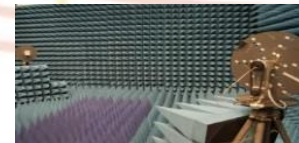
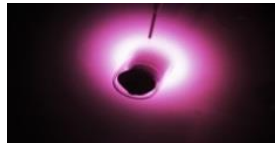
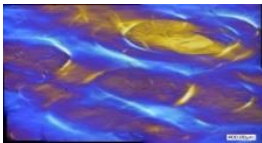


Needs and Challenges for the characterization of dielectrics at nanoscale

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Some needs for (nano)dielectrics characterization at nanoscale

Expected quantities

Information from and limitation of scanning probe methods

Information from and limitation of classical space charge measurement methods

Needs for characterization on interfaces at nanoscale: 3 drivers

- Physics of interface**
- Take benefit from new processes for tailoring external/internal interfaces**
- Devices in power engineering /energy challenges involving thinner layers**



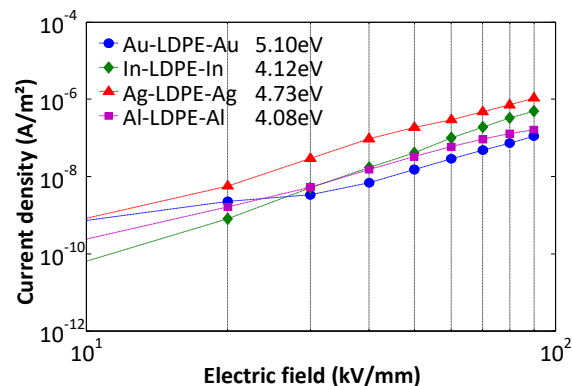
Needs: insulator/electrode contact

Electronic properties of interfaces at insulator/electrode contact of paramount importance

- i/ Low amount of intrinsic carrier density in insulations viewed as wide band gap materials and the dominance of injection process in transport**
- ii/ Reports on substantial dependence of apparent conduction on the interface and electrode conditions, including nature of the electrode material, roughness, surface properties**
- iii/ Electrical double layer in electrode/dielectric liquid interface**
- iv/ The need for realistic boundary conditions when developing drift-diffusion models of transport**

Examples of interface-mediated macroscopic behavior

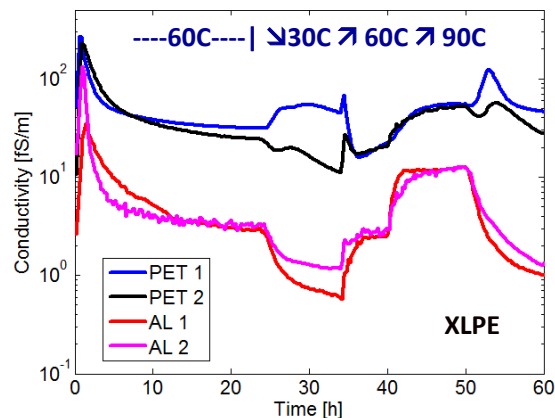
Influence of electrode metal on apparent conduction



M. Taleb et al. Proc IEEE-CEIDP, p.112, 2009

No clear correlation to the work function

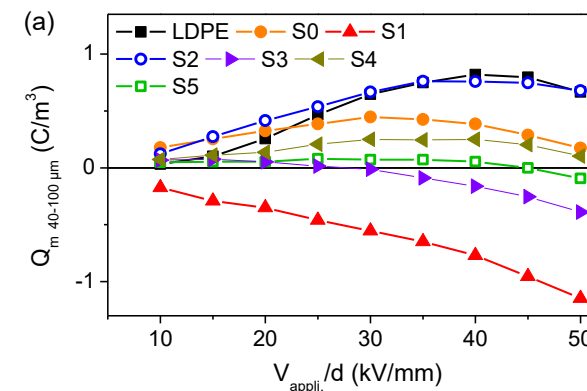
Influence of molding process on apparent conduction



H. Ghorbani et al. Proc. IEEE-ICD, p. 1179, 2016

Impact on both XLPE and LDPE

Interface modification (Ag NPs, fluorination) vs. space charge



L. Milliere et al. J. Phys. D 49 , 015304, 2016

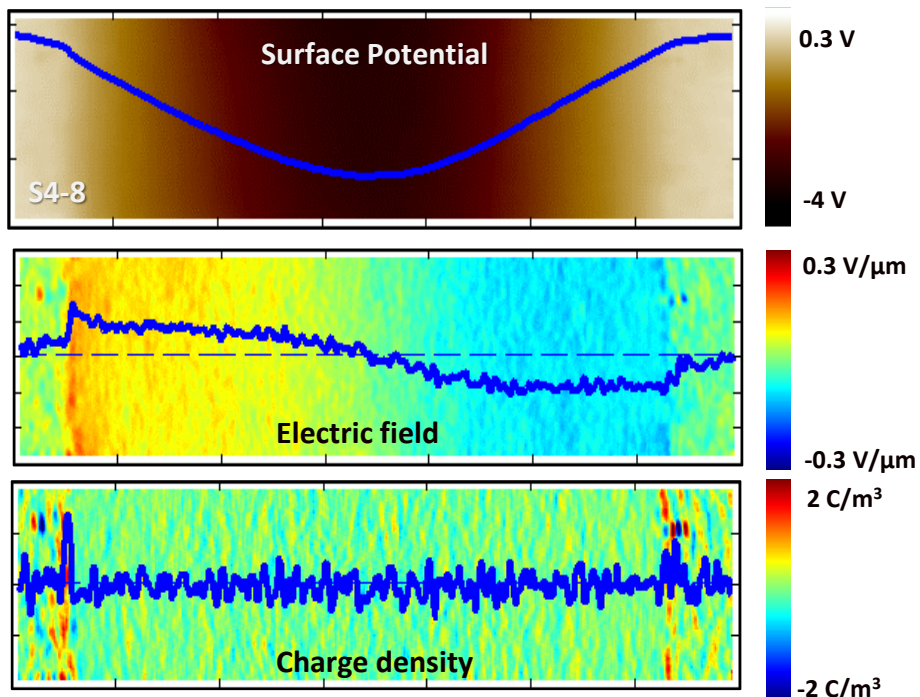
Efficient screening effect due to charge trapping

How is material characterization affected by the used electrodes?

What tools to implement to identify characteristic interface properties?

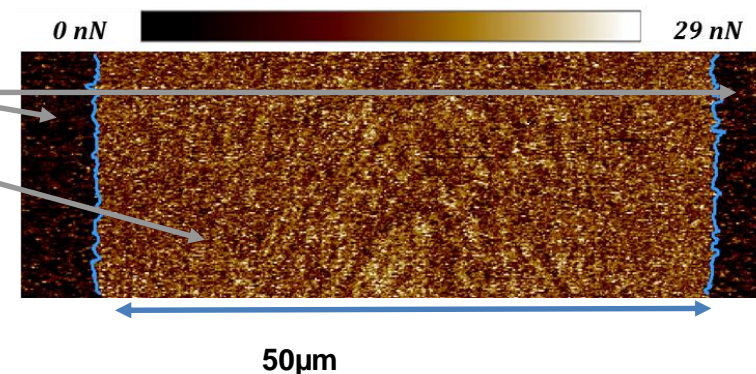
Process-dependent potential profiles along cross-sections

Surface potential by KPFM



Electrodes: Semiconducting polymer

Bulk: low density polyethylene



Theoretically, the surface potential corresponds to:

- The work function for metals
- The Fermi level for semiconductors (depends on doping)

Need to be measured in equilibrium state.

Smooth variation of the potential that would reflect internal charge build-up

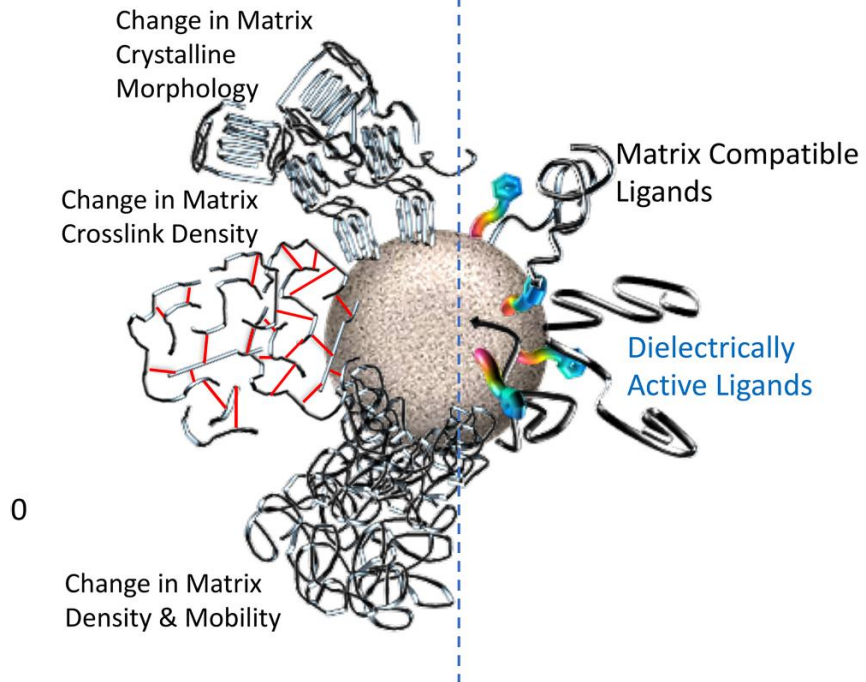
The response depends on the cover-layer used for press-moulding the samples

2nd derivative of voltage does not provide clear view of charge

Polymer matrix / inorganic nanoparticles

Intrinsic Interfaces

Extrinsic Interfaces



Nanodielectrics may have specific properties owing to the large specific area brought by nanoparticles and by specific properties of these interface (interphase) regions

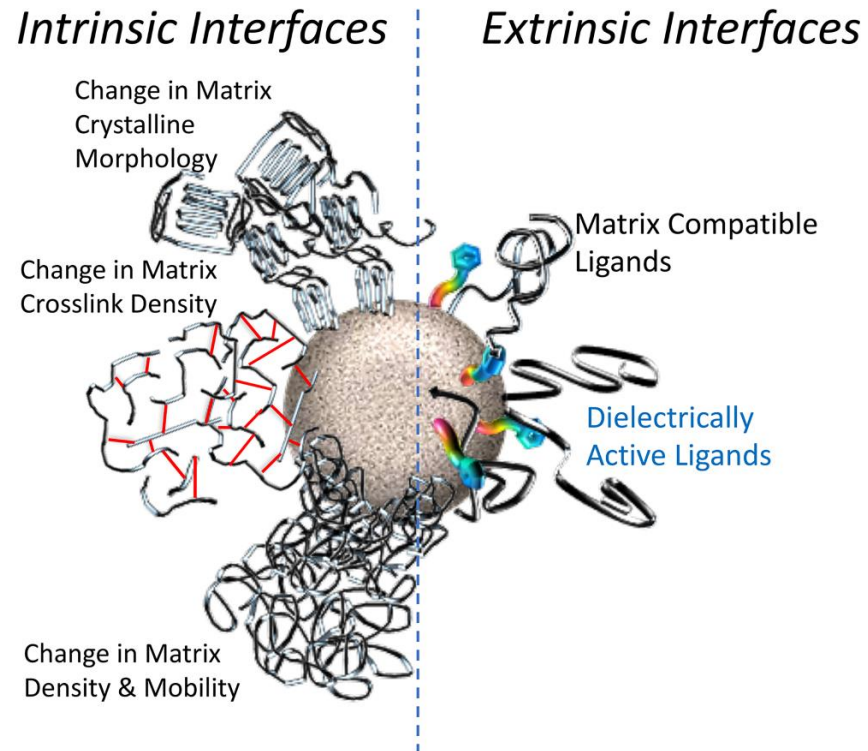
This interfacial region can be categorized into two regions:

- intrinsic interphase, as a result of changes in the matrix structure, chain mobility, cross-link density, with possibly double layer response as suggested by Lewis;
- extrinsic interfacial region, as a result of ligands on the surface of the fillers, particularly resulting from functionalization. This extrinsic interphase region can be extensive / tailored / controlled for building a charge trapping, or region of altered conductivity or permittivity.

L. S. Schadler and J. Keith Nelson, *Polymer nanodielectrics—Short history and future perspective*, J. Appl. Phys. 128, 120902, 2020

T. J. Lewis, *Nanometric dielectrics*, IEEE Trans. Dielectr. Electr. Insul. 1, 812, 1994

Polymer matrix / inorganic nanoparticles



Improving of dielectric properties was extensively reported with properly dispersed nanoparticles:

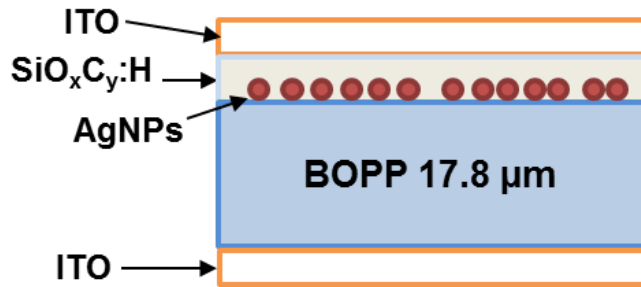
- improved in resistance to discharge-induced erosion
- improved in breakdown field
- decrease in macroscopic conductivity
- apparent decrease in charge accumulation

→ Due to ‘trapping at interfaces’

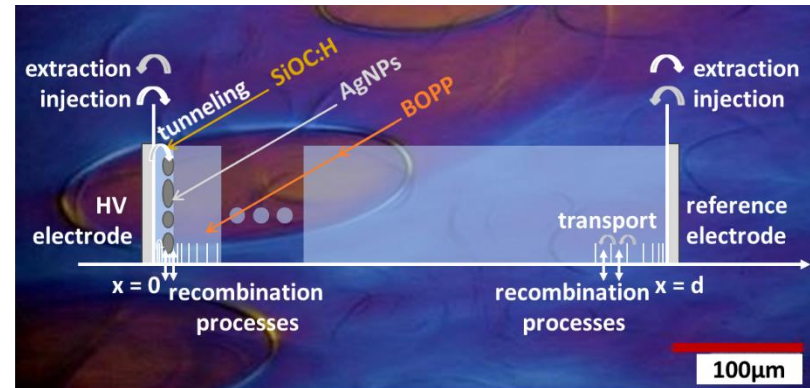
→ **Is it a bulk effect or a surface effect?**

→ Need high spatial resolution charge probing method

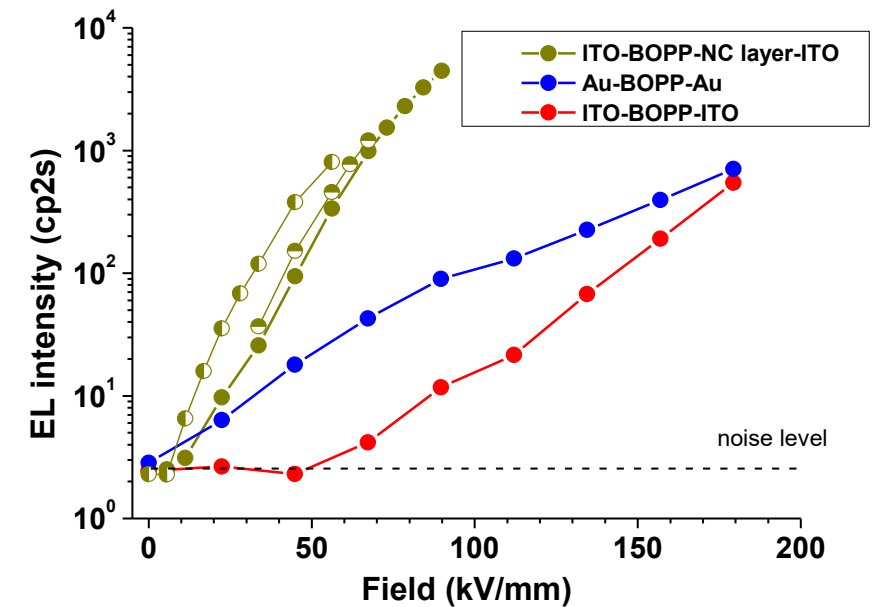
Interface tailoring of dielectrics: AgNPs/SiOC:H stack deposited on BOPP



⇒ DC stress: mitigation of charge injection with the tailored NC layer



⇒ Electroluminescence under ac-stress



Strong increase of ac-EL signal with the NC layer ($\times 100^+$)

⇒ Behaviour controlled by charge-exchange mechanisms. Light detected even at low field
 Expectancy to probe the charge state of metallic NPs

Requirements for nm to sub- μm scale resolution

Power Engineering

- wiring of motors (typically 20-40 μm thickness), insulation/passivation in power electronics (some μm)
- polymer capacitors: thicknesses 5-20 μm

Aerospace Applications

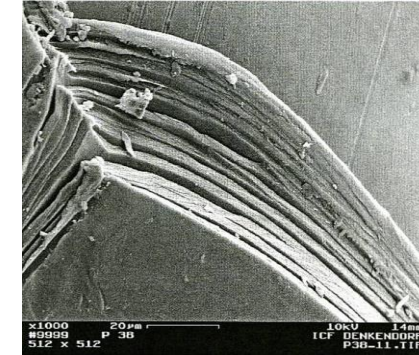
- thin dielectrics in outer parts of spacecraft (some 10s of μm)
- tribo-electrification; flow electrification phenomena
- corona charging analysed so far as surface charging phenomena

Electronics

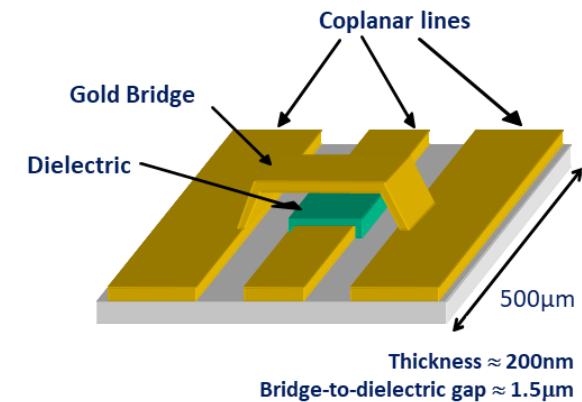
- integrated and miniaturized devices: actuators, MEMS, lab-on-a-chip, dosimeters, passivation layers...
... with μm or sub- μm scale
- microelectronics: SiO_2 gates, MOS devices (nm scale)

Energy harvesting

- Photovoltaics
- piezo-, pyro-electric thin films in energy conversion



PAI/PEI enamelled wire insulation, 13 layers, total: 50 μm . Image $\approx 100 \times 100 \mu\text{m}^2$





What needs to be probed?

Local properties in a dielectric with 'high' spatial resolution

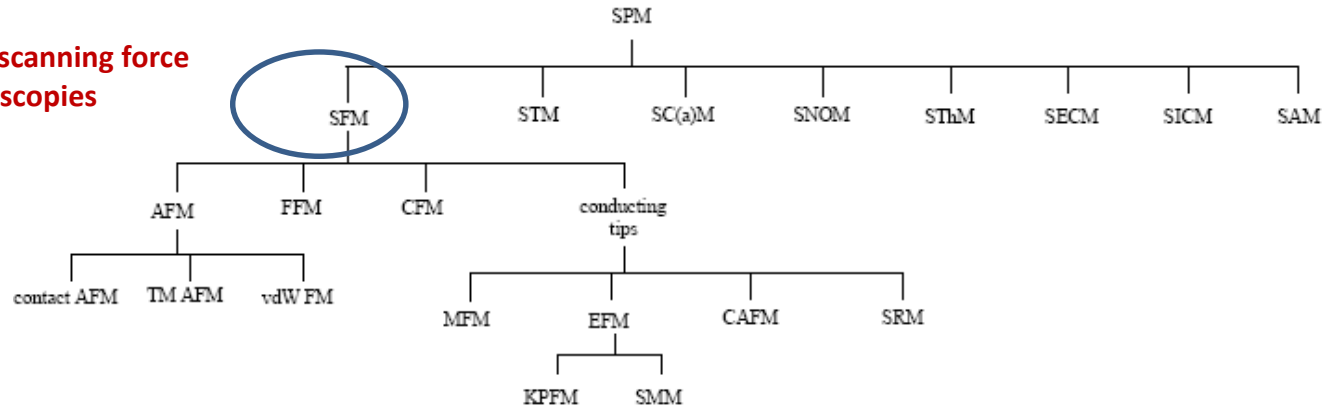
- Conductivity
- Permittivity
- Electrical charge amount / trapping processes

Interfaces/interaction between materials

- Energy levels
- Charge exchange mechanisms: 'injection barrier' in a wide bandgap material?
- Topology / composition / structure / defects

Electrical Modes of Scanning Probe Methods constitute on way of providing new information

SFM: scanning force microscopies



Of specific interest

-EFM: electrostatic force microscopy: maps of field distribution → permittivity distribution

KPFM: Kelvin probe force microscopy: maps of surface potential

CAFM: conducting atomic force microscopy

Peak Force – QNM Quantitative Nanomechanical: topography and mechanical properties (adhesion-deformation-Young modulus) mapping

SPM: scanning probe microscopy

STM: scanning tunnelling microscopy

SFM: scanning force microscopy

SC(a)M: scanning capacitance microscopy

AFM: atomic force microscopy

SNOM: scanning near-field optical microscopy

FFM: friction force microscopy

SThM: scanning thermal microscopy

CFM: chemical force microscopy

SECM: scanning electrochemical microscopy

TM AFM: tapping mode AFM

SICM: scanning ion conductance

vdWFM: van der Waals force microscopy

SAM: scanning acoustic microscopy

MFM: magnetic force microscopy

EFM: electrostatic force microscopy

KPFM: Kelvin probe force microscopy

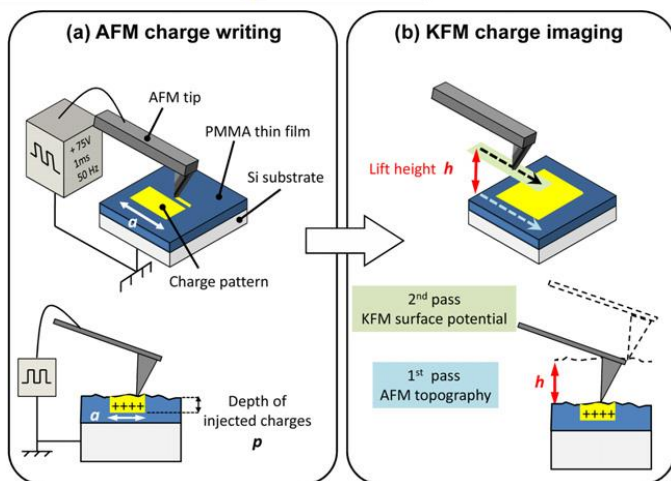
SMM: scanning Maxwell stress microscopy

CAFM: conducting atomic force microscopy

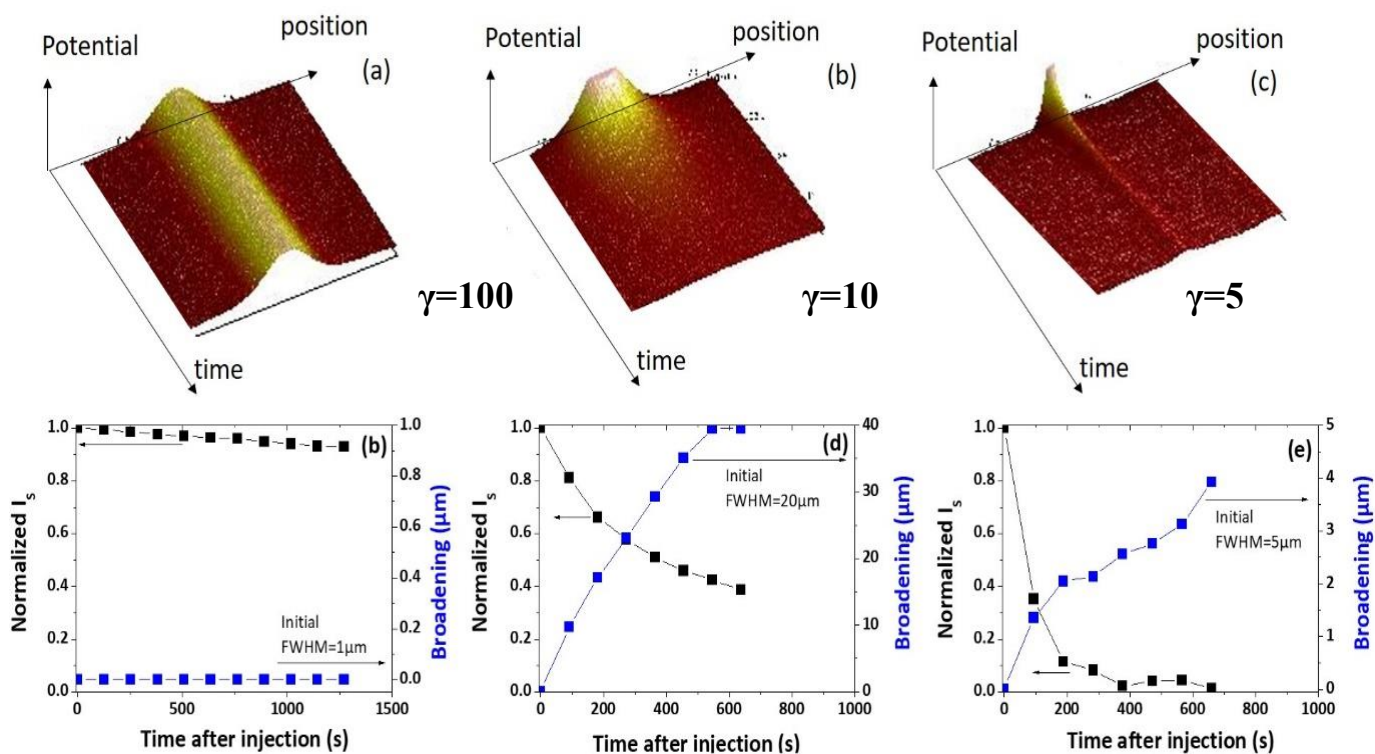
SRM: scanning resistance microscopy

GC Stevens and P Baird, *Nano- and meso-measurement methods in the study of dielectrics* IEEE-Trans. Dielectr. Electr. Insul. 12, 979, 2005

Charging by AFM tip/ Probing by KPFM



Surface potential profile decay and related features after charge injection for SiO_xN_y layers elaborated by plasma process with different gas ratio γ



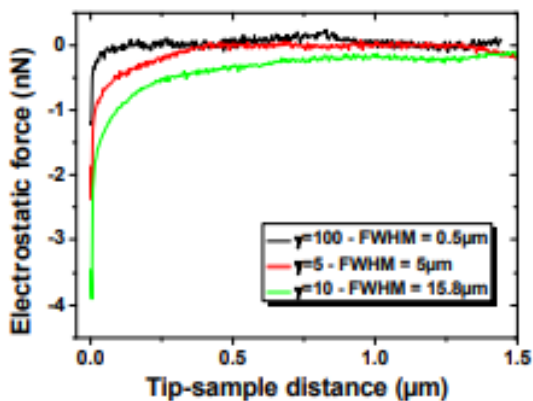
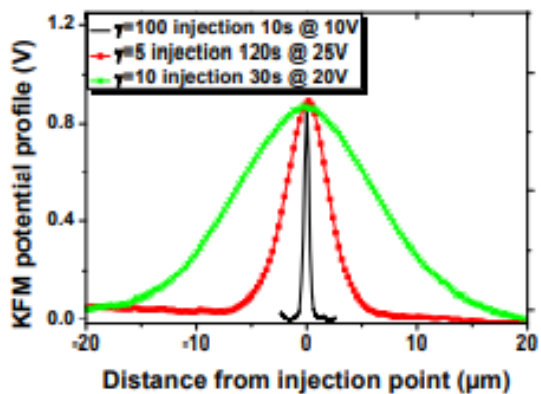
- Obvious dependence of the response with material nature is obtained
- Potential decay in time and peak broadening in time can be quantified
- However, turning to quantitative charge determination is not easy:
 - needs electrostatic modelling for establishing the link between potential and charge distribution;
 - field divergence in the charging step;
 - potential peak width dependent on lift height ;
 - not easy not distinguish in-depth evacuation of charges from lateral spreading.

⇒ As such the method remains essentially a surface analysis

C. Villeneuve-Faure et al, *Space charge at nanoscale...*, Chap 9 in *Electrical Atomic Force Microscopy for Nanoelectronics*, Springer, 2019

Routes to in-depth resolution

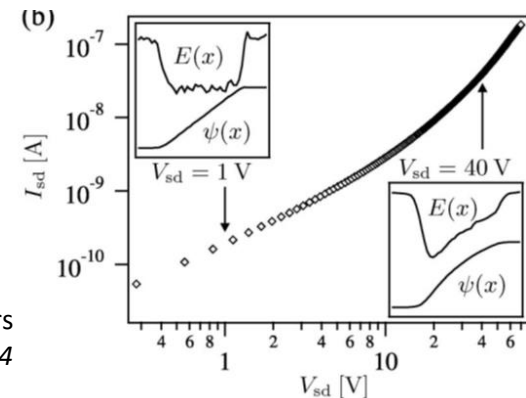
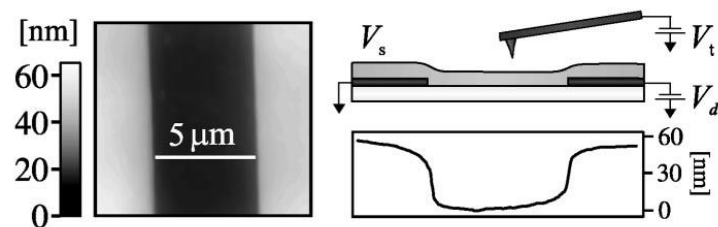
Combining KPFM and FDC



Finite Element Modelling +
strong hypotheses on the
distribution of charges

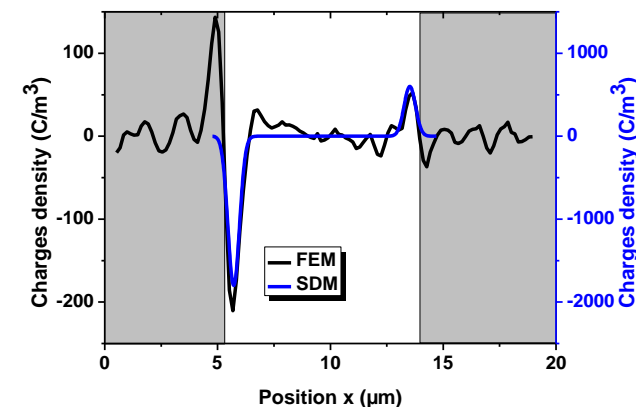
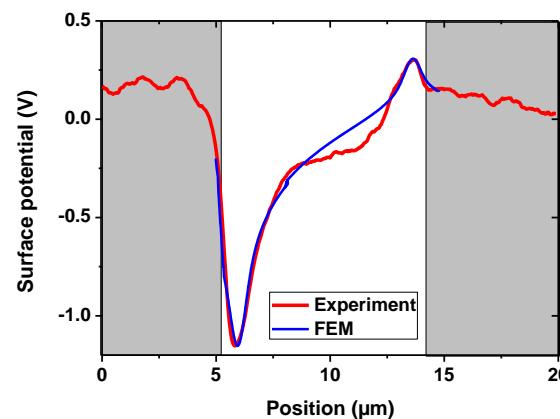
3D space charge density

Working with cross-sections



Field distribution in semiconductors

W.R. Silveira, J.A. Marohn, *Phys. Rev. Lett.* 93, 116104, 2004



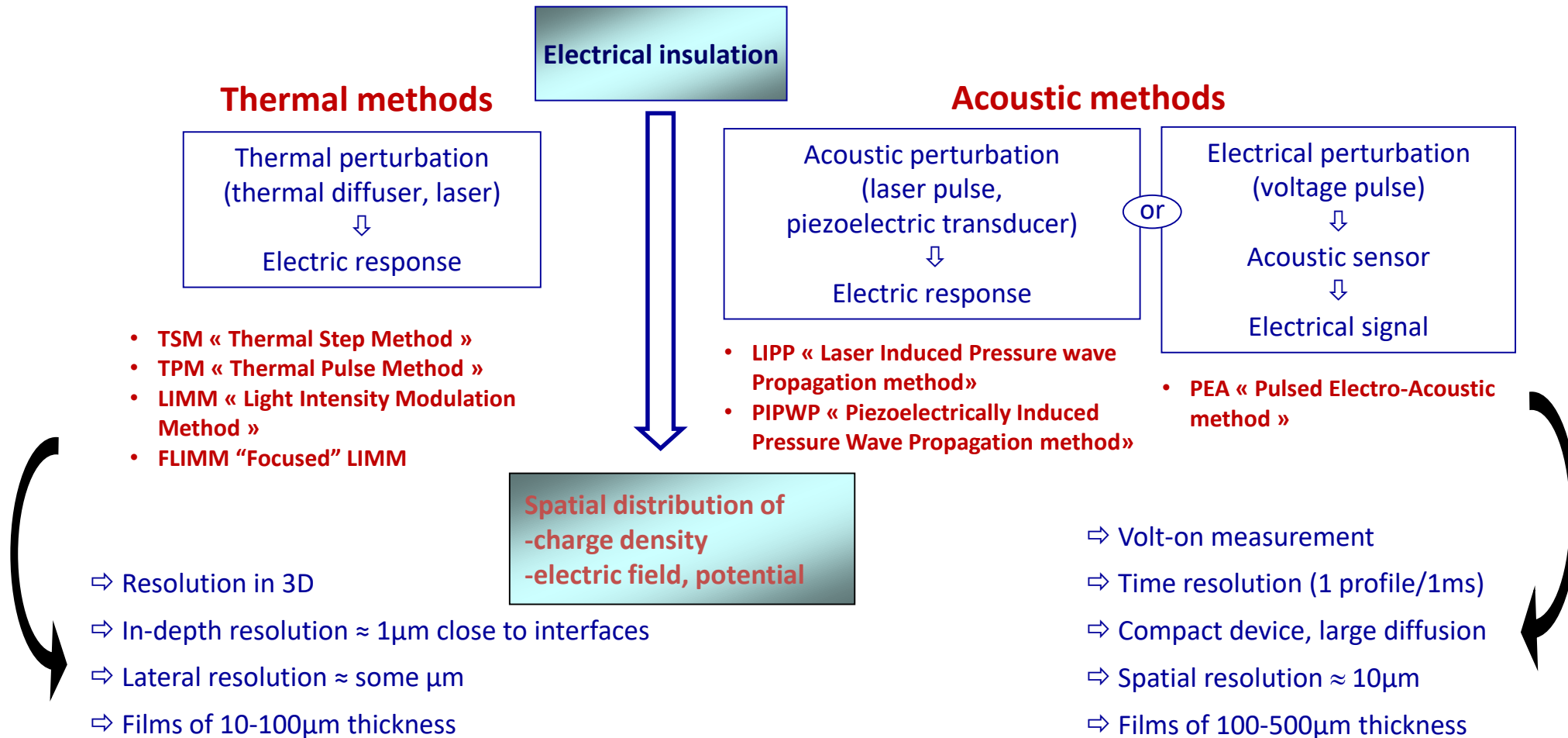
Surface potential profile and charge profiles: in SiNx / crosssection charged under -40V

C. Villeneuve-Faure, L. Boudou, K. Makasheva and G. Teysseire *Nanotechnology* 28, 505701, 2017

'Perturbative' space charge measurement methods

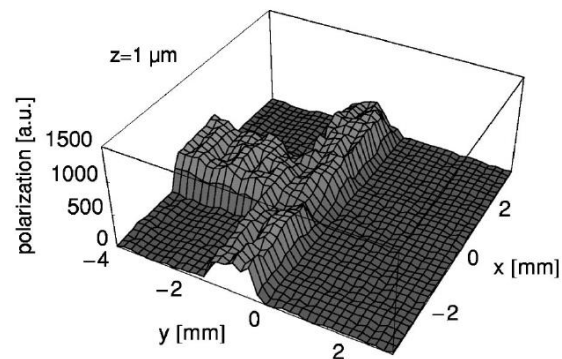
Provide in-depth charge distribution density

Different families depending upon the nature of the excitation



Thermal methods

In-depth resolution: 0.5 μm close to interfaces (Thermal Pulse Method, on a ferroelectric polymer)



A. Mellinger et al. *Appl. Phys. Letts.* 86, 082903, 2005

Lateral resolution: 2 μm (FLIMM)

Bottlenecks:

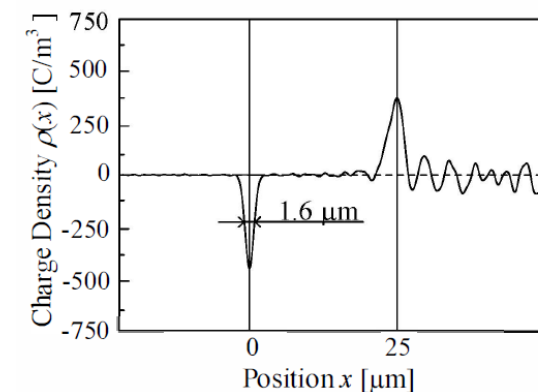
- Signal to noise ratio
- decrease of the resolution with depth
- bandwidth of amplifying circuits 30kHz in (F)LIMM
- need for accurate and simultaneous measurement of surface temperature profile for accurate deconvolution
- lateral temperature diffusion by electrodes

Electro-acoustic method

Resolution: 1.6 μm with a 1 μm thick piezo PVDF sensor

Capacitive charge in 25 μm thick PET

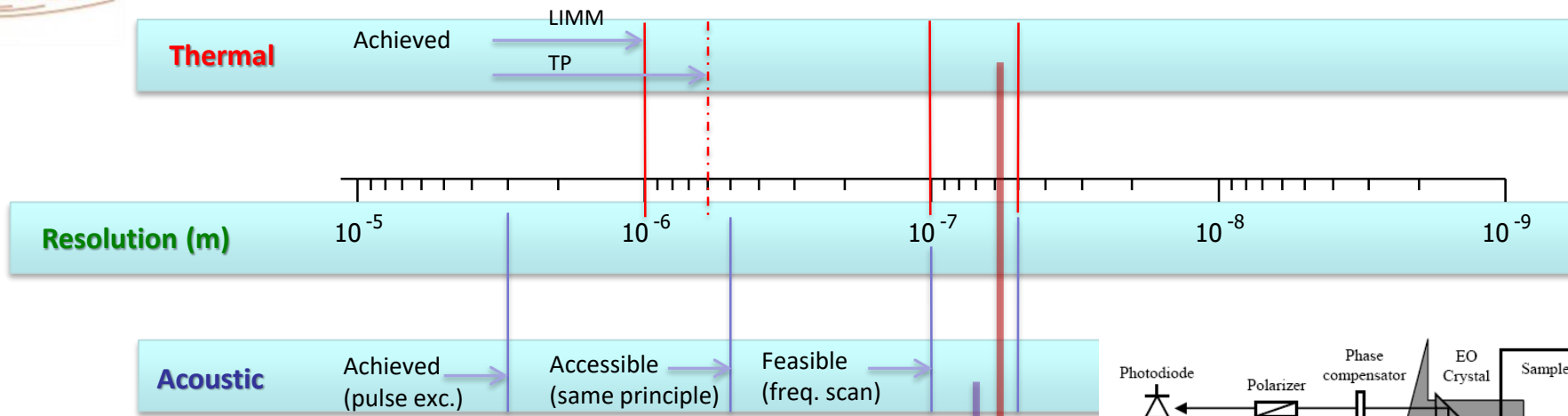
K. Kumaoka, T. Kato, H. Miyake,
Y. Tanaka, *Proc. IEEE-ISEIM*, 389, 2014



Bottlenecks:

- attenuation and dispersion of acoustic waves by the sample (sample-dependent; may impose thin sample);
- optimized deconvolution method
- intrinsic response from the piezo sensor: geometry; electromechanical losses
- acoustic impedance mismatch
- bandwidth of amplifying circuit (500MHz in ordinary PEA system)
- impedance matching of 50ps pulse to sample capacitance;
- low coupling factor of piezo polymer sensors + unknown high frequency behaviour
- SNR

'Perturbative' space charge measurement methods



Feasibility study: 60nm resolution in SiO₂

G. Dagher, S. Holé, and J. Lewiner, IEEE-TDEI 13, 1036, 2006

Thermal / pulse wave

Excitation: fs laser pulse
 → thermal pulse / 100MHz
 + Thermally-induced elastic waves / 10ps

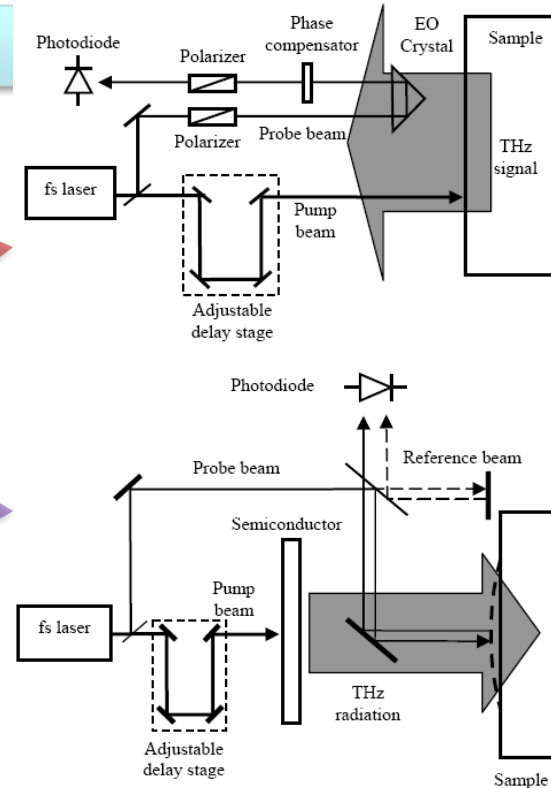
Detection (for both)
 EM field probed by EO crystal
 (ZnTe Pockels cell, sub-THz frequency)

Electro-acoustic method

Excitation: fs laser pulse
 → SC crystal (AsGa) → THz EM pulses
 70ps laser pulse / 1ps field pulses

Detection
 Surface displacement by interferometry
 sub-ps time / 60fm resolution

-Still far from being demonstrated
 -Heavy instrument



Needs for characterization on interfaces at nanoscale with drivers

- Technology evolution towards thinner layers in power engineering /energy applications
- Refine physics at interfaces
- Improve our ways of tailoring internal and outer interfaces

Scanning probe techniques

- do provides information at nanoscale
- however it remains essentially surface analysis tools

Perturbative methods with in-depth resolution

- Convenient
- Bottlenecks with improving spatial resolution



Workshop on NanoElectroAcoustics for Dielectric Materials Characterization

NMDC Conference, Vancouver, B.C., Canada, Dec 12th 2021

Needs and Challenges for the characterization of dielectrics at nanoscale –

Gilbert Teyssedre, CNRS-Laplace Toulouse gilbert.teyssedre@laplace.univ-tlse.fr

Measurement of electric charge distribution in insulators with elastic waves: A search for high spatial resolution

Stéphane Holé, Sorbonne Université

Nanoscale ultrasonic characterization using ultrafast and near-field optical methods

James B. Spicer, Johns Hopkins University