall motion are usually modeled as an elastic interface pinned by a random potential that is generated by defects, which appear to be strongly sample-dependent and affected by various elastic, microstructural and other extrinsic effects. Theoretically, connecting the 0 K, first-principles-based, microscopic quantities of a sample with finite-temperature, macroscopic properties such as the coercive field is critical for material design and device performance; and the lack of such a connection has prevented the use of techniques based on ab initio calculations for high-throughput computational materials discovery. Here we use molecular dynamics simulations of 90° domain walls (separating domains with orthogonal polarization directions) in the ferroelectric material PbTiO3 to provide microscopic insights that enable the construction of a simple, universal, nucleation-and-growth-based analytical model that quantifies the dynamics of many types of domain walls in various ferroelectrics. We then predict the temperature and frequency dependence of hysteresis loops and coercive fields at finite temperatures from first principles. We find that, even in the absence of defects, the intrinsic temperature and field dependence of the domain-wall velocity can be described with a nonlinear creep-like region and a depinning-like region. Our model[1] enables quantitative estimation of coercive fields, which agree well with experimental results for ceramics and thin films. This agreement between model and experiment suggests that, despite the complexity of ferroelectric materials, typical ferroelectric switching is largely governed by a simple, universal mechanism of intrinsic domain-wall motion, providing an efficient framework for predicting and optimizing the properties of ferroelectric materials.

[1] S.Liu, I.Grinberg & A.M. Rappe, “Intrinsic FE switching from 1st principles”, Nature, 534, 360-3 (2016).

**Jan Seidel**
*School of Materials Science and Engineering, UNSW Sydney, Sydney NSW 2032 (Australia)*

# Domain walls and phase boundaries for memory applications

Topological structures in functional materials, such as domain walls and skyrmions, see increased attention due to their properties that can be completely different from that of the parent bulk material[1] and provide interesting prospects for memory applications[2]. I will discuss recent results on multiferroic phase boundaries and domain walls in BiFeO3[3-6] using SPM, TEM and theory, and discuss future prospects[7, 8].

[1] J. Seidel (ed.), *Topological structures in ferroic materials: domain walls, skyrmions and vortices*, ISBN: 978-3-319-25299-5, Springer, Berlin (2016).

[2] P. Sharma, et al., Science Advances 3, e1700512 (2017).

[3] P. Sharma, et al., Advanced Electronic Materials 2, 1600283 (2016).

[3] J. Seidel, et al., Advanced Materials 26, 4376 (2014).

[4] Y. Heo, et al., Advanced Materials 26, 7568 (2014).

[5] Y. Heo et al., ACS Nano 11, 2805 (2017).

[6] P. Sharma, et al., Advanced Materials Interfaces 3, 1600033 (2016).

[7] N. Faraji et al., Applied Physics Letters 110, 213108 (2017).

[8] J. Seidel, Nature Nanotechnology 10, 190 (2015).

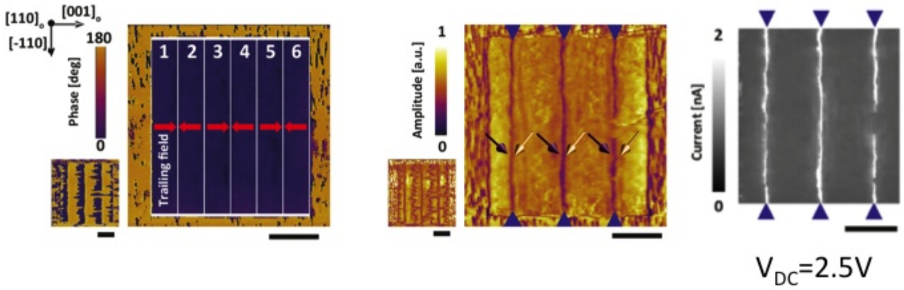


***Figure**: Conductive charged domain walls, formed and reconfigured in $BiFeO_3$ films[10].*

**Nava Setter**

*Faculty of Engineering, EPFL, 1015 Lausanne, (Switzerland) and Department of Materials Science and Engineering, Tel Aviv University, 69978 Ramat Aviv (Israel)*

# Insight into Structure, Properties, and Mobility of Ferroelectric Domain Walls

As interfaces that can be displaced in-situ, ferroelectric domain walls are a source of continuous fascination. We have been studying during the past 5 years some of their properties and internal structure and learning how to control domain wall patterns and ultimately functionalize them. Among the obtained results are dense patterns of arrays of domains and domain walls having <10 nm width /periodicity [1, 2], controlled displacement of domain walls[3, 4, 5], charged domain walls with metallic conductivity inside the insulating matrix[6, 7] and their controlled creation and density[8, 9] and demonstrated reconfigurability[10]. It has been found also that tailored bent neutral domain walls can be electrically conductive, and this metallic conductivity is sustained to ultra-low temperature (testifying the metallic nature of the conductivity)[11, 12]. In addition, we have evidenced ferroelectric boundaries in non-ferroelectric, antiferroelectric materials[13], evidenced polarization rotation across wide walls [14], demonstrated ferroelectric switch for propagation of ferromagnetic domain walls at room temperature [15], and showed the possibility of elastic interaction between non-ferroelastic domain walls[16], promising new possibilities for domain-wall control.

[1] L. Feigl et al., Nature Commun. (2014).
[2] M. Mtebwa et al. APL (2015).
[3] L. McGilly et al., Nature Nanotech (2015).
[4] L. McGilly et al. Nano Ltters (2016).
[5] L. McGilly et al. Ad Fun Materials (2017).
[6] T. Sluka et al., Nature Commun (2012).
[7] T. Sluka et al., Nature Commun. (2013).

[8] P. Bednyakov et al. Sci. Reports (2015).

[9] P. Bednyakov et al. Adv. Materials (2016).

[10] A. Crassous et al. Nature Nanotech (2015).

[11] I. Stolichnov et al. Nano Letters (2015).

[12] X.-K. Wei et al., ACS Appl. Mat. & Interfaces (2016).

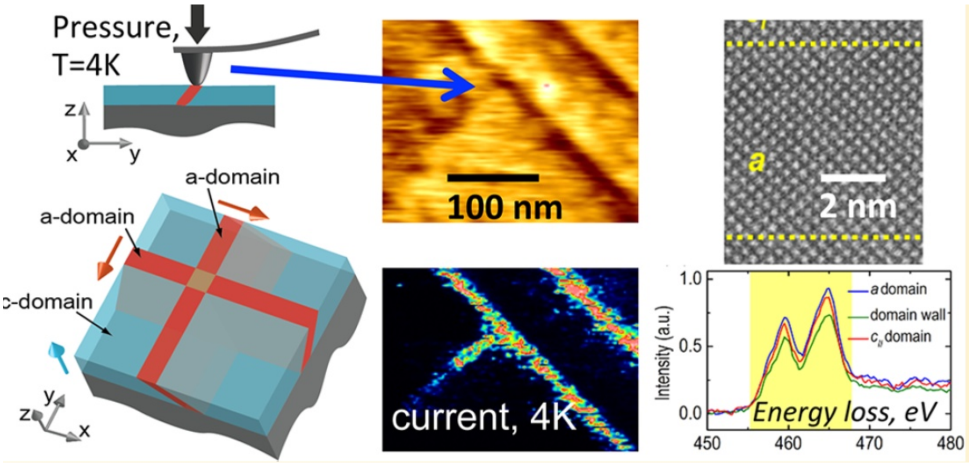
[13] X.-K. Wei et al., Nat. Commun (2014).

[14] X. -K. Wei et al., Nat. Commun (2016).

[15] Z. Huang et al. (2015).

[16] K. Shapovalov et al. PRL (2014).

*__Figure__: Ultrafine domain, written by pressure at 40K (left), shows conductivity (bottom, middle) and its confinement to the domain walls (bottom, right)[11].*



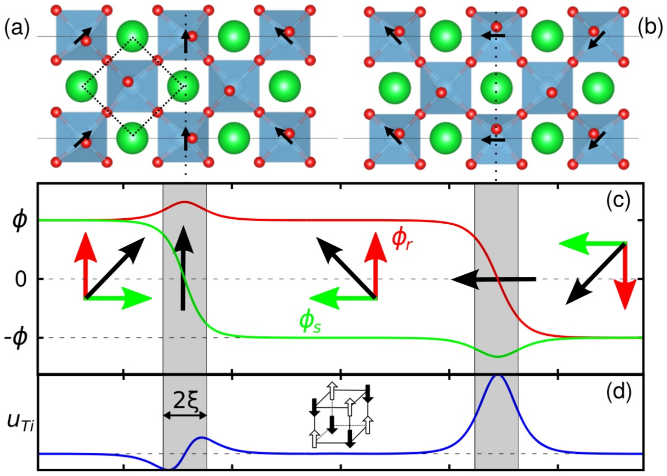


***Figure:***
*(a-b) Schematic illustration of the two different types of twin boundaries (TB) considered in this work, respectively "head-to-head" HH (a) and "head-to-tail" HT (b); the dashed square indicates the primitive cell of the cubic reference phase; the arrows indicate the local antiferrodistortive tilt vector. (c) Evolution of the amplitude of the tilt order parameter across the two TBs. A local decomposition of the tilt vector (black arrows) into longitudinal (green) and transverse (red) components is also shown. The shaded area indicates the nominal wall thickness. (d) Amplitude of the secondary antiferroelectric (AFE) distortion mode,* $u_{Ti}$*, in arbitrary units. The inset illustrates the AFE character of the Ti displacements, resembling spins in a G-type antiferromagnet.*

**Massimiliano Stengel**
*Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), Campus UAB, 08193 Bellaterra, and Institució Catalana de Recerca i Estudis Avançats (ICREA), 08010 Barcelona (Spain)*

# Macroscopic polarization from antiferrodistortive cycloids in ferroelastic $SrTiO_3$

Ferroelastic twin walls have received considerable attention in the past few years, as they are characterized by a net dipole moment even if the parent material is nonpolar. Several models have been proposed to rationalize this observation, ranging from flexoelectricity to improper ferroelectricity, but a fundamental theory of the effect is still missing. In this talk I will first give a brief overview of the technical and conceptual challenges that one has to face to describe gradient-mediated couplings from the perspective of microscopic electronic-structure theory. Next, by using ferroelastic twins in $SrTiO_3$ as a testcase, I will show how these challenges can be successfully overcome, leading to a physically consistent, quantitatively predictive description of domain wall-induced polarity. In particular, I will discuss two new mechanisms that crucially contribute to P: a direct "rotopolar" coupling to the gradients of the antiferrodistortive oxygen tilts, and a trilinear coupling that is mediated by the antiferroelectric displacement of the Ti atoms. Remarkably, the rotopolar coupling presents a strong analogy to the mechanism that generates a spontaneous polarization in cycloidal magnets, thereby allowing for a breakdown of macroscopic inversion symmetry (and therefore a macroscopic polarization) in a periodic sequence of parallel twins. These results open new avenues towards engineering pyroelectricity or piezoelectricity in nominally nonpolar ferroic materials.

***Funding:***
*Ministerio de Economia, Industria y Competitividad (MINECO-Spain), Grants No. FIS2013-48668-C2-2-P, MAT2016-77100-C2-2-P, and SEV-2015-0496; Generalitat de Catalunya (Grant No. 2014 SGR301); European Research Council (ERC) under the European Union's Horizon 2020 research and innovation program (Grant Agreement No. 724529).*

[1] Andrea Schiaffino and Massimiliano Stengel, Phys. Rev. Lett. 119, 137601 (2017).

**David Vanderbilt**
*Department of Physics and Astronomy, Rutgers University, Piscataway, New Jersey (USA)*

# Ferroelectric reversal and domain walls in corundum derivatives

The known ferroelectric (FE) materials $LiNbO_3$ and $LiTaO_3$ can be regarded as derived from the $A_2O_3$ corundum structure with cation ordering. Here we consider other binary $ABO_3$ and more general ternary $A_2BB'O_6$ corundum derivatives as an extended class of potential FE materials, motivated by the fact that some members of this class have recently been synthesized using high-pressure growth techniques (see, e.g., [1]). We first identify structure types within this class that allow polarization reversal between equivalent structures. These structures are all strongly polar with large spontaneous polarizations, but a material cannot be considered ferroelectric unless the polarization is reversible by the application of an electric field. This in turn is determined by the height of the energy barrier for domain reversal.

We therefore report first-principles calculations of the energy barriers for FE reversal in a range of representative materials from this class. These are computed first for the case of coherent bulk reversal, and then also for he more realistic case of reversal via motion of 180° domain walls. Our calculations predict the orientation and formation energies of domain walls. For $A_2BB'O_6$, which are chiral, and for materials with magnetic ions, which are multiferroic, we also discuss important couplings between polarization, magnetization, and chirality at the domain walls. In particular, we find an interesting extrinsic magnetoelectric effect in the case of certain magnetic corundum derivatives. Finally, we also identify the mechanisms for migration of domain walls and their migration barriers.

We expect our results[2, 3] to assist

***Collaborators:***
*Meng Ye*

***Acknowledgements:***
*This work was supported by ONR grants N-00014-12-1-1035 and N-00014-16-1-2591.*

in the experimental search for new FE materials in the corundum derivative family.

[1] SM.-R. Li, M. Croft, P. W. Stephens, M. Ye, D. Vanderbilt, M. Retuerto, Z. Deng, C. P. Grams, J. Hemberger, J. Hadermann, W.-M. Li, C.-Q. Jin, F. O. Saouma, J. I. Jang, H. Akamatsu, V. Gopalan, D. Walker, and M. Greenblatt. “$Mn_2FeWO_6$ : a new $Ni_3TeO_6$-type polar and magnetic oxide.” Advanced Materials 27, 2177 (2015).

[2] M. Ye and D. Vanderbilt. “Ferroelectricity in corundum derivatives.” Phys. Rev. B 93, 134303 (2016).

[3] M. Ye and D. Vanderbilt. “Domain walls and ferroelectric reversal in corundum derivatives.” Phys. Rev. B 95, 014105 (2017).

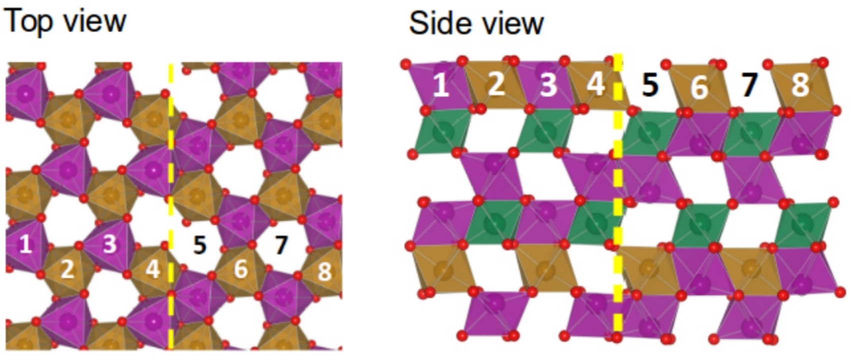


***Figure***: *Top and side view of one of the considered domain-wall structures in an $A_2BB'O_6$ corundum-derivative ferroelectric material.*

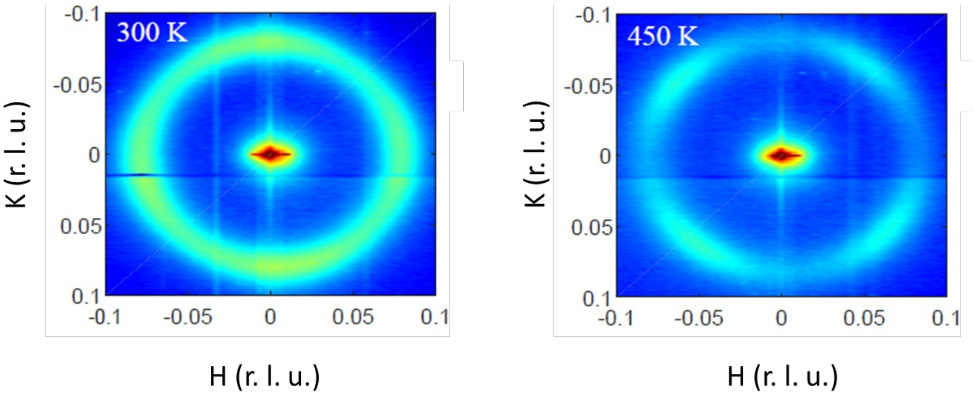


***Figure:*** *Reciprocal space maps at different temperatures for a PbTiO3/SrTiO3 superlattice illustrating the change in preferred domain wall orientation from {100} walls at 300 K to {110} walls at 450 K.*

**Pavlo Zubko**

*London Centre for Nanotechnology and Department of Physics and Astronomy, University College London, 17-19 Gordon Street, London WC1H 0AH (UK)*

# Imaging ferroelectric domain structures with X-ray nanodiffraction

Thin film epitaxy and heterostructuring provide a powerful method for controlling the electrostatic and mechanical boundary conditions and thereby engineering a wide variety of regular, dense ferroelectric and ferroelastic domain structures. Such dense domains are especially interesting in the light of the recent discoveries of new properties inherent to domain walls and interesting behaviour emerging from their dynamics [1,2]. At the same time, there are many difficulties associated with the non-destructive characterization of domain structures at the nanoscale, particularly at high temperatures and in buried layers, where scanning probe microscopy becomes very challenging. Techniques that extend our capability to characterise ferroelectric domain structures are therefore highly sought after.

Here we have employed X-ray nanodiffraction to study ferroelectric and ferroelastic domain structures in $PbTiO_3$ thin films and superlattices. In superlattices composed of alternating, ultrathin layers of $PbTiO_3$ and $SrTiO_3$, dense ferroelectric stripe domains form in response to the depolarisation fields induced by the lack of screening charges [3]. Using synchrotron X-ray diffraction, we have found that these stripe domains have a preferred domain wall orientation that rotates from {100} walls at low temperature to {110} walls at higher temperatures (Fig. 1). Local diffraction measurements performed with a nanofocused X-ray beam were then used to map the variation of domain wall orientations across the

***Collaborators:***

*M. Hadjimichael*
*E. Zatterin*
*S. J. Leake*
*S. Fernandez-Pena*

***Acknowledgements:***

*his work was supported by ONR grants N-00014-12-1-1035 and N-00014-16-1-2591.*

sample. Measurements in the vicinity of an extended structural defect reveal a strong preferential alignment of domain walls along features associated with the defect. Local 3D reciprocal space maps were further used to map out the special distribution of the out-of-plane strain and its corresponding gradients associated with the defect. The same technique was then applied to image the ferroelastic domain structures in $PbTiO_3$ films under tensile strain. These films contain bundles of periodic a1/a2 and a/c domains that give rise to diffraction satellites due to the domain periodicity and Bragg peak splitting due to the twinning. By selecting different regions of interest in reciprocal space, the variation of intensity arising from different domain types can be mapped across the sample using a nanofocused beam, thereby imaging the different ferroelastic domain bundles nanodomains. Such images of domain bindles can then be directly compared with data obtained using piezoresponse force microscopy.

[1] G. Catalan et al. Rev. Mod. Phys. 84, 119 (2012).

[2] P. Zubko et al. Nature 534, 524 (2016); A. Bratkovsky, A. Levanyuk, Phys. Rev. B 63, 132103 (2001).

[3] S. K. Streiffer et al. Phys. Rev. Lett. 89, 067601 (2002); D. Fong et al. Science 304, 1650 (2004) ; P. Zubko et al. Phys. Rev. Lett. 104, 187601 (2010).

# Venue: Museum of the Jewish People

Our Workshop takes place at the Zeevi Auditorium of the Museum of the Jewish People, inside the Campus of Tel Aviv University.
The Museum presents and displays the unique and ongoing 4,000 year-old story of the Jewish people—past, present and future. It opened in 1978 thanks to the vision of Nahum Goldmann, president of the World Jewish Congress 1954-1977. In 2005, the Israeli Knesset passed the Beit Hatfutsot Law that defines Beit Hatfutsot as "the National Center for Jewish communities in Israel and around the world".
The Museum displays permanent and temporary exhibitions. Among the permanent ones, there is a world renowned collection of synagogue models. At the time of our Workshop, the temporary ones will include an exhibition on the life and influence of Bob Dylan, one of the greatest american poets and musicians of our time.
More details can be found at: https://www.bh.org.il/.

# Maps: TAU Campus and Surroundings

Workshop Venue:
The Museum of the Jewish People

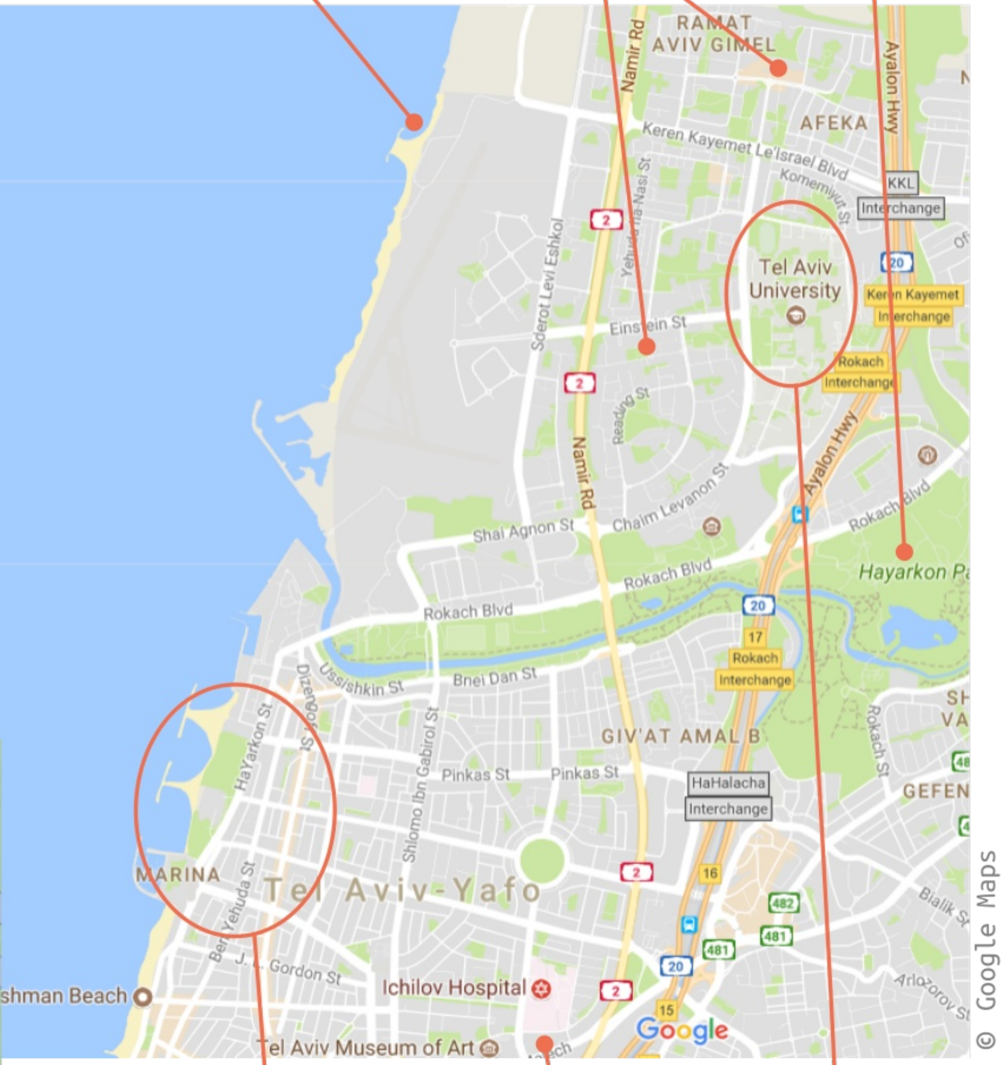
Einstein Dorms

Broshim Dorms
(next to a few restaurants
and one supermarket)

Leisure:
Tel Baruch Beach

Leisure:
Shopping Malls

Leisure:
Large Park

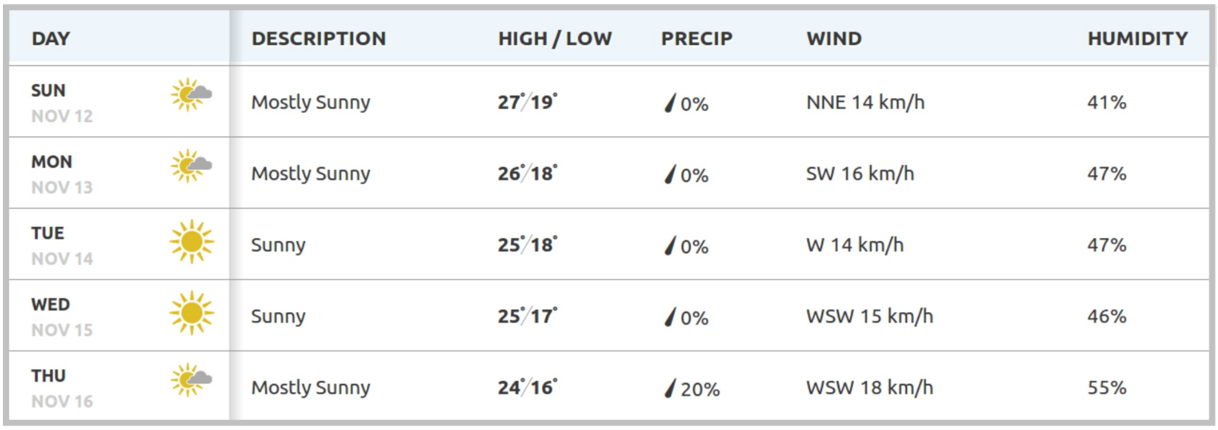


Workshop Hotels
(shuttle available
to and from TAU Campus;
restaurants and shops
in the Port area)

TAU Campus
(see details in the map
on the left)

Workshop Banquet:
Lilyot Restaurant
(South of here: downtown Tel Aviv)

# Transportation & Weather



| DAY | | DESCRIPTION | HIGH / LOW | PRECIP | WIND | HUMIDITY |
|---|---|---|---|---|---|---|
| SUN<br>NOV 12 | ⛅ | Mostly Sunny | 27°/19° | 0% | NNE 14 km/h | 41% |
| MON<br>NOV 13 | ⛅ | Mostly Sunny | 26°/18° | 0% | SW 16 km/h | 47% |
| TUE<br>NOV 14 | ☀ | Sunny | 25°/18° | 0% | W 14 km/h | 47% |
| WED<br>NOV 15 | ☀ | Sunny | 25°/17° | 0% | WSW 15 km/h | 46% |
| THU<br>NOV 16 | ⛅ | Mostly Sunny | 24°/16° | 20% | WSW 18 km/h | 55% |

© The Weather Channel (retrieved Nov 7)

**Taxi from the airport:**
There is a line of regular taxis outside the terminal of Ben Gurion Airport. The price of a ride to the Workshop hotels or dorms, or to the Tel Aviv Campus is between 150 and 200 NIS (1 NIS = 0.25 EUR).

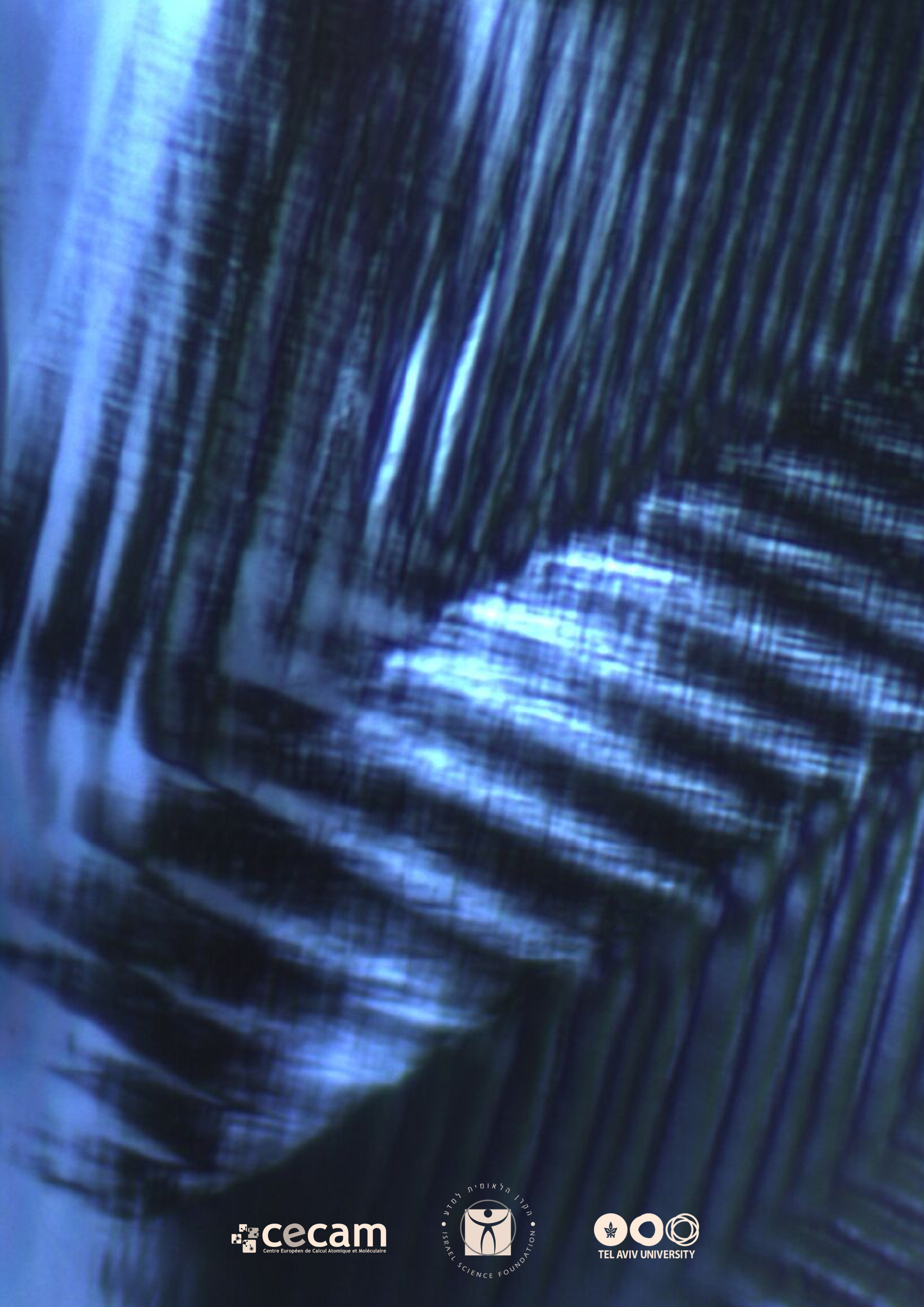
**Taxi applications:**
The most used one in Israel is probably Gett (Uber and Lyft do not operate in Israel in the way they do in other countries).

**Reaching TAU Campus by shuttle:**
A shuttle from the Workshop hotels to TAU Campus and back will be provided before and after the Workshop sessions.

**Reaching TAU Campus by car:**
The Rokach Boulevard exit of Ayalon Highway is the nearest to the Tel Aviv Campus.

**Reaching TAU Campus by bus:**
Lines 74, 86, 572, 274, 604, and 475 of the Egged bus company stop near the campus. Lines 7, 13, 24, 25, 27, 45, 49, and 112 of the Dan bus company have also nearby stops.

**Reaching TAU Campus by train:**
The Tel Aviv University train station is within walking distance of the campus (around 10 minutes, uphill). For additional information, see the Israel Railways website.



cecam
Centre Européen de Calcul Atomique et Moléculaire

הקרן הלאומית למדע
ISRAEL SCIENCE FOUNDATION

TEL AVIV UNIVERSITY