ELECTRONIC CERAMICS DEPARTMENT

K-5

The Electronic Ceramics Department is active in the research of the synthesis, properties and applications of ceramic materials for electronics and energetics, mainly complex multifunctional materials and structures that can perform multiple functions (multifunctional materials). The materials of interest include piezoelectrics, ferroelectrics, relaxors, multiferroics, conductive oxides, low-dimensional magnets and cuprate superconductors. The emphasis is on the creation of properties by the synthesis and structure on the nano-, micro- and macro-levels. The group also works on the principles of basic technologies for ceramic pressure sensors, ceramic MEMS and flexible electronics.

We continued with the work on **lead-free piezoelectrics**. In collaboration with colleagues from the Technical University of Darmstadt, Germany, we studied the uniaxial deformation of the ferroelectric KNbO₃ single crystal in [010] and [101] orientations at room temperature. The mechanical dislocation and ferroelectric domain structure were investigated using piezo-response force microscopy (PFM). The results suggest that the dislocations act as nucleation and pinning sites for the ferroelectric domains, leading to a local increase in the density of the ferroelectric domain walls.

We continued with the work on **polycrystalline BiFeO**₃. We performed a comparative study of uncharged and charged domain walls (DWs) in BiFeO₃ ceramics from the point of view of the atomically resolved strain and structure using scanning-transmission electron microscopy. We showed that the uncharged $\{100\}$ pseudo cubic (pc)-

type DWs have a larger associated lattice strain than the charged-"tail-to-tail" {100}pc-type DWs, and we were able to explain the result as a pure intrinsic lattice mismatch. As the {100}pc-type DWs have been experimentally shown to be intrinsically different in strain distribution and structure, we assume that their role in the switching mechanism will be different, depending on their particular strain and charged state.

Together with colleagues from the Norwegian University of Science and Technology, Trondheim, Norway, we published a review paper on the alternating-current (AC) properties of domain walls, which includes our work on $BiFeO_3$ ceramics, in particular the implications of the domain-wall conductivity in the emergent macroscopic piezoelectric response.

In collaboration with colleagues at the National Institute of Chemistry, EPFL, Lausanne, Switzerland, Materials Center Leoben, Austria, and the Tokyo Institute of Technology, Japan we conducted an atomic resolution study using aberration-corrected scanning transmission electron microscopy complemented by Raman spectroscopy and directly revealed, visualized, and quantitatively described static 2–4-nm polar nanoclusters in the nominally nonpolar cubic phases of **barium titanate-based ceramics**. These results helped us understand the atomic-scale structure of disordered materials and may help clarify ambiguities about the dynamic-versus static nature of nano-sized clusters [Figure 1].

 $(Na_{1/2}Bi_{1/2})TiO_3$ -BaTiO₃ (NBT-BT) lead-free piezoceramics are of interest for high-power piezoelectric applications where the hardening of the electromechanical response is of paramount importance. In collaboration with colleagues from Technical University Darmstadt, Germany, we conducted a study on NBT-BT ceramics aimed at identifying the hardening efficiency of the so-called composite approach. The approach consists of dispersing second-phase inclusions, in this case ZnO, in the ceramics to provide pinning effects and thus reduce the domain-wall displacements and the associated losses. With the support of higher harmonic polarization measurements,

A sthe { 100 }pc-type DWs have been experimentally shown by different in strain distribution and structure, we assume in the switching mechanism will be different, depending on strain and charged state. ith colleagues from the Norwegian University of Science , Trondheim, Norway, we published a review paper on the ent (AC) properties of domain walls, which includes our



HAADF STEM image

Figure 1: High-angle annular dark-field (HAADF) and annular bright-field (ABF) STEM image of $Ba_{o,o}Sr_{o,4}$ TiO₃ in [110] zone axis with corresponding Ti vs (Ba, Sr), O vs (Ba, Sr), and O vs Ti displacements presented in the form of polar plots. The inset in b shows the perovskite unit cell viewed along [110] axis. Dashed rectangles mark areas where displacements were determined. Dashed circles mark areas where majority of the individual types of displacements are present. Displacements of cations and anions do not coincide, indicating noncubic structure.

performed in our lab, we were able to explain the superior thermal stability of the mechanical quality factor in NBT-BT/ZnO composites as a consequence of robust mechanical pinning effects provided by the inclusions.

In collaboration with colleagues from Friedrich-Alexander-University Erlangen-Nürnberg, Germany, we used piezoforce microscopy (PFM) and transmission electron microscopy to investigate the domain structure of $(Na_{1,2}Bi_{1,2})$ TiO₂ ceramics. Ferroelectric domain switching was observed by applying a sufficiently large electric field, no change in domain configuration was observed in the samples subjected to uniaxial compressive stresses up to 750 MPa.

In ferroelectrics, the mechanisms of hardening by acceptor doping are commonly associated with domain-wall pinning effects provided by oxygen vacancies. In lead-based relaxor ferroelectrics, these mechanisms are com-

PhD students Oana-Andreea Condurache and Matej SadI won four prizes for presenting their dissertation results at international conferences.

plicated by the nano-polar structure of these materials and their dynamic contribution to the electromechanical properties. To shed light on this issue, we systematically investigated the hardening effects by Mn doping in Pb(Mg,Nb)O,-PbTiO, (PMN-PT). We found that the oxygen-vacancies-related pinning manifests similarly both in the ergodic and non-ergodic relaxor

phases of the PMN-PT. By reducing the freezing temperature, Mn doping was found to be an efficient approach to improving the thermal stability of the electrocaloric response.

Poling-field-induced changes in the PMN-PT relaxor ferroelectric ceramics around the morphotropic phase boundary (MPB) were investigated, with a detailed examination of the effects of the poling procedure on the piezoelectric and dielectric response, as well as the response of the crystal lattice and the ferroelectric domains. We found that AC poling on the monoclinic (M) side of the phase diagram is more effective than DC poling from the point of a much lower AC field being needed to obtain a similar d33 response as with DC poling. This result was supported by in-situ XRD measurements of the 30 PT M composition, which show a large-strain response at significantly lower AC than the DC field. Furthermore, the Cm M phase possesses low-angle nanodomain walls



Figure 2: Multifunctional cantilevers as working elements in solidstate cooling devices: microstructures of the sandwich-like structure of cantilevers in cross-section, consisting of two Pb(Mg,Nb)O₃ ceramic layers and platinum, sintered at different temperatures. When an electric field (E) is applied to the upper ceramic layer, the cantilever bends, as shown schematically in the bottom inset. Experimental setup for testing the proof-of-concept device with cantilevers arranged in a cascade structure is shown on the right.

and exhibits a gradual "cascade-like" motion of the domain walls that starts immediately at low fields (~2 kV/cm) and saturates roughly at 15 kV/cm, contributing to large strains in the material. The tetragonal-like M phase, however, shows sudden domain switching roughly at the coercive field, with no obvious lattice strain up to 30 kV/cm.

In collaboration with colleagues from the Condensed Matter Department, JSI, and from the Technical University Darmstadt, Germany, we investigated the electrocaloric (EC) properties of 0.9Pb(Mg_{1/2}Nb_{2/2})O₂-0.1PbTiO₂ (PMN-10PT) ceramics prepared by the conventional columbite route and mechanochemical synthesis. The samples were able to withstand DC electric fields up to 115 kV/cm, resulting in very high EC temperature changes ($\Delta T_{\rm FC}$) of 2.37 K at 107 °C. In contrast, the ceramics prepared by the columbite route could only withstand ~ 60 kV/cm and therefore exhibited a much lower $\Delta T_{\rm FC}$. A detailed examination of the microstructure revealed that the respective samples contained a semi-crystalline secondary phase at the triple junctions of the grains, which presumably provided an easy pathway for current propagation. In contrast, in the PMN-10PT obtained by mecha-

nochemical synthesis, the grain boundaries were clean and MgO inclusions were identified. These microstructural features contributed to a higher electrical breakdown field of the ceramics.

Despite the challenges of practical implementation, EC cooling remains a promising technology because of its good scalability and high efficiency. To provide a further step in this application area, we fabricated multifunctional

Prof. dr. Goran Dražić received a Zois Award for top achievements in the field of the transmission electron microscopy of materials.

cantilever structures made of relaxor ferroelectric PMN-PT, which were stacked in a cascade, forming a proof-of-concept device. Functional testing of the structure revealed that the key element of the device's performance is the poor heat transfer through the cantilever contacts. The study thus clearly showed that further engineering will have to be focused on lowering the thermal contact resistance for an efficient EC operation of the cantilever cascade [Figure 2].

Together with collaborators from McMaster University, Canada, Oakridge National Laboratory in the US, and ANSTO's Australian Synchrotron, we continued the research on cuprate superconductors, $La_{16-x}Nd_{04}Sr_{x}CuO_{4}$ (Nd-LSCO). We first used resonant X-ray scattering to measure the evolution of electronic nematicity and charge density wave order with hole doping. We found that electronic nematicity - a rotational symmetry breaking of the electronic structure - is associated with the onset of the pseudo-gap phase. Nd-LSCO exhibits a substantial decrease in electronic nematicity, either by increasing the temperature to the onset of the pseudo-gap phase or by increasing the doping through the pseudo-gap quantum critical point.

Moreover, using elastic and inelastic neutron scattering measurements on single crystals of x = 0.125, 0.19, 0.24, and 0.26 we showed that two-dimensional, quasistatic, parallel spin stripes have an onset at temperatures such that the parallel spin stripe phase extends beyond p_* and envelops the entirety of the superconducting ground states in this system. Our measurements of 2D TN and the onset of 2D parallel stripes at optimal and high hole-doping levels in Nd-LSCO, allowed us to complete the phase diagram for 2D parallel stripes and examine their relation to superconductivity.

In the framework of our studies on **low-dimensional magnetism**, we continued our investigations of **manganates**. We performed the synthesis, structural and magnetic characterisation of the Ba_{1-x}La_{1-x}MnO₄₊₈ ($0 \le x \le 0.5$) series. We found that each member of the Ba_{1-x}La_{1-x}MnO₄₊₈ series exhibits the same spin-glass behaviour previously found in the x = 0.2. Moreover, Tg varies with x reaching a maximum of 26.4(4) K for x = 0.20 [Figure 3].

Furthermore, using X-ray absorption near-edge structure (XANES) studies we found out that the oxidation state of Mn in the Ba_{1-x}La_{1+x}MnO_{4+δ} samples varies with *x*: for $x \le 0.2$ Mn is in +3.0(1) oxidation state only, whereas a mixed +2/+3 oxidation state was found for x > 0.2. The origin of the spin-glass state in the Ba_{1-x}La_{1+x}MnO_{4+δ} series shows two regimes depending on the oxidation state of the Mn.

In collaboration with the Department of Advanced Materials, JSI, we investigated the relaxor ferroelectric domain structure in the epitaxial $0.67Pb(Mg_{1/3}Nb_{2/3})O_3-0.33PbTiO_3$ (PMN-33PT) films on different substrates. PFM analysis was used to show that the domain structure of PMN-33PT films is sensitive to the compressive strain induced in the films. A relaxor-like behaviour was observed at a strain state below 1.1%, while irregularly shaped ferroelectric domains were observed at a higher compressive strain (> 1.9%). The results suggest that epitaxial strain engineering could be an effective approach to tailor and improve the functional properties of **relaxor ferroelectric thin films**.

Patterning of nanostructures of functional-oxide materials by inkjet printing of solution-based inks was studied with colleagues from the Condensed Matter Physics Department, JSI, Faculty of Mathematics and Physics, University of Ljubljana, and CENN Nanocenter. The importance of contact line mobility– either pinned or mobile – on the deposit morphology, either dome-like, flat or ring-like, was addressed. By adjusting the ink solvent composition and controlling the substrate wetting behaviour, deposits with a uniform thickness in the nanometre range could be printed.

Within the field of chemical solution deposition of lead-free ferroelectric thin films our focus was on barium-titanate-based solid solutions with an enhanced ferro- and piezoelectric response.

We continued the research of **thick films** of environmentally benign piezoelectrics based on $\rm K_{0.5}Na_{0.5}NbO_3$ on ceramic substrates for energy harvesting and ultrasound transducer applications. The research was conducted within the Proteus project in collaboration with researchers from the University of Tours, France.

We studied how sintering in different atmospheres affects the structural, microstructural, and functional properties of ~30-µm-thick films of $K_{0.5}Na_{0.5}NbO_3$ (KNN) modified with 0.38 mol% $K_{5.4}Cu_{1.3}Ta_{10}O_{29}$ and 1 mol% CuO. The films were **screen printed** on platinized alumina substrates and sintered at 1100 °C in oxygen or in the air with or without the packing powder. Thick films sintered in oxygen exhibit a piezoelectric d_{33} coefficient of 64 pm/V and an effective thickness coupling coefficient k_t of 43%, as well as very low mechanical losses of less than 0.5%, making them promising candidates for lead-free piezoelectric energy-harvesting applications. The study was conducted in collaboration with researchers from the University of Tours, France.

We continued with the preparation of **thick films using an aerosol deposition method**. The aerosol deposition system is a part of the Laboratory for the Ultracool Preparation of Complex Oxides, for which the financial support was granted by **the Director's fund ULTRACOOL project**. We focused on optimization of processing parameters of functional 0.9Pb(Mg_{1/3}Nb_{2/3})



Figure 3: Ba_{1-x}La_{1-x}MnO₄₊₆ ($0 \le x \le 0.4$) series exhibits a rare anisotropic spin-glass behaviour. The freezing temperature, T_g , varies with x, reaching a maximum of 26.4(4) K for x = 0.20. The oxidation state of Mn is also dependent on x.

Prof. dr. Mojca Otoničar and Prof. dr. Tadej Rojac were awarded 'Excellent in science' by the Slovenian research agency in the field of electronic components and materials, for disclosing the influence of polar disorder on the response dynamics of ferroelectric materials. The results of this study were published in the prestigious international journal Advanced Functional Materials.



Figure 4: Room temperature energy storage properties of PMN-10PT thick films deposited on stainless steel; recoverable energy density (U_{ree}) and efficiency $(\mathbf{\eta})$ as a function of electric field (E). Inset (bottom): microstructure of the sample in cross-section taken with a scanning electron microscope. Inset (top): Measurement of polarization (P) as a function of E.

 O_3 -0.1PbTiO_3 (PMN-10PT) thick films deposited on stainless steel. The as-deposited films withstand electric fields of 900 kV/cm and exhibit promising room-temperature energy-storage properties: the recoverable energy density reached 7.0 J/cm³ with an energy-storage efficiency of ~70%. A post-deposition stress relaxation by annealing at 500 °C further improved the recoverable energy density, leading to 9.8 J/cm³ with an efficiency of ~80% [Figure 4]. The energy-storage performance exhibited excellent temperature stability up to 200 °C and electric-field cycling stability up to 16 million cycles.

In collaboration with the Laboratory for Refrigeration and District Energy, the Faculty of Mechanical Engineering, University of Ljubljana we prepared multilayer composites using the aerosol-deposition method. The composite was made of $Al_2O_3/Al/Al_2O_3$ layers on a magnetocaloric gadolinium substrate. Such ceramic-metal multilayers represent a simple, reliable, and cost-effective approach to functionalizing and protecting existing magnetocaloric substrates and provide an excellent starting point for the development of future electrowetting-on-dielectric devices.

We progressed the research on the **cold sintering** of functional oxides in our **ULTRACOOL** laboratory, expanding the sintering from BiFeO₃ ceramics to (K,Na)NbO₃ perovskites and composites with piezoelectric polymers (PVDF). While the optimization of all parameters for the successful cold sintering of the ceramic compounds is still an ongoing process, the first measured electromechanical properties of the sintered ceramics are very promising and show a great perspective of cold-sintered oxides for actuator and energy-storage applications. Preliminary studies show that the main benefits of the cold sintering of ceramics are, besides the energy savings due to the low-temperature processing, their dielectric breakdown strength that allows high voltages applied to the materials without their disintegration, as well as high dielectric permittivity and low dielectric losses.

A miniature ozone generator in the form of a **monolithic three-dimensional ceramic structure** was fabricated by low-temperature co-fired ceramic (LTCC) technology utilising the principle of electric discharge. A multilayered ceramic structure with dimensions of 63.6 mm × 41.8 mm × 1.3 mm included integrated electrodes, buried channels and cavities in micro and millimetre scales. The highest ozone concentration in the LTCC-based ceramic device was around 1.1 vol. % at a voltage of about 7 kV and an oxygen flow rate of 10 ml/min. Its yield is comparable to much larger ozone generators available on the market. The LTCC technology was also implemented to fabricate a 3D structure with a buried cavity for the radio-frequency dielectric heating of polar liquids. The power used to heat water in the cavity with the volume of 0.3 mL ranges from 5 to 40 W. This novel application of dielectric heating could enable the miniaturisation of microfluidic systems. Furthermore, LTCC and high-temperature cofired ceramic (HTCC) materials were tested to fabricate three-dimensional power modules. A dual-pulse electrical test of the power modules confirmed, the quality of metallization, wire bonding, and assembly in combination with selected materials. The research was conducted in collaboration with the company KEKO Equipment and Centre of Excellence NAMASTE.

As part of the **KET4CP** project "Manufacturing of invisible interconnections from solutions of low-cost transparent conduction oxides by screen printing", with project partners RC eNeM and the Institute of Solid State Physics from Latvia, we developed a novel method for fabricating transparent zinc oxide-based thin films on glass from low-cost precursor solutions by chemical solution deposition and screen-printing processes. A high optical transmittance of over 90 % and electrical conductivity of 0.002 S/cm were obtained for 150-nm-thick films processed by screen printing. The solution-derived screen-printing process was successfully demonstrated in the large-scale production line of the company RC eNeM. The Slovenian project partners were awarded the Silver Recognition of the Zasavje Regional Chamber of Commerce in 2021 for the invention.

In collaboration with the Condensed Matter Physics Department, JSI, we studied the memory effect in polydomain liquid-crystal elastomer particles dispersed in a polymer. We confirmed the applicability of flow-induced shear stress in the alignment and deformation of liquid crystal elastomer microparticles in a viscous resin by analysing the rheological behaviour of the composite in terms of temperature and shear-rate-induced changes.

In collaboration with the Condensed Matter Department, JSI, and colleagues from Morocco and France, we investigated lead-free piezoelectric composites for energy harvesting. The composites consist of $Ba_{0.85}Ca_{0.15}Zr_{0.10}Ti_{0.90}O_3$ nanoparticles embedded in a biodegradable polymer. The piezoelectricity and ferroelectricity of the nanoparticles before and after embedding in the polymer matrix were determined by PFM. The maximum power density achieved in the prepared samples was 7.5 mW/cm³.

In collaboration with the Slovenian company Lotrič Metrology d.o.o., we developed an economical process for producing a non-biological fluid for testing protective medical equipment according to EN 14683 in accordance with ISO 22609: 2004. The technical improvement "Process of preparing a fluid for testing medical protective equipment" was registered.

Some outstanding publications in the past year

- Kuščer, Danjela, Drnovšek, Silvo, Levassort, Franck. Inkjet-printing-derived lead-zirconate-titanate-based thick films for printed electronics. Materials & design. 2021, vol. 198, str. 109324-1-109324-9. ISSN 0264-1275. DOI: 10.1016/j.matdes.2020.109324. [COBISS.SI-ID 36937475]
- Benčan, Andreja, Oveisi, Emad, Hashemizadeh, Sina, Veerapandiyan, Vignaswaran K., Hoshina, Takuya, Rojac, Tadej, Deluca, Marco, Dražić, Goran, Damjanović, Dragan. Atomic scale symmetry and polar nanoclusters in the paraelectric phase of ferroelectric materials. *Nature communications*. 2021, vol. 12, no. 1, str. 3509-1-3509-9, ilustr. ISSN 2041-1723. https://www.nature.com/articles/s41467-021-23600-3, DOI: 10.1038/s41467-021-23600-3. [COBISS.SI-ID 65810179]
- Bradeško, Andraž, Fulanović, Lovro, Vrabelj, Marko, Matavž, Aleksander, Otoničar, Mojca, Koruza, Jurij, Malič, Barbara, Rojac, Tadej. Multifunctional cantilevers as working elements in solid-state cooling devices. *Actuators*. [Online ed.]. 2021, vol. 10, no. 3, str. 58-1-58-13. ISSN 2076-0825. DOI: 10.3390/ act10030058. [COBISS.SI-ID 55105027]
- 4. Šadl, Matej, Condurache, Oana, Benčan, Andreja, Dragomir, Mirela, Prah, Uroš, Malič, Barbara, Deluca, Marco, Eckstein, Udo, Hausmann, Daniel, Khansur, Neamul Hayet, Webber, Kyle Grant, Uršič, Hana. Energy-storage-efficient 0.9Pb(Mg_{1/3}Nb_{2/3})O₃-0.1PbTiO₃ thick films integrated directly onto stainless steel. Acta materialia, ISSN 1359-6454. [Print ed.], Dec. 2021, vol. 221, str. 117403-1-117403-11, ilustr., doi: 10.1016/j. actamat.2021.117403. [COBISS.SI-ID 81773059]
- Otoničar, Mojca, Bradeško, Andraž, Salmanov, Samir, Chung, C. C., Jones, Jacob L., Rojac, Tadej. Effects of poling on the electrical and electromechanical response of PMN-PT relaxor ferroelectric ceramics. Open ceramics. 2021, vol. 7, 100140-1-100140-14, ilustr. ISSN 2666-5395. DOI: 10.1016/j.oceram.2021.100140. [COBISS.SI-ID 67640323]

Awards and Appointments

- 1. Oana-Andreea Condurache: Awarded 3rd place at the Virtual Workshop Contest: YCN Pitch me your Idea!, Young Ceramicst Network Pitch Contest
- 2. Oana-Andreea Condurache: Awarded 3rd place at the Student Paper Contest, 27th Annual Meeting the Slovenian Chemical Society (SKD 2021)
- 3. Danjela Kuščer: Silver Award for Innovation: "Manufacture of transparent electrodes from solutions of affordable conductive oxides using screen printing" awarded by Chamber of Comerce Zasavje
- 4. Andreja Benčan Golob, Goran Dražić, Barbara Malič, Mojca Otoničar, Tadej Rojac, Hana Uršič Nemevšek, Achievement for the paper "Connecting the Multiscale Structure with Macroscopic Response of Relaxor Ferroelectrics" was included in the selection Excellent in science ARRS 2021
- Matej Šadl: "Alessandro de Vita" award for curiosity and multidisciplinary approach at Crossnano Crossborder Workshop in Nanoscience and Nanotecnology 2021
- 6. Matej Šadl: Awarded 2nd place at student competition "2021 Joint ISAF ISIF-PMF virtual conference"

Patent granted

1. Vid Bobnar, Barbara Malič, Aleksander Matavž, A method for producing polymeric surface modification layers, SI25887 (A), Slovenian Intellectual Property Office, 31. 03. 2021.

INTERNATIONAL PROJECTS

- H2020 ATHENA; Implementing Gender Equality Plans to Unlock Research Potential of RPOs and RFOs in Europe Prof. Barbara Malič
 - European Commission
- H2020 QMatCh; Towards Quantum States of Matter via Chemistry under Extreme Conditions Asst. Prof. Mirela Dragomir
 - European Commission
- Cold Sintering of Complex Oxide Materials Dr. Mojca Otoničar
- Slovenian Research Agency
- 4. Low Bandgap Ferroelectric Solar Cell Absorbers: Synthesis and Characterization

Prof. Hana Uršič Nemevšek Slovenjan Research Agency

- Slovenian Research Agency
 Interface Stability of Piezoelectric Ceramic Oxides Prof. Tadej Rojac
- Slovenian Research Agency
- Environmental Benign Sodium Potassium Niobate-based Thick Films for Piezoelectric Energy Harvesting Applications Prof. Danjela Kuščer Hrovatin
- Slovenian Research Agency 7. Multiferroics for Solid-State Cooling Applications Prof. Hana Uršič Nemevšek Slovenian Research Agency



- Processing Structure Properties Study of Environmentally Friendly Piezoelectric 8 Nanoparticles of Tailored Surface Morphology Prof. Andreja Benčan Golob
 - Slovenian Research Agency
- 9 High-Pressure Synthesis and Characterization of Selected Ferroics Dr. Kristian Radan Slovenian Research Agency
- 10. Crystal Growth and Magnetic Properties of Double Perovskites Asst. Prof. Mirela Dragomir
- Slovenian Research Agency
- Porous Lead-Free Relaxor Ferroelectric Films for Energy Storage 11. Prof. Hana Uršič Nemevšek
- Slovenian Research Agency
- 12. Environment-Friendly Ferroelectric Oxide Thin Films for Energy Harvesting and Energy Storage Applications Prof. Barbara Malič
 - Slovenian Research Agency
- 13. Realizing In-Situ Studies of Dynamic Mechanisms in Ceramic Oxides in the Reducing Environment in a Transmission Electron Microscope Prof. Andreja Benčan Golob
 - Slovenian Research Agency
- 14. Engineering the Microstructure and Performance of Lead-Free Piezoelectrics for Energy Harvesting
 - Prof. Barbara Malič
 - Slovenian Research Agency
- 15. Environment-Friendly Processing of Lead-Free Functional-Oxide Thin Films for Micro-Electro-Mechanical Systems (MEMS) Applications Prof. Barbara Malič
- Slovenian Research Agency
- Smart Design of New Multifunctional Composites with Optimized Energy Transfer 16. Across Interfaces between the Components (SoMwOT) Prof. Barbara Malič Slovenian Research Agency

RESEARCH PROGRAMME

Electronic Ceramics, Nano-, 2D and 3D Structures 1 Prof. Barbara Malič

VISITORS FROM ABROAD

- Maria Karypidou, Aristotle University of Thessaloniki, Thessaloniki, Greece, July 2-August 29, 2021
- Maximilian Gehringer, Technische Universität Darmstadt, Darmstadt, Germany, 2 September 1 - October 29, 2021
- 3. Reham Elsurhafa, University of Ankara, Ankara, Turkey, September 13-November 24, 2021

STAFF

Researchers

- Prof. Andreja Benčan Golob 1.
- 2. Dr. Mirela Dragomir
- Prof. Goran Dražić 3.
- Prof. Danjela Kuščer Hrovatin 4 Dr. Kostja Makarovič'
- 5. 6. Prof. Barbara Malič, Head
- Dr. Mojca Otoničar
- Prof. Tadei Rojac 8
- Prof. Hana Uršič Nemevšek 9
- Postdoctoral associates
- 10. Dr. Uroš Prah, on leave since 15.06.21
- 11. Dr. Kristian Radan, 01.02.21, transferred to Department K1
- Postgraduates
- 12. Matic Belak Vivod, B. Sc.
- 13. Oana Andreea Condurache, M. Sc.

- Prof. Brahim Dkhil, Centrale Supélec, Université Paris-Saclay, Paris, France, December 6. 19 - 21, 2019
- 16. Samir Salmanov, M. Sc. Matej Šadl, B. Sc. 17.

14. Sabi William Konsago, M. Sc.

18. Lia Šibav, B. Sc.

15. Barbara Repič, B. Sc.

- 19. Katarina Žiberna, B. Sc.
- Technical officers
- 20. Silvo Drnovšek, B. Sc. 21. Brigita Kmet, B. Sc.
- 22. Maja Koblar, B. Sc.
- Technical and administrative staff
- 23. Andrei Debevec
- 24. Tina Ručigaj Korošec, B. Sc.

Note: * part-time JSI member

Prof. Barbara Malič Advanced inorganic and organic thin films with enhanced electrically-induced response Prof. Barbara Malič

6 The quest for high-temperature superconductvity and exotic magnetism in fluoridoargentates(II) Asst. Prof. Mirela Dragomir

R & D GRANTS AND CONTRACTS

TCCbuilder: An open-source simulation tool for thermal control circuits

Electrocaloric elements for active cooling of electronic circuits

In situ atomic level Quantitative Scanning Transmission Electron Microscopy of

- Designing functionality of lead-free ferroelectrics through domain wall engineering Prof. Andreja Benčan Golob
- 8 The cool way to polarize Dr. Mojca Otoničar

Functional Materials

Prof. Barbara Malič

Multicaloric cooling

Prof. Andreja Benčan Golob

Prof. Hana Uršič Nemevšek

1

2.

3

4

5

- 9 Engineering of relaxor ferroelectric thin films for piezoelectric and energy storage applications
 - Prof. Tadej Rojac
- Structures of elusive noble-gas compounds elucidated by 3D electron diffraction Asst. Prof. Mirela Dragomir
- 11 All in One: Multi-caloric and Multi-scavenging Elements for Green Future Prof. Hana Uršič Nemevšek
- Enhanced piezoelectricity via structural disorder in polycrystalline relaxor ferroelectrics Prof. Tadej Rojac
- MIcrofluidic Sensor System for PESticides detection (MISS PES) 13. Prof. Danjela Kuščer Hrovatin
- Flexible elements with multi-physical properties Prof. Hana Uršič Nemevšek
- 15. Process intensification for the continuous synthesis of high purity hydrogen peroxide using a micro-scale electrocatalytic reactor Prof. Barbara Malič
- - 4. Matthieu Fricaudet, Universite Paris-Saclay, Paris, France, October 22 - 29, 2021
 - Maxime Vallet, Universite Paris-Saclay, Paris, France, November 25 December 4, 2021

BIBLIOGRAPHY

ORIGINAL ARTICLE

- 1. Luka Pavko *et al.* (19 authors), "Toward the continuous production of multigram quantities of highly uniform supported metallic nanoparticles and their application for synthesis of superior intermetallic Pt-alloy ORR electrocatalysts", *ACS applied energy materials*, 2021, **4**, 12, 13819–13829.
- 2. Klemen Bohinc, Karla Korade, Katarina Jerin, Nikolina Lešić, Marijana Đaković, Goran Dražić, Jean-François Dufrêche, Davor Kovačević, "Experimental and theoretical study of morphological and charging properties of truncated octahedron and cubic ceria nanoparticles: implications for biomedical applications", ACS applied nano materials, 2021, 4, 2, 1434-1444.
- 3. Matej Šadl *et al.* (12 authors), "Energy-storage-efficient $0.9Pb(Mg_{1/3}Nb_{2/3})O_3 0.1PbTiO_3$ thick films integrated directly onto stainless steel", *Acta materialia*, 2021, **221**, 117403.
- 4. Andraž Bradeško, Lovro Fulanović, Marko Vrabelj, Aleksander Matavž, Mojca Otoničar, Jurij Koruza, Barbara Malič, Tadej Rojac, "Multifunctional cantilevers as working elements in solid-state cooling devices", Actuators, 2021, **10**, 3, 58.
- 5. Neamul Hayet Khansur, Udo Eckstein, Hana Uršič, Matej Šadl, Martin Brehl, Alexander Martin, Kevin Riess, Dominique de Ligny, Kyle Grant Webber, "Enhanced electromechanical response and thermal stability of $0.93(Na_{1/2}Bi_{1/2})TiO_3 0.07BaTiO_3$ through aerosol deposition of base metal electrodes", *Advanced materials interfaces*, 2021, **8**, 11, 2100309.
- 6. Tony Schenk, Andreja Benčan, Goran Dražić, Oana Condurache, Nathalie Valle, Brahime El Adib, Naveen Aruchamy, Torsten Granzow, Emmanuel Defay, Sebastjan Glinšek, "Enhancement of ferroelectricity and orientation in solution-derived hafnia thin films through heterogeneous grain nucleation", *Applied physics letters*, 2021, **118**, 16, 162902.
- 7. Tadej Žumbar, Alenka Ristić, Goran Dražić, Hristina Lazarova, Janez Volavšek, Albin Pintar, Nataša Zabukovec Logar, Nataša Novak Tušar, "Influence of alumina precursor properties on Cu-Fe alumina supported catalysts for total toluene oxidation as a model volatile organic air pollutant", *Catalysts*, 2021, **11**, 2, 252.
- Aleksandar Miletić, Peter Panjan, Miha Čekada, Lazar Kovačević, Pal Terek, Janez Kovač, Goran Dražić, Branko Škorić, "Nanolayer CrAlN/TiSiN coating designed for tribological applications", *Ceramics international*, 2021, 47, 2, 2022-2033.
- 9. Vlad Alexandru Lukacs *et al.* (11 authors), "Preparation and properties of porous BaTiO₃ nanostructured ceramics produced from cuboidal nanocrystals", *Ceramics international*, 2021, **47**, 13, 18105-18115.
- Omeir Khalid, Alexander Spriewald Luciano, Goran Dražić, Herbert Over, "Mixed Ru_xIr_{1-x}O₂ supported on rutile TiO₂: catalytic methane combustion, a model study", *ChemCatChem*, 2021, **13**, 18, 3983-3994.
- 11. Stanislav Kurajica, Katarina Mužina, S. Keser, Goran Dražić, Ivana Katarina Munda, "Assessment of cell toxicity and oxidation catalytic activity of nanosized zinc-doped ceria UV filter", *Chemical and biochemical engineering quarterly*, 2021, 35, 2, 157-164.
- 12. Peter Panjan, Aljaž Drnovšek, Goran Dražić, "Influence of growth defects on the oxidation resistance of sputter-deposited TiAlN hard coatings", *Coatings*, 2021, **11**, 2, 123.
- 13. Hana Uršič, Marko Vrabelj, Mojca Otoničar, Lovro Fulanović, Brigita Rožič, Zdravko Kutnjak, Vid Bobnar, Barbara Malič, "Influence of synthesis-related microstructural features on the electrocaloric effect for $0.9Pb(Mg_{1/3}Nb_{2/3})O_3 0.1PbTiO_3$ ceramics", *Crystals*, 2021, **11**, 372.
- 14. Ruggero Vigliaturo, Sabrina M. Elkassas, Giancarlo Della Ventura, Günther Redhammer, Francisco Ruiz-Zepeda, Michael J. O'Shea, Goran Dražić, Reto Gieré, "Multi-scale characterization of glaucophane from Chiavolino (Biella, Italy): implications for international regulations on elongate mineral particles", *European journal of mineralogy*, 2021, **33**, 1, 77-112, .
- 15. Anica Peter, Leonard Mihaly Cozmuta, Camelia Nicula, Anca Mihaly Cozmuta, Cătălina Mihaela Talaşman, Goran Dražić, Antonio Peñas, Antonio Jesus Calahorro, Gianni Sagratini, Stefania Silvi, "Chemical and organoleptic changes of curd cheese stored in new and reused active packaging systems made of Ag-graphene-TiO₂-PLA", *Food chemistry*, 2021, **363**, 130341.

- 16. Nejc Rozman, Peter Nadrah, Renaud Cornut, Bruno Jousselme, Marjan Bele, Goran Dražić, Miran Gaberšček, Špela Kunej, Andrijana Sever Škapin, "TiO₂ photocatalyst with single and dual noble metal co-catalysts for efficient water splitting and organic compound removal", *International Journal of Hydrogen Energy*, 2021, **46**, 65, 32871-32881.
- Andreja Jelen, Primož Koželj, Darja Gačnik, Stanislav Vrtnik, Mitja Krnel, Goran Dražić, Magdalena Wencka, Zvonko Jagličić, Michael Feuerbacher, Janez Dolinšek, "Collective magnetism of a singlecrystalline nanocomposite FeCoCrMnAl high-entropy alloy", *Journal of alloys and compounds*, 2021, 864, 158115.
- Lukas Riemer, Li Jin, Hana Uršič, Mojca Otoničar, Tadej Rojac, Dragan Damjanović, "Dielectric and electro-mechanic nonlinearities in perovskite oxide ferroelectrics, relaxors, and relaxor ferroelectrics", *Journal of applied physics*, 2021, **129**, 5, 054101.
- Oana Condurache, Goran Dražić, Naonori Sakamoto, Tadej Rojac, Andreja Benčan, "Atomically resolved structure of step-like uncharged and charged domain walls in polycrystalline BiFeO₃", *Journal of applied physics*, 2021, **129**, 5, 054102.
- 20. Mihail Slabki, Lalita Kodumudi Venkataraman, Tadej Rojac, Jürgen Rödel, Jurij Koruza, "Thermal stability of the electromechanical properties in acceptor-doped and composite-hardened $(Na_{1/2}Bi_{1/2})TiO_3 BaTiO_3$ ferroelectrics", *Journal of applied physics*, 2021, **130**, 1, 014101.
- 21. Primož Koželj *et al.* (13 authors), "Spin-glass magnetism of the nonequiatomic CoCrFeMnNi high-entropy alloy", *Journal of Magnetism and Magnetic Materials*, 2021, **523**, 167579.
- 22. Andraž Bradeško *et al.* (12 authors), "Implications of acceptor doping in the polarization and electrocaloric response of 0.9Pb(Mg_{1/3}Nb_{2/3})0₃ - 0.1PbTiO₃ relaxor ferroelectric", *Journal of materials chemistry. C, Materials for optical and electronic devices*, 2021, 9, 3204-3214.
- Katarina Mužina, Stanislav Kurajica, Goran Dražić, Patrick Guggenberger, Gordana Matijašić, "True doping levels in hydrothermally derived copper-doped ceria", *Journal of nanoparticle research*, 2021, 23, 8, 149.
- 24. Anca Peter *et al.* (13 authors), "Modifying the silver-titania nanocomposites with carbonaceous materials to remove the pollutants from domestic waste water", *Journal of nanoscience and nanotechnology*, 2021, **21**, 4, 2278-2291.
- 25. Bing Ma, Matfas Blanco, Laura Calvillo, Lingjing Chen, Gui Chen, Tai-Chu Lau, Goran Dražić, Julien Bonin, Marc Robert, Gaetano Granozzi, "Hybridization of molecular and graphene materials for CO_2 photocatalytic reduction with selectivity control", *Journal of the American Chemical Society*, 2021, **143**, 22, 8414-8425.
- 26. Hermina Hudelja, Bernd Wicklein, Danjela Kuščer, Andraž Kocjan, "Triggering the aqueous interparticle association of $\gamma - Al_2O_3$ hierarchical assemblies using divalent cations and cellulose nanofibers", *Journal of the European ceramic society*, 2021, **41**, 1, 590-598.
- 27. Marion Höfling, Maximilian Trapp, Lukas Porz, Hana Uršič, Enrico Bruder, Hans-Joachim Kleebe, Jürgen Rödel, Jurij Koruza, "Large plastic deformability of bulk ferroelectric KNbO₃ single crystals", *Journal of the European ceramic society*, 2021, **41**, 7, 4098-4107.
- 28. Alfredo Blázquez Martínez, Nicolas Godard, Naveen Aruchamy, Cosme Milesi-Brault, Oana Condurache, Andreja Benčan, Sebastjan Glinšek, Torsten Granzow, "Solution-processed BiFeO₃ thin films with low leakage current", *Journal of the European ceramic society*, 2021, **41**, 13, 6449-6455.
- 29. Danjela Kuščer, Silvo Drnovšek, Franck Levassort, "Inkjet-printingderived lead-zirconate-titanate-based thick films for printed electronics", *Materials & design*, 2021, **198**, 109324.
- 30. Saide Umerova, Danjela Kuščer, Matej Bobnar, Nikita Derets, Boštjan Zalar, Andraž Rešetič, "Shear flow-controlled shape memory of polymer resin dispersed liquid crystal elastomer microparticles", *Materials & design*, 2021, 207, 109836.
- Anca Peter *et al.* (13 authors), "Morpho-structural and chemical characterization of paper based materials with functionalized surface", *Materials chemistry and physics*, 2021, 267, 124693.
- 32. Vesna V. Vodnik *et al.* (12 authors), "Development of genistein-loaded gold nanoparticles and their antitumor potential against prostate cancer cell lines", *Materials science & engineering. C, Materials for biological applications*, 2021, **124**, 112078.

- 33. Matej Šadl, Urban Tomc, Hana Uršič, "Investigating the feasibility of preparing metal-ceramic multi-layered composites using only the aerosol-deposition technique", *Materials*, 2021, 14, 16, 4548.
- 34. Brigita Kmet, Danjela Kuščer, Soma Dutta, Hana Uršič, Aleksander Matavž, Franck Levassort, Vid Bobnar, Barbara Malič, Andreja Benčan, "Screen printed copper and tantalum modified potassium sodium niobate thick films on platinized alumina substrates", *Materials*, 2021, 14, 23, 7137.
- Kostja Makarovič, Darko Belavič, Matjaž Vidmar, Barbara Malič, "A 3D LTCC-based ceramic microfluidic system with RF dielectric heating of liquids", *Materials*, 2021, 14, 23, 7396.
- 36. Kostja Makarovič, Darko Belavič, Barbara Malič, Andreja Benčan, Franci Kovač, Janez Holc, "Small ozone generator fabricated from low temperature Co-fired ceramics", *Microelectronics international*, 2021, 38, 1, 1-5.
- 37. Marija Tkalčević, Jordi Sancho-Parramon, Lovro Basioli, Matej Bubaš, Goran Dražić, Peter Nadazdy, Peter Siffalovica, Maja Mičetić, "3D networks of nanopores in alumina: structural and optical properties", *Microporous and mesoporous materials*, 2021, **325**, 111306.
- Zouhair Hanani *et al.* (12 authors), "Lead-free nanocomposite piezoelectric nanogenerator film for biomechanical energy harvesting", *Nano energy*, 2021, 81, 105661.
- 39. Edi Radin, Goran Štefanić, Goran Dražić, Ivan Marić, Tanja Jurkin, Anđela Pustak, Nikola Baran, Matea Raić, Marijan Gotić, "Solid-state dispersions of platinum in the SnO₂ and Fe₂O₃ nanomaterials", *Nanomaterials*, 2021, **11**, 12, 3349.
- 40. Jingkun Li *et al.* (16 authors), "Identification of durable and non-durable FeN_x sites in Fe-N-C materials for proton exchange membrane fuel cells", *Nature Catalysis*, 2021, 4, 1, 10-19.
- 41. Andreja Benčan, Emad Oveisi, Sina Hashemizadeh, Vignaswaran K. Veerapandiyan, Takuya Hoshina, Tadej Rojac, Marco Deluca, Goran Dražić, Dragan Damjanović, "Atomic scale symmetry and polar nanoclusters in the paraelectric phase of ferroelectric materials", *Nature communications*, 2021, **12**, 1, 3509.
- Mojca Otoničar, Andraž Bradeško, Samir Salmanov, Chingchang Chung, Jacob L. Jones, Tadej Rojac, "Effects of poling on the electrical and electromechanical response of PMN-PT relaxor ferroelectric ceramics", *Open ceramics*, 2021, 7, 100140.
- Anže Abram, Goran Dražić, "Structural and photocatalytic properties of hydrothermally-prepared boehmite/TiO₂ coatings", *Open ceramics*, 2021, 7, 100153.
- 44. Junning Li *et al.* (11 authors), "Doping-induced polar defects improve the electrocaloric performance of Ba_{0.9}Sr_{0.1}Hf_{0.1}Ti_{0.9}O₃", *Physical review applied*, 2021, **16**, 1, 014033.
- Mirela Dragomir, Iztok Arčon, Paul A. Dube, Jeremiah C. Beam, Andrew P. Grosvenor, Graham King, John E. Greedan, "Family of anisotropic spin glasses Ba_{1-x}La_{1+x}MnO₄₊₈", *Physical review materials*, 2021, 5, 7, 074403.
- 46. Qianli Ma, Kirrily C. Rule, Zachary W. Cronkwright, Mirela Dragomir, Gabrielle Mitchell, Evan M. Smith, Songxue Chi, Alexander I. Kolesnikov, Matthew B. Stone, Bruce D. Gaulin, "Parallel spin stripes and their coexistence with superconducting ground states at optimal and high doping in La_{1.6-x}Nd_{0.4}Sr_xCuO₄", *Physical review research*, 2021, **3**, 2, 023151.
- 47. Kevin Ries, Neamul Hayet Khansur, Alexander Martin, Andreja Benčan, Hana Uršič, Kyle Grant Webber, "Stress- and frequency-dependent properties of relaxor-like sodium bismuth titanate", *Physical review. B*, 2021, **103**, 9, 094113.
- 48. Tjaša Gornik, Sudhirkumar Shinde, Lea Lamovšek, Maja Koblar, Ester Heath, Börje Sellergren, Tina Kosjek, "Molecularly imprinted polymers for the removal of antidepressants from contaminated wastewater", *Polymers*, 2021, **13**, 1, 120.

- 49. Naman K. Gupta *et al.* (12 authors), "Vanishing nematic order beyond the pseudogap phase in overdoped cuprate superconductors", *Proceedings of the National Academy of Sciences of the United States of America*, 2021, **118**, 34, e2106881118.
- 50. Jamal Belhadi, Urška Trstenjak, Hana Uršič, Nina Daneu, Jieun Kim, Zishen Tian, Gertjan Koster, Lane W. Martin, Matjaž Spreitzer, "Growth mode and strain effect on relaxor ferroelectric domains in epitaxial 0.67Pb(Mg_{1/3}Nb_{2/3})O₃ – 0.33PbTiO₃/SrRuO₃ heterostructures", *RSC advances*, 2021, **11**, 3, 1222-1232.
- 51. Vesna Ribić, Aleksander Rečnik, Goran Dražić, Matejka Podlogar, Zorica Branković, Goran Branković, "TEM and DFT study of basal-plane inversion boundaries in SnO₂-doped ZnO", *Science of sintering*, 2021, 53, 2, 237-252.
- 52. Arij Marzouki, M. Yao, Samir Salmanov, Vincent Loyau, Adel Megriche, Brahim Dkhil, Mojca Otoničar, "New approach for designing bulk multiferroic composites made of two perovskite oxides with enhanced direct magnetoelectric coupling", *Scripta materialia*, 2021, **194**, 113673.
- 53. Kristina Radinović, Jadranka Milikić, Una Stamenović, Vesna V. Vodnik, Mojca Otoničar, Srečo D. Škapin, Biljana Šljukić Paunković, "Tailoring gold-conducting polymer nanocomposites for sensors applications: proof of concept for As(III) sensing in aqueous media", *Synthetic metals*, 2021, **278**, 116834.

REVIEW ARTICLE

 Shujun Zhang, Barbara Malič, Jing-Feng Li, Jürgen Rödel, "Lead-free ferroelectric materials: prospective applications", *Journal of materials research*, 2021, 36, 5, 985-995.

PUBLISHED CONFERENCE CONTRIBUTION

- Blaž Mikuž, Jan Kren, Anil Kumar Basavaraj, Danjela Kuščer, "Influence of seeding particles on particle image velocimetry measurements in single-phase turbulent pipe flow", In: NENE 2021, 30th International Conference Nuclear Energy for New Europe, September 6-9, Bled, Slovenia, Proceedings, Nuclear Society of Slovenia, 2021, 614.
- 2. Aleksander Sešek, Tadej Skuber, Kostja Makarovič, "Optimizacija izdelave močnostnih modulov ter termična analiza", In: ERK 2021, 30th International Electrotechnical and Computer Science Conference, Portorož, Slovenija, 20-21 September 2021, Proceedings, (Zbornik Elektrotehniške in računalniške konference 30), Slovenska sekcija IEEE, Fakulteta za elektrotehniko, 2021, 5-9.

INDEPENDENT COMPONENT PART OR A Chapter in a Monograph

- 1. Danjela Kuščer, "Screen printing", In: *Encyclopedia of materials: technical ceramics and glasses*, Elsevier, 2021, **1**, 227-232.
- 2. Barbara Malič, Mojca Otoničar, Kristian Radan, Jurij Koruza, "Lead-free piezoelectric ceramics", In: *Encyclopedia of materials: technical ceramics and glasses*, Elsevier, 2021, **3**, 358-368.

Patent

1. Vid Bobnar, Barbara Malič, Aleksander Matavž, *A method for producing polymeric surface modification layers*, S125887 (A), Urad RS za intelektualno lastnino, 31. 03. 2021.