Atomically engineered oxide heterointerfaces: new opportunities for nanoionics and nanoelectronics

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O.34

A new approach for tayloring tunability and permittivity values by using grain size reduction at nanoscale

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The nonlinear dielectric properties of ferroelectrics were intensively studied due to their potentiality for miniaturization of microwave components and integration in microelectronic circuits. Such applications demand a level of tunability of $n = \varepsilon(E)/\varepsilon(0)$ >1.5, moderate permittivity below 1000 and losses below 3%. The large majority of publications in the field of tunable materials report the fulfilling of technological requirements by adding to the ferroelectric a low permittivity linear dielectric. We propose a new approach to use the grain size reduction at nanoscale in dense $BaTiO_3$ - based ceramics for tailoring the dielectric and tunability properties.

The tunable properties of pure and substituted $BaTiO_3$ - based nanostructured ceramics with grain size of (1 μm - 90 nm) were investigated. A reduced permittivity down to ~500 and almost linear $e(E)$ dependence in a large field range is typical to fine grained nanoceramics. The observed behavior was explained by considering the nanostructured ceramic as a composite formed by ferroelectric grains and low-permittivity non-ferroelectric grain boundaries, whose volume fraction increases when reducing grain size. An original approach was developed to calculate the local field distribution in the nanostructured ceramic with realistic microstructure using a combination of finite element and Monte Carlo models. The experimental data combined with theoretical model show that the reduction of grain size in ferroelectric dense materials can be an effective alternative route to accomplish the tunability applications requirements.

O.35

Structural and electrical properties of a new $Ba_2NdTi_{2+x}Nb_{3-x}O_{15-x/2}$ solid solution with tetragonal tungsten bronze phase

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A ferroelectric tetragonal tungsten bronze (TTB) phases of general formula $Ba_2RETi_2Nb_3O_{15}$: RE = Nd, Sm, were previously prepared by two low temperature routes, sol-gel synthesis and solvothermal synthesis[1]. The relative permittivity *vs* temperature data for these samples exhibits two unusual features with ferroelectric materials: first, a significant hysteresis between heating and cooling cycles in their phase transition behaviour, and second, a huge depression in the Curie-Weiss temperature T_0, which is very different from the behaviour usually observed in first order ferroelectrics. Both effects were attributed to the complex nature of their TTB-related crystal structures.

In this work, based on these studies, a wide range of solid solution of general formula $Ba_2NdTi_{2+x}Nb_{3-x}O_{15-x/2}$ (BNTN) with TTB phase were prepared by low temperature solvothermal synthesis. Extensive solid solution with oxygen deficiencies forms over the range 0 < x < 1.5. The existence of this BNTN solid solution with oxygen deficiency has not been reported previously. X-ray diffraction (XRD), scanning electron microscopy (SEM) and Powder neutron diffraction (ND) were used to determine the solid solution limit. According impedance measurements, samples were electrically-homogeneous, and highly insulating. The total conductivity increases when the Ti/Nb ratio increases. Besides, the relative permittivity shows a large dependence on the composition: the *Tc* and relative permittivity values decrease when x increases.

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O.36

Lessons from $Bi_2Ti_2O_7$: One step closer to finally understanding dielectric relaxation in pyrochlores

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Bismuth titanate with the pyrochlore-like stoichiometry ($Bi_2Ti_2O_7$) has been the focus of several investigations by various groups around the world. Over the last 40 to 50 years, these investigations have led to several very conflicting and contradicting observations. To help resolve these, here, dense phase-pure $Bi_2Ti_2O_7$ polycrystalline ceramic pellets were obtained via co-precipitation synthesis methods followed by microwave sintering techniques. Density functional theory was used to model the structure of the pyrochlore, from which the theoretical X-ray diffraction patterns were compared with experimental patterns and confirmed the phase purity in the powder as well as in the pellets. As such, this maiden achievement allowed for primary thermophysical, crystallographic, and dielectric characterization of this ceramic compound. Discrepancies among reports in literature regarding the structure, stability, and supposed ferroelectricity of this material are discussed and clarified. As a result, a modification to the phase diagram of the Bi_2O_3-TiO_2 system is proposed. In addition, and contrary to prior reports, the dielectric characterization of $Bi_2Ti_2O_7$ reveals a linear dielectric with high permittivity values at room temperature (115 at 500 kHz), and more remarkably, a decrease in permittivity with decreasing temperature unlike the typical dielectric relaxation in ceramics. Based on experimental data and computational simulations, and by contrasting the dielectric response of $Bi_2Ti_2O_7$ with that of well known bismuth pyrochlroe compounds, the necessary and sufficient chemical and crystallographic conditions for dielectric relaxation in pyrochlores will be presented.

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Atomically engineered oxide heterointerfaces: new opportunities for nanoionics and nanoelectronics

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In recent years, the advance in fabrication of oxide heterostructures with atomically engineered interfaces provides new opportunities for both nanoelectronics and nanoionics. Well-defined oxide heterointerfaces, where an oxide is in intimate contact with another, can exhibit exceptional electronic, ionic, optical, and catalytic properties that are remarkably different from those of their bulk counterparts by establishing a new equilibrium of the distribution of electrons or ions across the heterointerface. Here, we will firstly present our efforts in epitaxial growth of atomically flat thin films of oxygen-ions conductors, for example, gadolinia-doped ceria[1]. Secondly, we show that oxygen-redistribution related redox reactions at oxide interfaces can provide an effective way to create two-dimensional electron gases (2DEGs) between two insulating complex oxides[2], the basis for a new generation of all-oxide electronic devices. By careful oxygen engineering, we obtain 2DEGs with unprecedented mobilities greater than 1.4×10^5 $cm^2V^{-1}s^{-1}$ at these interfaces based on insulating complex oxides.

Reference:

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O.38

Multiferroic epitaxial thin films and heterostructures. Strain effects.

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During the last years, multiferroic materials have received much attention due to possible integration in advanced applications, such as data storage, communications or even energy harvesting. Most of those applications require using thin films and thus, development of multiferroic thin films and related heterostructures has been a hot area of research. Broadly speaking multiferroic thin films activity has focused either on single-phase multiferroic or more complex heterostructures involving materials with distinct ferroic (ferroelectric FE and magnetic M) orders. Whereas in the former an intimate coupling of FE and M orders can be expected and the unavoidable substrate-induced strain can only contribute to modify the magnetoelectric ground state, in the later strain is thought to be the coupling mechanism between FE and M phases.

In this presentation we shall overview the role of substrate and interface-induced strain effects on single phase multiferroic materials ($YMnO_3$) and on bilayers of ferromagnetic/ ferroelectric materials ($CoFe_2O_4/BaTiO_3$).

Materials are gown by pulsed laser deposition on suitable substrates. It will be shown that $YMnO_3$ can be grown having a cycloidal magnetic structure that give rise to ferroelectricity with the rare property of its polarization direction being switchable by 90° with appropriate magnetic field. Furthermore, substrate-induced strain dramatically modifies the magnetic order and cancels ferroelectricity, by changing the Mn-O-Mn bond topology.

On the other hand, it will be shown that strain in multilayers plays a dual role. It clamps the film to the substrate but also allows to transmit any film-lattice modifications through the structure. Using this approach, we will show that the dielectric permittivity of $BaTiO_3$ can be modified by a suitable magnetic field applied to $CoFe_2O_4/BaTiO_3$ heterostructures. The relevance of $CoFe_2O_4/BaTiO_3$ stacking order on the observed magnetoelectric coupling will be discussed.