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SÉMINAIRE

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**Ferroelectric-based composite ceramics: design by local field
engineering and tailoring the functional properties by microstructure
control**

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Ferroelectric oxides represent the main functional materials in various microelectronics devices, due to their high permittivity and low loss, switchable polarization by suitable fields, non-linear dielectric character (tunability) suitable for wireless applications, piezo- and pyroelectric properties. For extending the range of applications or for improving their properties, their combination with other types of functional materials is currently proposed. Ferroelectrics present macroscopic properties strongly dependent upon the electric field. Therefore, it is highly important to estimate the dielectric and ferroelectric properties in ferroelectric inhomogeneous materials and in ferroelectric-based composites by taking into account for the fields locally acting inside the material, which are dependent on the microstructures. The proposed approach was to use finite element modeling (FEM) to develop models for realistic microstructures. Two examples of composite materials will be presented:

(i) Ferroelectric-dielectric composites for tunable applications

Ferroelectrics usually show a very high tunability $\epsilon(0)/\epsilon(E)$, but they have a hysteretic response and high permittivity which is detrimental for certain microwave applications. The approach of diluting the ferroelectric material either by reducing the ceramic grain size (nanoceramics) or by their mixing with a low-permittivity dielectric in composites results also in an undesired reduction of tunability. FEM calculations showed that a field inhomogeneity in such composites can give rise even to an enhanced tunability, in specific microstructures or even to a linear field dependent $\epsilon(E)$ in nanostructured ceramics, which would be a very important property for tunability applications. This idea was checked for BaTiO₃ dense nanostructured ceramics prepared by ultrafine powders and sintered by spark plasma sintering down to 90 nm grain size and for PZT porous ceramics with anisotropic porosity, while the possibility to simultaneous increase of both permittivity and tunability by the use of metallic nanoparticles as fillers in a polar low-dielectric matrix was proposed.

(ii) Ferroelectric-magnetic composite ceramics with strain-mediated magnetoelectric coupling.

Bulk magnetoelectric composites are expected to combine multifunctional properties for emergent electromagnetic applications like miniaturized microwaves antenna, metamaterial characteristics, magnetoelectric sensing and bi-tunable characteristics: $\epsilon, \mu = f(E, H)$. Calculations showed that for a better magnetoelectric coupling and high tunability, (0-3) composites are desired. The preparation and properties of a few types of core-shell composites formed by ferroelectric (BaTiO₃, PZT) and magnetic (ferrites, Fe oxides) phases produced by solid state mixing and wet in-situ processing will be shown. Since nanoscale interface reactions can drive to new functional properties, a better control of such reactions and a large ratio of active interfaces can be derived in core-shell structures formed by Mag@BaTiO₃ (where Mag= α -Fe₂O₃ or Ni,ZnFe₂O₄). With appropriate sintering, di- or multiphase composites can be produced: (a) di-phase compositions with fully isolated magnetic regions within BaTiO₃ matrix: (0-3) connectivity, and (b) multi-phase compositions. Variable amounts of secondary phases with contrasting magnetic coercivities (Fe₃O₄, BaFe₁₂O₁₉ and Ba₁₂Fe₂₈Ti₁₅O₈₄) were induced by controlled interface reactions. As result, wasp-waisted constricted M(H) loops were determined as result of the formation of different amounts of hard/soft phases at interfaces. Dielectric constant of 100-300 and low losses by comparison with other BaTiO₃-based composites and a reduced hopping conductivity were typically found as result of the isolation of the low-resistivity magnetic phase.