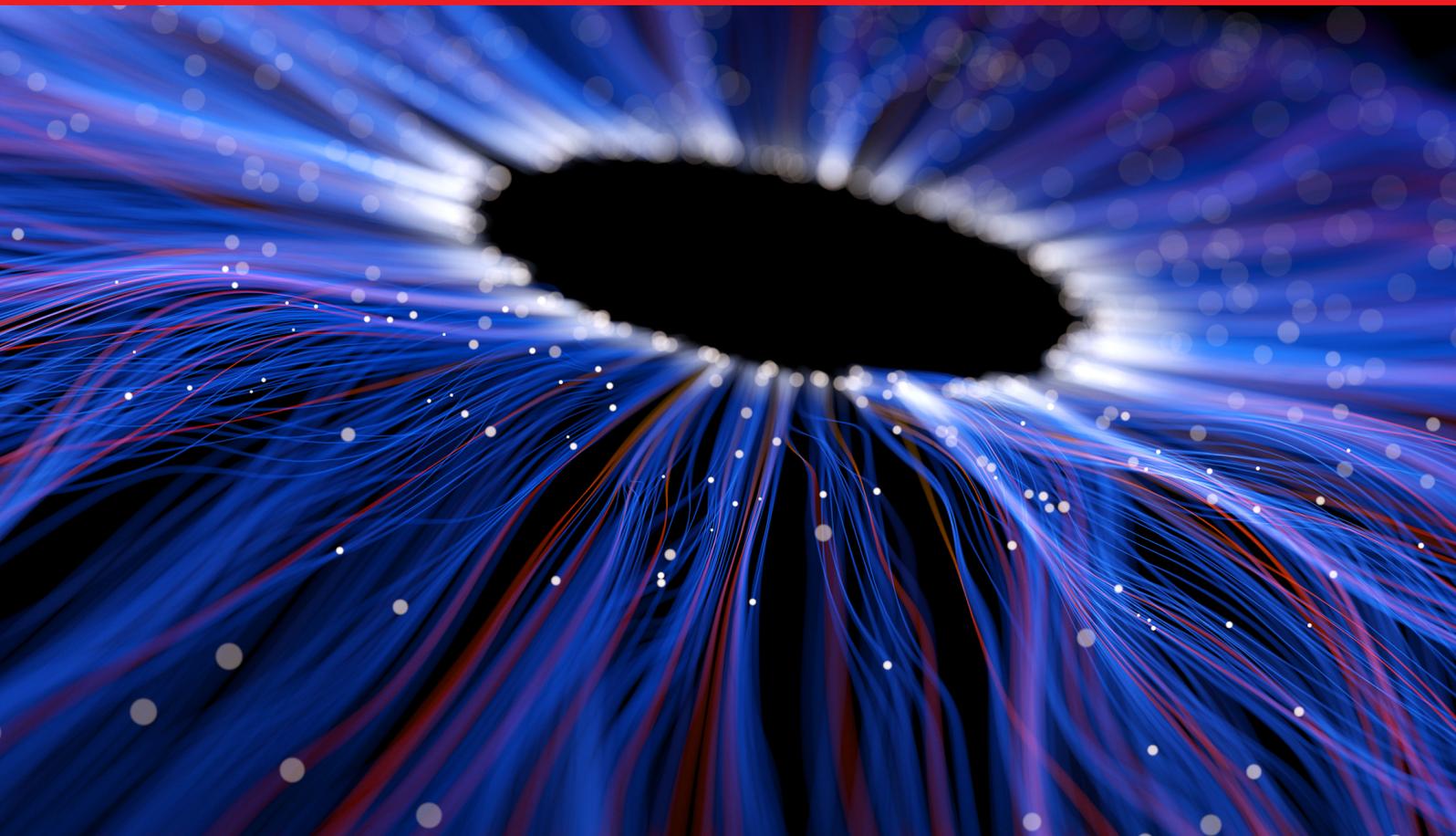


Abstract booklet



Dielectrics 2022

27-29 April 2022

<http://dielectrics2022.iopconfs.org>

Programme

Wednesday 27 April

- 12:30 Arrival, registration and lunch
- 13:30 Welcome
- Session 1 - Focus Topic 1 (Theory and Modelling of Dielectric Materials)**
- 13:40 **(Plenary) Modelling of Spin-Orbit Coupling Effects in Ferroelectrics**
Silvia Picozzi, CNR-SPIN, Italy
- 14:30 **Manipulation of point-defect spin orientation via ferroelectric switching from first principles**
Katherine Inzani, University of Nottingham, UK
- 14:50 **Oxygen vacancy defect of crystalline (α , β , and ϵ)-Ga₂O₃**
Chaiyawat Kaewmeechai, UCL, UK
- 15:10 Coffee break
- Session 2, Focus Topic 1 (Theory and Modelling of Dielectric Materials)**
- 15:30 **(Invited) Designing topology in oxides**
Sinéad Griffin, Lawrence Berkeley National Laboratory, USA
- 16:00 **An electron-injection based model of degradation in amorphous HfO₂**
Jack Strand, UCL, UK
- 16:20 **First principles DFT+U+J modelling of bandgaps and defect levels in TiO₂, ZrO₂ and HfO₂**
Daniel Lambert, Trinity College Dublin, Ireland
- 16:40 **Anharmonicity and entropic stabilisation in zirconia from first principles**
Kasper Tolborg, Imperial College London, UK
- 17:00 Drinks reception

Thursday 28 April

- 08:30 Arrival, registration and refreshments
- Session 3 - Focus Topic 2 (Advanced Characterisation of Dielectric Materials)**
- 09:00 **(Plenary) Space Charge Characteristics in Nanodielectrics**
George Chen, University of Southampton, UK
- 09:50 **(Invited) In operando x-ray spectroscopy of interfaces and devices**
Prof. Kai Rosnagel, University of Kiel, Germany
- 10:20 **What spins below the surface: HAXPES view of mixed valence manganites**
Tommaso Pincelli, Fritz Haber Institute, Germany
- 10:40 **Relaxor ferroelectricity in an average cubic (Pm-3m) phase of a lead-free smart material**
Digvijay Nath Dubey, Indian Institute of Technology (BHU), India

11:00 **Domain wall free enhanced photocatalytic reactions in nanoscale ferroelectric $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$**
Steve Dunn, LSBU, UK

11:20 Coffee break

Session 4 - Focus Topic 1 (Theory and Modelling of Dielectric Materials)

11:40 **(Mansell Davies Award Talk) Modelling dielectric constants with an explainable machine learning model**

Kazuki Morita, Imperial College London, UK

12:10 **Understanding electronic transport in semiconductors using high-throughput calculations and machine learning**

Alex Ganose, Imperial College London, UK

12:30 **Unravelling the impact of disorder on the polarizability and electronic properties of mixed-metal chalcogenides**

Adair Nicolson, UCL, UK

12:50 Lunch

Session 5 - Focus Topic 3 (Dielectrics for Biological Applications)

13:50 **(Plenary) Dielectrics for Biomedical Applications – putting their functionality to use *in-vivo***

Julia Glaum, Norwegian University of Science and Technology, Norway

14:40 **(Invited) Ferroelectric materials and scaffolds for tissue regeneration and implantable devices**

Dr. Hamideh Khanbareh, University of Bath, UK

15:10 **An Evolving Theory for Protein Dielectrophoresis**

Ronald Pethig, The University of Edinburgh, UK

15:30 Coffee break

Session 6 - Focus Topic 1 (Theory and Modelling of Dielectric Materials)

15:50 **A new take on Electrostriction**

Pierre-Eymeric Janolin, Université Paris-Saclay, France

16:10 **A New Approach to an Old Idea: Understanding the Fröhlich $B(T)$ Function**

Paul Ben Ishai, Ariel University

16:30 **A theory for like-charge attraction of polarizable ions**

Ho-Kei Chan, Harbin Institute of Technology, China

16:50 **Finite Size Effects in the Two-Dimensional XY Model of Ferromagnetism**

Yoonsang Kim, North London Collegiate School Jeju, South Korea

17:10 Finish

19:00 Dinner

Friday 29 April

08:30 Arrival, registration and refreshments

Session 7 - Focus Topic 4 (Dielectrics for Electronic Devices)

09:00 **(Plenary) Deposition of Metal Oxide Thin Films for Large-Area Electronics**

Andrew Flewitt, University of Cambridge, UK

09:50 **Approaching metal oxide dielectrics and semiconductors by solution and gas phase processing of molecular precursors**

Joerg Schneider, Technische Universitaet Darmstadt, Germany

10:10 **Study of compositional and thermal dependent negative permittivity of Sr₂Mn_{1-x}Nb_xO₄ (x = 0.1, 0.2, 0.3) solid solutions**

Gurudeo Nirala, Indian Institute of Technology (BHU), India

10:50 **Heteroepitaxially grown diamond for application in electronic devices**

Juergen Weippert, Fraunhofer IAF, Germany

11:10 Coffee break

Session 8 - Focus Topic 4 (Dielectrics for Electronic Devices)

11:30 **(Invited) Composite Design and Modelling for Novel Energy Harvesting and Sensing Structures**

Chris Bowen, University of Bath, UK

12:00 **Light-assisted ferroelectric domain switching for controlled modulation of current**

Gaurav Vats, Katholieke Universiteit Leuven, Belgium

12:20 **A detailed Study on Bulk Photovoltaic Effect in Ferroelectric System**

Subhajit Pal, QMUL, UK

12:40 **Solution processable high - k polymer nanocomposite for low cost energy storage device applications**

Suman Mandal, UCL, UK

13:00 Closing remarks

13:10 Lunch and departure

Wednesday 27 April 2022

Session 1 - Focus Topic 1 (Theory and Modelling of Dielectric Materials)

(Plenary) Modelling of Spin-Orbit Coupling Effects in Ferroelectrics

Silvia Picozzi

Consiglio Nazionale delle Ricerche CNR-SPIN, Italy

During the last couple of decades, the spin-orbit interaction has played an increasingly crucial role in condensed matter physics, thanks to its relevance as a rich microscopic mechanism from the fundamental point of view and as a driving force for innovative spintronic applications on the technological side. After a general overview on spin-orbit coupling (SOC), I will discuss two non-trivial aspects where this relativistic interaction gives rise to novel and exotic phenomena in ferroelectric materials. First, I will focus on the ab-initio modelling of (non-magnetic) ferroelectric semiconductors, where SOC leads to a tight link between Rashba spin-splitting, spin-texture and electric polarization, with the appealing perspective of electric-field control of spin-degrees of freedom and long-sought integration of spintronics with ferroelectricity. Second, I will discuss first-principles results for two-dimensional magnets, where SOC, mostly arising not from the 3d transition metal but rather from anionic p-orbitals, can play a relevant role in the emergence of multiferroicity and magnetoelectricity.

The author acknowledges the Italian Ministry of Research (MUR) for support to the project "TWEET: Towards Ferroelectricity in two dimensions", Grant n. 2017YCTB59

Manipulation of point-defect spin orientation via ferroelectric switching from first principles

Katherine Inzani

University of Nottingham, UK

A key advantage of spin-defect based quantum technologies is the potential to engineer the defect properties by modifying the atomic environment. Systems which exhibit coupling between magnetic and electric degrees of freedom provide a route to manipulate spins by applying an electric-field, suggesting the prospect of localized spin-control via electrostatic gating.

We have elucidated the fundamental limit of magnetoelectric coupling by electric-field control of magnetic dopants in ferroelectric hosts, demonstrating through combined first principles calculations and electron-paramagnetic resonance measurements that the spin directionality evolves following a switching path coupled to polarization switching.¹

We further leverage the versatility of the ferroelectric oxide platform by modifying the crystal field environment of the defect through epitaxial strain, low symmetry hosts and emergent topological polarization textures.² Results demonstrating spin-control in these systems provide an enhanced understanding of spin-charge coupling in defect systems and offer novel routes to tailored spin-control in ferroelectric crystals.

- [1] J. Liu, V. V. Laguta, K. Inzani, W. Huang, S. Das, R. Chatterjee, E. Sheridan, S. M. Griffin, A. Ardavan, R. Ramesh, Coherent electric field manipulation of Fe³⁺ spins in PbTiO₃. *Science Advances*, **7**, eabf8103 (2021).
- [2] K. Inzani, N. Pokhrel, N. Leclerc, Z. Clemens, S. P. Ramkumar, S. M. Griffin, E. A. Nowadnick, Manipulation of spin orientation via ferroelectric switching in Fe-doped Bi₂WO₆ from first principles. *Physical Review B*, **105**(5), 054434 (2022).

Oxygen vacancy defect of crystalline (α , β , and ϵ)- Ga_2O_3

Chaiyawat Kaewmeechai, and Alexander L. Shluger

University College London, UK

Ga_2O_3 is a transparent semiconductor possessing ultrawide bandgap (5 eV), a high breakdown electric field and a large Baliga's figure of merit (BFOM), which is a potential candidate for the power electronics. The three most stable phases of Ga_2O_3 are β , ϵ , and α respectively, which contain different portions in octahedron and tetrahedron. In this work, we investigate geometry optimizations and charge transition levels (CTL) of oxygen vacancy defect in these Ga_2O_3 phases using the density functional theory (DFT) with optimized PBE0-TC-LRC nonlocal functional satisfying the Koopmans' theorem. We found that the CTL change from 0 to +2 in Ga_2O_3 . Then, tetrahedron Ga atoms move toward the vacancy with the displacement ~ 0.3 Å for the 0-charge state but it moves outward the vacancy by around ~ 0.9 Å for +2 charge state respect to the perfect atomic position. While the α -phase that contains only octahedrons, two Ga atoms with loss the shorter bonds move toward and opposite the vacancy with the displacement ~ 0.3 Å for 0 and +2 charge state respectively. We discussed that the very large displacement is observed in tetrahedron Ga atoms especially +2 charge state due to weaker bond and lower density in tetrahedron-contained Ga_2O_3 (β and ϵ). Possessing tetrahedron in Ga_2O_3 also decreases density and electronic band gap of Ga_2O_3 significantly.

Session 2 - Focus Topic 1 (Theory and Modelling of Dielectric Materials)

(Invited) Designing topology in oxides

Sinéad Griffin

Lawrence Berkeley National Laboratory, USA

Topology has emerged as both a framework for understanding the fundamental physics of materials, and as a route to achieving sought-after properties such as giant magnetoresistance and chiral anomalies. In fact, connections between the origins of the universe and phase transitions in solid-state materials have been made by understanding the physics of topological defects that are common to both. With the ever-increasing litany of materials with topological phases, the next goal is to provide design routes for their control with experimentally accessible parameters. Oxides in particular are exciting for this on-demand design of topology since they already host highly-tunable orders such as ferroelectricity, ferromagnetic and superconductivity. In this talk, I will discuss how the conventional order parameters in oxides such as ferroelectric mode distortions and octahedral rotations can be used to tune topological order. I will provide two case studies looking at higher-order topological phases in perovskite oxides, and the interplay between real and reciprocal space topology in hexagonal manganites using first principles calculations and phenomenology theory. Finally, I will discuss the prospects of these materials as hosts for Majorana fermions and their potential as robust elements in topological quantum computers and as new classical information carriers.

An electron-injection based model of degradation in amorphous HfO₂

Jack Strand¹, Paolo La Torraca², Andrea Padovani³, Luca Larcher³, and Alexander Shluger¹

¹UCL, UK, ²University of Modena and Reggio Emilia, Italy, ³Applied Materials Inc, USA

We use a multi-scale modelling approach to study the time-dependent dielectric breakdown (TDDB) of amorphous (a-) HfO₂ based metal-oxide-metal (MOM) capacitors. In our proposed breakdown model, two types of defect are important: Firstly, the intrinsic bi-electron trap, which we have previously predicted to exist in a-HfO₂ [1]. Secondly, the oxygen vacancy, which is the primary defect responsible for degradation. Electron injection into bi-electron traps significantly reduces the activation energy for generation of oxygen vacancies [2]. Therefore, electron injection causes the number of vacancies to grow over time, eventually forming the percolation path responsible for dielectric breakdown. The majority component of electron transport through the dielectric occurs by multi-phonon trap assisted tunnelling (MPTAT) between O vacancies [3]. Energy parameters characterising the creation of oxygen vacancies and the MPTAT process are calculated using Density Functional Theory (DFT) employing the PBE0-TC-LRC hybrid density functional[4]. These parameters are then used to implement a device-level simulation of breakdown using the Ginestra® software[5].

The results demonstrate that the formation of O vacancies facilitated by electron injection into the oxide is a viable model for degradation in amorphous HfO₂. We further show the importance of the so-called "energetic correlation" effect where O vacancies locally increase the generation rate of additional vacancies, accelerating the oxide degradation process. This model gives realistic breakdown times and Weibull slope parameters, and provides a detailed insight into the mechanism of dielectric breakdown. It offers a new understanding of degradation mechanisms in oxides used in current MOSFET technology and can be useful for developing future resistive switching and neuromorphic nanodevices.

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First principles DFT+U+J modelling of bandgaps and defect levels in TiO₂, ZrO₂ and HfO₂

Daniel Lambert, and David O'Regan

Trinity College Dublin, Ireland

Density functional theory (DFT) modelling of dielectric materials using semi-local functionals is fast, but inaccurate for bandgaps when compared to computationally expensive hybrid functionals. Hubbard U corrections are often used to reduce bandgap inaccuracy at low cost, but the magnitude of these corrections is often chosen on an empirical basis, or turns out to be excessively large when calculated in situ. DFT+U+J [1] is a technique for incorporating explicit exchange interaction corrections within the DFT+U framework, and operated via first principles it has produced accurate bandgaps for several materials. It is yet untested for a wider range of calculations such as defect formation energies.

In our research we have first benchmarked the performance of DFT+U+J against alternative methodologies such as hybrid functionals, observing their effect on bandgap accuracy as well as effective mass, bond lengths and unit cell volume [1]. We then modelled the effect of oxygen vacancies in TiO₂, ZrO₂, and HfO₂ supercells, with and without DFT+U+J corrections, examining transition levels and defect formation energies.

We find that in the tested materials, the use of DFT+U+J can give band gap values that are close to experimental values, outperforming PBE, standard DFT+U, and in some cases even hybrid functionals. We demonstrate that, for selected materials, the DFT+U+J bandgap accuracy persists in oxygen vacancy simulations, and that defect transition levels are in line with previous hybrid-functional results. This opens a route to first-principles, fast computation of defects in large supercells, without sacrificing bandgap accuracy.

This publication has emanated from research supported in part by a research grant from Science Foundation Ireland (SFI) and is co-funded under the European Regional Development Fund under Prime Award Number 12/RC/2278_P2.

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Anharmonicity and entropic stabilisation in zirconia from first principles

Kasper Tolborg, and Aron Walsh

Imperial College London, UK

Stability of high temperature phases is of continuous importance in solid state chemistry with many materials exhibiting exciting properties in their high temperature polymorphs. Currently, the discovery of high temperature phases is largely serendipitous due to a lack of computational methods to predict stabilities at high temperature.

In this contribution, we investigate the high temperature stability of zirconia, which is one of the most studied oxide ceramics with several interesting properties in both pure and doped forms, including high hardness, high dielectric constant, ionic conductivity, and low thermal conductivity. At ambient conditions, zirconia is stable in its monoclinic polymorph, which transforms to tetragonal and cubic polymorphs at high temperatures [1].

For cubic zirconia, conventional harmonic phonon theory breaks down, and we must therefore apply anharmonic phonon theory to understand its high temperature properties [2]. Thus, applying the framework of self-consistent phonon theory including quasi-particle corrections [3,4], we show how the soft mode cubic-to-tetragonal transition develops. Within this framework, we also predict free energies of cubic and tetragonal phases, but this shows that the current level of theory is unable to predict the reverse transition upon heating.

To complete the picture, we apply ab initio molecular dynamics simulations at high temperature to show that even with inclusion of higher order anharmonicities, the tetragonal phase appears to remain thermodynamically stable. Instead, we show that the cubic polymorph is likely to be stabilised due to spontaneous defect formation, which is also the driving force for the well-known ionic conductivity at high temperatures.

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Thursday 28 April 2022

Session 3 - Focus Topic 2 (Advanced Characterisation of Dielectric Materials)

(Plenary) Space Charge Characteristics in Nanodielectrics

George Chen

University of Southampton, UK

Nanodielectrics, as a new branch of dielectrics, have attracted significant attention in last two decades as they have shown some improved dielectric properties that are important for high voltage insulation and energy storage applications. One of these improvements is the suppression of charge injection when nanodielectrics are subjected to higher electric fields. Several explanations have been proposed including the formation of deep traps after introducing nanoparticles in the material. However, mechanisms that are responsible for the charge suppression are not detailed. More importantly, the effect of charge suppression is strongly dependent on the amount of nanoparticles, i.e. loading concentration. In the present paper, a schematic model has been proposed based on deep trap concept. The results from two complimentary experiments have been presented to support the model. Further, a tunneling process has been introduced when the trapping sites become closer which is the case for high nanofillers concentration. Based on the new model, charge formation and dynamics in nanodielectrics with different loading concentrations can be better understood and electrical performance explained.

(Invited) In operando x-ray spectroscopy of interfaces and devices

Kai Rossnagel

Kiel University / DESY, Germany

Device functionality arises from electron behavior at interfaces. A thorough bottom-up understanding of this behavior is essential not only to continued miniaturization and sustained performance, but also to the exploration of new emergent materials, devices, or forms of electronics. Synchrotron- and free-electron-laser-based x-ray spectroscopies are uniquely powerful tools to provide such understanding as they can image and deconstruct interfacial electronic structure and dynamics at their fundamental dimensions.

Here, we will highlight recent developments in the application of x-ray spectroscopies to interfaces under operating and far-from-equilibrium conditions. The spectroscopic probes include near-edge x-ray absorption fine structure (NEXAFS) spectroscopy, hard x-ray photoelectron spectroscopy (HAXPES), and angle-resolved photoelectron spectroscopy (ARPES), in modalities with high spectral (meV), spatial (nm to μm), or temporal (fs) resolution. The investigated materials range from oxides (SiO_2 , PZT, PMN-PT, VO_2) to layered transition-metal dichalcogenides (MoS_2 , TiSe_2 , TaS_2).

Two different *in operando* approaches will be demonstrated. First, electrical gating and current flow in device-like structures will be used to probe barrier heights at dielectric and ferroelectric interfaces, drive a strain-induced insulator-to-metal transition in VO_2 , and tune electronic structures in quasi-2D materials. Second, we will show how stroboscopic optical pumping can provide direct and incredibly detailed dynamical information on ultrafast interfacial charge transfer and concurrent responses of electronic and atomic structures at a molecule-quasi-2D material interface. The results presented open up exciting perspectives for future *in operando* x-ray spectroscopies at diffraction-limited storage rings and high-repetition-rate x-ray free-electron lasers.

What spins below the surface: HAXPES view of mixed valence manganites

Tommaso Pincelli¹, Gian Marco Pierantozzi², Aleksandr Yu. Petrov², Francesco Offi³, Francesco Borgatti⁴, Giancarlo Panaccione²

¹Fritz Haber Institut, Germany, ²Istituto Officina dei Materiali - CNR, Italy, ³Dipartimento di Scienze - Università Roma Tre, Italy, ⁴Istituto per lo Studio dei Materiali Nanostrutturati (ISMN) - CNR, Italy

Hole-doped rare-earth manganites display peculiar phenomena originating from the competition between charge, spin, and orbital order parameters [1]. The key functionality arises from the d-states of Mn that form narrow, crystal-field split bands where double exchange hybridization and Jahn-Teller correlation mechanisms compete to create, respectively, magnetic-metal and polaronic-insulator states. This makes manganites highly responsive to external stimuli such as electric, magnetic and strain fields, a behaviour highly sought-after in the new generation of functional materials. Manganites in the magnetic metallic phase also possess a very high degree of spin polarization of the Fermi surface (half-metallicity), which make them promising candidates for spintronic applications.

We present here the insight provided by advanced characterization based on hard X-ray photoemission [2], which allows peering below the surface into the electronic structure. We first focus on $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$, the widest bandwidth manganite. Employing multiple photon energies, we are able to evaluate the critical thickness of electronic hybridization, finding a significant evolution up to 10 unit cells from the surface [3]. With the same approach, we explore the structure of the valence band, identifying elemental and orbital contributions previously neglected [4].

We then extend the characterization in the time domain. This allows us to disentangle energetically close mechanisms by assessing their timescales. In LSMO, we identify the slow collapse of magnetization, a signature of half-metallicity, and we link it to the evolution of delocalized electronic states. We show that the double-exchange interaction remains active for several hundreds of picoseconds, suggesting a slow timescale evolution of electronic correlation [5]. Polaronic insulating states can be observed in the narrower bandwidth compound $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$ under biaxial strain conditions [6], which offer a fascinating phase diagram: we characterize the electronic structure [7] and observe a hidden phase accessible via ultrafast optical excitation.

Note: the works described here are the result of a large international collaboration whose participants will be acknowledged to full extent in the oral presentation; the author list (limited by space constraints) reports only the key contributors.

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Relaxor ferroelectricity in an average cubic (Pm-3m) phase of a lead-free smart material

Digvijay Nath Dubey¹, Gurvinderjit Singh², and Saurabh Tripathi³

¹Indian Institute of Technology (BHU) , India, ²Laser and Functional Materials Division, Raja Ramanna Centre for Advanced Technology, India, ³Department of Physics, Indian Institute of Technology (BHU), India

Relaxor ferroelectrics (or relaxors) are the class of materials which are characterized by a strongly broadened frequency dependent dielectric permittivity peak, and exhibit the ability to maintain the thermally stable high dielectric and electromechanical properties over a wide range of temperature. Owing to such fascinating characteristics, relaxors find wide applications in the field of actuators, sensors, capacitors etc [1]. However, the dominance in the field of relaxors is still of lead-based ceramics, which are toxic in nature [2]. Therefore, the need for the replacement of lead-based ceramics has intensified the search of lead-free alternatives in recent years [3]. In this regard we have developed a perovskite (ABO₃) based ‘A’ site tunable lead-free relaxor ferroelectric system *viz.*, (Ba_{1-x}Ca_x)Sn_{0.11}Zr_{0.05}Ti_{0.84}O₃; (0 ≤ x ≤ 0.20) exhibiting an average cubic (Pm-3m) structure at room temperature as inferred from x-ray diffraction studies [4]. Despite having cubic phase, the so developed smart materials have shown a slim hysteresis loop when subjected to PE loop measurements. Such a contrapositive behaviour is found to be owed by the presence of locally off-centered atoms corresponding to ‘A’ and ‘B’ site of the perovskite structure. Further, our analysis have revealed a cooperative behaviour between the dipoles driven by the ‘A’ and ‘B’ site atoms, which has been confirmed from Raman spectroscopic measurements. The short-range correlations among the off-centered displacements of the cations corresponding to ‘A’ and ‘B’ sites, and hence the number, and size of polar nano regions (PNRs) have shown an increasing trend as a function of Ca²⁺(x), which has been clearly revealed by the enhancement of ferroelectric polarization and the dielectric relaxation behaviour of the developed ceramics. The increase in strength of the PNR's via Ca²⁺(x) content has been found to be analogous to the decreasing temperature behaviour of relaxor ferroelectrics. The presence of a slim hysteresis loop along with broad and diffuse dielectric nature has made the ceramic a potential candidate for various energy storage applications [4].

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Domain wall free enhanced photocatalytic reactions in nanoscale ferroelectric Ba_xSr_{1-x}TiO₃

Steve Dunn¹, Yaqiong Wang², Man Zhang³, Jianguo Liu⁴, Haibin Zhang⁵, and Haixue Yan³

¹London South Bank University, UK, ²Xi'an Jiaotong University, China, ³Queen Mary University of London, UK, ⁴Tsinghua University, China, ⁵China Academy of Engineering Physics, China

Ferroelectric materials exhibit anomalous behaviour due to the presence of domains and domain walls which are related to the spontaneous polarisation inherent in the crystal structure. Control of ferroelectric domains and domain walls has been used to enhance the performance of devices such as ultrasound, pyroelectric detectors and photovoltaic systems with renewed interest in nanostructuring ferroelectric materials for energy applications. The application of ferroelectric materials in photoactive applications has gained interest due to the discovery of inherent polarisation in organic metal halide perovskite photosensitisers. Inherent properties associated with a ferroelectric can double a photocatalytic reaction rate and sustain a carrier lifetime of 100's of ms. However, there remains a lack of understanding on the contributions from domain and domain on enhanced photoactivity. In this work we compare samples of nanostructured Ba_xSr_{1-x}TiO₃ with and without a polar domain structure. Our study reveals that the observed enhanced photoactivity of perovskite ferroelectric materials stems from the inherent polarisation (and associated field) of the domain and not the presence of domain walls. This new understanding of the

underlying physics of materials with a spontaneous dipole opens a door to enhance the performance of light induced energy harvesting systems.

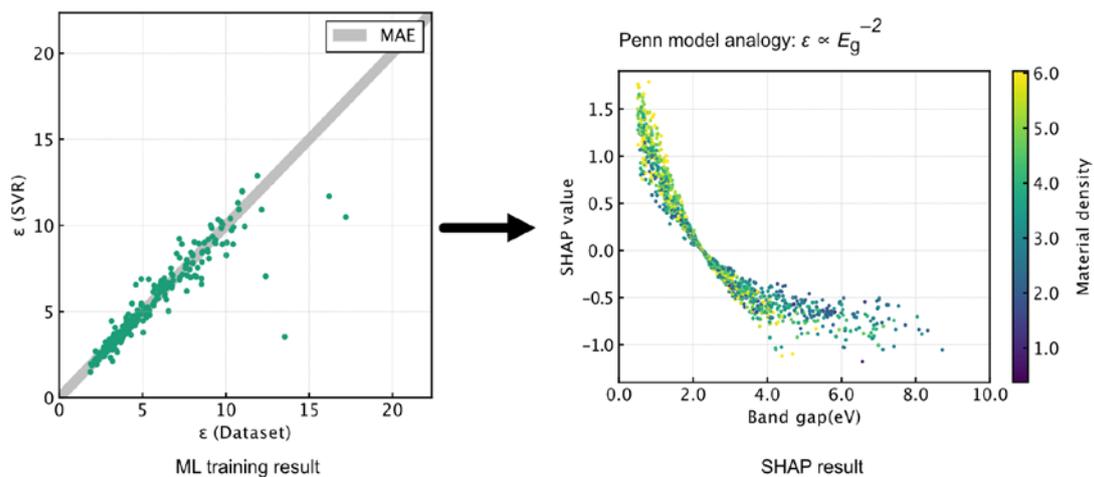
Session 4 - Focus Topic 1 (Theory and Modelling of Dielectric Materials)

(Mansell Davies Award Talk) Modelling dielectric constants with an explainable machine learning model

Kazuki Morita¹, Daniel Davies¹, Keith Butler², and Aron Walsh¹

¹Imperial College London, UK, ²Rutherford Appleton Laboratory & University of Reading, UK

In the past decade, machine learning (ML) has emerged as a powerful tool in materials science, however, their predictions are often difficult to interpret, and their reliability was repeatedly questioned. In this study, we train a ML model on a dataset of dielectric constants and use game theory to derive the reasoning behind the prediction. The Shapley Additive Explanation (SHAP) analysis used in this work allows one to obtain the quantitative contribution of each features towards the final ML prediction. Using this method, we show that the ML model is capturing trends similar to that found in conventional theoretical models, such as Penn model, but with more generality. Furthermore, through close comparison between our density functional perturbation theory and the data set, we were able to distinguish between dataset error and ML error, which are often difficult to disentangle. We report that errors could originate from an unusual property of materials or under-converged calculation settings.



Understanding electronic transport in semiconductors using high-throughput calculations and machine learning

Alex Ganose

Imperial College London, UK

The temperature dependence of experimental mobility is commonly used as a predictor of the dominant scattering mechanism in thermoelectric materials. However, if the scattering mechanism is determined incorrectly, this can lead to wildly inaccurate predictions of materials properties, frustrating efforts to optimise devices. In this work, I use a combination of high-throughput workflows and machine learned materials properties to generate a dataset of 24,000 mobility calculations. Based on this dataset, I demonstrate that the temperature-dependence of mobility is not a reliable indicator of the dominant scattering mechanism and instead reveal that many materials long considered to be dominated by deformation-potential scattering are instead controlled by polar optical phonons. This work highlights the potential for data driven approaches to provide insights for materials discovery and optimisation.

Unravelling the impact of disorder on the polarizability and electronic properties of mixed-metal chalcogenides

Adair Nicolson, Seán Kavanagh, and David Scanlon

University College London, UK

Mixed-metal mixed-anion systems have seen a significant rise in interest as ‘perovskite-inspired materials’, with desirable dielectric properties such as strong polarization and optical absorption. They are also expected to combine the excellent stability seen in metal chalcogenide solar cells with the well-known performance of hybrid halide perovskite solar cells.^[1]

$\text{Sn}_2\text{SbS}_2\text{I}_3$ is a promising solution-processed photovoltaic absorber having achieved efficiency above 4% in initial devices.^[2] Theoretical work predicts that the material family of $\text{A}_2\text{BCh}_2\text{X}_3$ mixed-metal chalcogenides could also be ferroelectric, with $\text{Sn}_2\text{SbS}_2\text{I}_3$ having strong lattice polarization and large dielectric constants.^[3]

However, this family has not been rigorously explored resulting in confusion in the literature regarding the structure of these materials, with some works observing disorder in room temperature crystals.^[4] Without a proper description of the structure, prediction of the electronic properties cannot be accurately performed. Understanding the extent of the disorder in these systems is of key importance due to its tendency to quench favourable properties such as macroscopic polarisation.

Using Density Functional Theory, Cluster expansion and Monte Carlo techniques, we have systematically examined the cation disorder in $\text{Sn}_2\text{SbS}_2\text{I}_3$ for the first time and will discuss its likely impact on the potential for this material family to produce ferroelectric and photovoltaic devices.

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Session 5 - Focus Topic 3 (Dielectrics for Biological Applications)

(Plenary) Dielectrics for Biomedical Applications – putting their functionality to use *in-vivo*

Julia Glaum

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Dielectric materials are at the heart of many biomedical applications and devices that improve patients' lives on an everyday basis. They enable diagnostic and surgical tools, mimic body functions and serve for therapeutic purposes. While in some applications dielectric materials are mainly chosen due to their structural and mechanical properties (e.g. in total joint replacements), in other cases their functional characteristics are directly utilized (e.g. in ultrasound devices). The advancements in miniaturization of electronics allow the transfer of established transducer technologies to *in vivo* applications. This enables a wide range of implantable sensors based on dielectric materials that help to monitor various processes inside the body. Many body functions are as well based on transduction-type processes, with the body constantly responding to a variety of stimuli (mechanical, chemical, acoustic,...). Electric signals are transferred into muscle contraction and body motion, optical and acoustic signals become electric signals that are interpreted by the brain and help us orient ourselves in the world. Mimicking such tissue responses using dielectric implant materials is a rapidly growing research field that opens up a wide range of novel *in vivo* applications. In that spirit, this presentation will focus on application areas for dielectric materials that aim to either replace the functionality of damaged or dysfunctional tissue, or that provide therapeutic stimuli *in vivo*.

(Invited) Ferroelectric materials and scaffolds for tissue regeneration and implantable devices

Hamideh Khanbareh, Vlad Jarkov, and Zois Michail Tsikriteas

University of Bath, UK

Ageing, disease and injury can cause permanent tissue damage, particularly in the case of organs which have limited healing capabilities, such as the central nervous system (CNS) and cartilage. Electrical stimulation can enhance cell regeneration by altering the cellular response to stimulate repair. This has been shown on conductive materials, powered by batteries. However, once the batteries have exceeded their lifetime, they require surgical replacement, exposing patients to health risks. The challenge for the long-term use of such devices is a reliable source that generates electricity from human-activity-induced thermal and vibrational energy.

Ferroelectric materials possess an inherent polarisation, thus surface charge that varies upon a change in pressure and temperature, through piezoelectricity and pyroelectricity, respectively. Therefore, they can interact with cells through electrical signals to stimulate repair. However, common ferroelectric materials such as lead zirconate titanate (PZT) and polyvinylidene-difluoride (PVDF) contain toxic or non-biodegradable components and are thus not favourable for implantation. In this work we show successful fabrication and characterisation of biocompatible ferroelectric polymer composites in the form of thin membranes and 3D porous scaffolds.

Thin ferroelectric membranes of Polydimethylsiloxane (PDMS)-potassium sodium lithium niobate (KLN) have been prepared via in-situ poling-dielectrophoresis to obtain high piezoelectric sensitivity while maintaining flexibility. A significant improvement in piezoelectric properties of quasi 1-3 composites is achieved by a combination of dielectrophoretic alignment of the ceramic particles and poling process. The degree of structuring as well as the functional properties of the in-situ structured and poled composites are enhanced significantly compared to those of the conventionally manufactured structured composites.

3D porous scaffolds of poly (lactic acid) (PLA) and chitosan KNLN have also been produced by freeze-casting technique; using the anisotropic solidification of a suspension to achieve aligned porosity for guiding cell outgrowth. Piezoelectric particles of KNLN of varying weight fractions (10, 20, 30 and 40%) have been integrated and poled using corona technique. The resultant composites show enhanced average pore size and interconnectivity as well as piezoelectricity ideal for soft tissue regeneration applications. This research is the first to report the fabrication and material characterisation of highly porous PLA-KNLN and chitosan-KNLN composites prepared by freeze casting. Morphological characterisation of the obtained microstructures has been performed and linked to the processing parameters and final properties. A poling study has also been performed to achieve piezoelectric properties. The highly interconnected aligned structures with ideal mean pore size have been obtained with PLA and chitosan freeze-cast scaffolds and enhanced with the incorporation of KNLN, up to 20wt% desirable for soft tissue applications.

An Evolving Theory for Protein Dielectrophoresis

Ronald Pethig

The University of Edinburgh, UK

Dielectrophoresis (DEP) studies have progressed from the microscopic scale of cells and bacteria, through the mesoscale of virions to the molecular scale of DNA and proteins [1]. Beginning in 1994 with the pioneering work of Washizu *et al* [2] at least 20 different globular proteins have now been studied for their DEP responses [3]. Analyses of these responses have employed the so-called Clausius-Mossotti (CM) function, even though its theoretical derivation fails to describe the situation for nanoparticles such as proteins that possess an intrinsic dipole moment and other subtle physico-chemical attributes [4, 5]. The CM function of DEP is in fact an *analogue* of (but not the same as) the CM-relation that formed the bedrock of classical dielectric theory [*e.g.*, 6] used to describe the electrical polarization of proteins [*e.g.*, 7].

In this presentation the derivations of the *molecular* CM-relation of dielectrics and the *macroscopic* CM function of DEP will be summarized and discussed in terms of a critical assessment of the work reported to date for proteins [8]. The current approaches [9, 10] for an evolving theory of protein DEP will also be presented.

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Session 6 - Focus Topic 1 (Theory and Modelling of Dielectric Materials)

A new take on Electrostriction

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All dielectrics exhibit electrostriction: they all expand or contract under the application of an electric field. The induced strain is proportional to square of the induced polarisation, in contrast to piezoelectricity where the deformation is proportional to the field but only exists in non-centrosymmetric materials.

Electrostriction has long been overshadowed by piezoelectricity because of its linearity and potentially larger effect despite the numerous advantages of electrostriction: it exists at all temperature (no T_c), there is no need to polarise the sample, no intrinsic hysteresis, little to no temperature dependence, and electrostriction exists in a wealth of environmentally-friendly materials. In addition, electrostriction is a major contributor to the electro-optic response of strain-induced birefringence and other elasto-optic, photo-elastic, or piezo-optic effects. Electrostriction also contributes to third-order non-linear optical response. As such, it is a coupling mechanism leading to stimulated Brillouin and Rayleigh scatterings. Furthermore, electrostriction is used to create laser-induced grating, for example to carry out phonon spectroscopy. Outside physics, electrostriction is, for example, responsible for the higher values of protein hydration that plays a major role to understand the protein structure and biological functions; or in electrochemistry where it is a source of internal stress in anodic oxide films, thereby affecting their breakdown.

With the recent discovery of "giant" electrostrictors, there has been a renewed interest in a phenomenon that remain little understood despite having been discovered (just) before piezoelectricity. As the recent claims of "giant" electrostrictions are by far not the first ones to be made, we took a critical look at the literature and propose a quantitative definition of "giant" electrostriction. This enabled us to point out the interest of giant electrostrictors beyond their current use.

In order to substantiate the current empirical proxy used to predict the electrostrictive performances of materials, we have derived analytical expressions of the electrostrictive coefficients as a function of the elastic and dielectric properties of the materials.

We have also derived a methodology to calculate electrostriction from ab initio that is more accurate, faster and more consistent than the few former attempts that relied on the application of an electric field. This methodology also enables to get insights into the mechanism presiding over the electrostrictive response.

In addition, we have proposed a scheme to induce extremely large electrostrictive responses at room temperature in thin films, hoping to promote electrostriction as an environmentally-friendly alternative to piezoelectrics micro and nano-electromechanical devices.

This presentation therefore aims to clarify some misconceptions about electrostriction, to provide a better understanding of its effects, its origin and to provide tools to predict, explain, and exploit this ubiquitous phenomenon.

A New Approach to an Old Idea: Understanding the Fröhlich B(T) Function

Paul Ben Ishai

Ariel University, Israel

In 1949 Herbert Froelich published the Theory of Dielectrics¹. While it is a relatively slim volume, it is widely regarded as one of the fundamental theoretical texts in the science of dielectrics. One of the more thorny subjects he covered was the meaning of the dielectric strength and its relationship to dipole correlation within the material. His conclusion was a reworking of ideas first put forward by Onsager² and Kirkwood³. It was condensed into a function, commonly called the B(T) function, that linked the macroscopic parameters of a dielectric relaxation to the microscopic state of the dipole-dipole interaction, responsible for that relaxation. While elegant, the final equation could only be related to correlated, uncorrelated or anti-correlated dipoles, in terms of an averaged angle, $\langle \vartheta \rangle$, such that $\langle \mathbf{m} \cdot \mathbf{m} \rangle \sim 1 + z \langle \cos \vartheta \rangle$, where $\langle \mathbf{m} \cdot \mathbf{m} \rangle$ is the ensemble averaged scalar multiplication of the dipole moments and z is the number of nearest neighbours.

We present a new derivation of the B(T) function based on the concepts of a correlation length, rather than relative dipole-dipole directions. Using this idea we show that B(T) function actually presents population distributions of dipole moments, each within its own correlation radius. These lengths can then be used to define one of the less tangible concepts in dielectric physics, namely the mesoscale. The paper will show some examples in complex systems of the approach and discuss the implication of the new approach compared to the traditional view.

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A theory for like-charge attraction of polarizable ions

Ho-Kei Chan

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A theory [Journal of Electrostatics 105, 103435 (2020)] that explains how like-charge attraction of polarizable ions could occur is presented. Based on an effective-dipole description, the theory predicts that, for a pair of spherical polarizable ions in vacuum, like-charge attraction is possible but opposite-charge repulsion could not occur, where any occurrence of like-charge attraction is necessarily conditional upon dissimilarities in net charge or polarizable volume between the particles. It also predicts a correlation between the nature of interaction and the alignments of effective-dipole moments. The findings, which echo recent theoretical predictions of like-charge attraction for a pair of dielectric spheres in vacuum, suggest that this counter-intuitive phenomenon could be realized in systems of different length scales through similar mechanisms of particle dissimilarities.

Finite Size Effects in the Two-Dimensional XY Model of Ferromagnetism

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The two-dimensional XY Model is a simplified model of ferromagnetism in statistical mechanics that exhibits a phase transition at non-zero temperature. Because it is not computationally viable to simulate systems of realistic size, the XY Model necessarily restricts the system size to a computable dimension, which alters expectation values of emerging physical quantities of the system. Accordingly, this research employs the Monte-Carlo Metropolis algorithm to investigate finite size effects on physical quantities at the model's critical temperature. Comprehensive analyses are performed for lattice sizes between 10×10 and 300×300 . Results demonstrate that power laws govern finite size effects on the autocorrelation times of both energy density and squared magnetisation, whose exponents are 1.5 and 1.7 respectively. We propose that the autocorrelation time of magnetisation experiences greater finite size effects than the autocorrelation time of energy because the energy is computed from neighbouring spins whilst magnetisation reflects the global dipole moment. In addition, we find that the specific heat and magnetic susceptibility exhibit, respectively, weak variations and non-power law increase with increasing lattice size.

Friday 29 April 2022

Session 7 - Focus Topic 4 (Dielectrics for Electronic Devices)

(Plenary) Deposition of Metal Oxide Thin Films for Large-Area Electronics

Andrew Flewitt

Cambridge University, UK

When we think of electronics, we perhaps most commonly focus on silicon wafer based technology. This has been dominated by Moore's Law where the drive is to increase the performance of microprocessors or memories through increasing the number of transistors whilst maintaining approximately the same area of silicon. In terms of the economics of production, this is achieved by minimising the cost per device and has been primarily realised by reducing the physical size of Metal-Oxide-Semiconductor Field Effect Transistors (MOSFETs).

However, in the display industry, the situation is very different. In this case, we need to be able to produce a layer of thin film transistors (TFTs) over the whole display area, and as the drive has been towards larger and lower cost displays, so the metric for economic production is instead minimising the cost per unit area. This characterises the field of large-area electronics, which has been born out of the display and solar cell industries, but which, as a result is improvement in device performance in recent years, now also includes other application areas such as smart packaging.

The rise of metal oxide thin films as an alternative to silicon thin film technology in TFTs has been a key development over the last ten years. As a result, there has been a move away from chemical vapour deposition to sputtering and atomic layer deposition for production of these metal oxide materials over large areas. These deposition techniques will be compared with a focus on metal oxide thin films for TFTs. This includes both the key deposition parameters which are important and the consequent microstructure and TFT performance that results.

Approaching metal oxide dielectrics and semiconductors by solution and gas phase processing of molecular precursors

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Functional metal oxide semiconductors (MOS) and high- k dielectrics possess a tremendous potential to replace the conventional amorphous silicon/silicon dioxide material combination for next generation electronics, due to the realization of low-voltage operations and enhanced TFT performance characteristics. Solution-processing enables a low-cost and large-area fabrication of metal oxide based materials like semiconductors and dielectrics to be employed in thin-film transistors. Their use may present a simple approach to fine-tune the electrical properties of metal oxides by modification of the materials composition and improvement of the processing conditions.

The employment of most frequently used precursors such as various metal salts or processes like the sol gel approach often requires high decomposition temperatures, subtle control of the reaction conditions (e.g. pH) or the addition of stabilizers or other additives. In our approach we set out to synthesize well-defined molecular precursors first, which then allow to fabricate high quality functional metal oxide thin films from such easy to use, off the bench reagents. The developed molecular oxide precursors display various desirable chemical properties including air-stability, high solubility in aqueous or organic solvents, the requirement of reduced decomposition temperatures and the ability to be processable via photoactivation.^{1,2,3} Along another avenue of research, gas phase techniques like atomic layer deposition (ALD) allow the precise control of semiconductor and dielectric material deposition of thin films as needed for thin film transistors. In this realm we were interested to see if controlled doping with dielectrics can be used to modify the charge carrier density and oxygen deficiency in oxide semiconductors, the latter being foremost responsible for modulating the electronic parameters in thin film metal oxide transistors.^{4,5}

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Study of compositional and thermal dependent negative permittivity of $\text{Sr}_2\text{Mn}_{1-x}\text{Nb}_x\text{O}_4$ ($x = 0.1, 0.2, 0.3$) solid solutions

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Materials with negative permittivity have recently become a research hotspot due to their potential applications in electromagnetic shielding, novel capacitance, coil-less inductor design, stretchable sensors, etc. Herein, from the perspective of intrinsic properties of mono-phase materials, a few compositions of the system $\text{Sr}_2\text{Mn}_{1-x}\text{Nb}_x\text{O}_4$ ($x = 0.1, 0.2, 0.3$) were synthesized in the air by the solid-state ceramic route. A change in the sign (positive to negative) of the permittivity above a particular temperature (T_c) at all the measuring frequencies is speculated for the compositions with $x = 0.1$ & 0.2 and observed in $x = 0.3$. The negative permittivity is analyzed using the Drude-Lorentz model. It was found that negative permittivity is caused by the plasmonic oscillations of thermally excited free charge carriers. The donor substitution of Nb has altered the valence state of Mn, analyzed by XPS spectra, thereby altering electrical conductivity. These studies highlight dependencies of negative permittivity on composition and temperature, which makes the $\text{Sr}_2\text{Mn}_{1-x}\text{Nb}_x\text{O}_4$ system a promising candidate for switching elements in the radio frequency (10 Hz -2 MHz) region.

Heteroepitaxially grown diamond for application in electronic devices

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Fraunhofer IAF, Germany

Diamond offers a set of unique properties such as its high thermal conductivity of $>2\text{kW/Km}$, an intrinsic resistivity of $10^{16}\Omega\text{cm}$ and its high Young's modulus of 900GPa . Epitaxial growth by microwave-plasma chemical vapour deposition (MPCVD) offers the opportunity to produce both intrinsic as well as semiconducting diamond by the inclusion of proper dopants such as Boron for p-type or phosphorous for n-type doping. Since homoepitaxy on substrates grown by high-pressure-high-temperature (HPHT) synthesis is limited to sample areas $<1\text{cm}^2$, heteroepitaxy is currently the only way to get to wafer-scale substrate sizes, for which the current maximum diameter is 9cm [1]. For the growth of (100)-oriented substrates, the layer system Dia/Ir/YSZ/Si(100), in which YSZ stands for Yttria-Stabilized Zirconia, has been shown to be the most suitable by finite-element method calculations[2].

Over the past years, our institute has developed a variety of applications for homoepitaxially grown diamond, see e.g. [3,4], which we will successively attempt to transfer to heterodiamond. We have tested the applicability of our heterodiamond in lithographic microstructuring procedures as well as by fabricating pseudo-vertical Schottky diodes with boron-doped diamond (BDD) for which we present the first results here.

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Session 8 - Focus Topic 4 (Dielectrics for Electronic Devices)

(Invited) Composite Design and Modelling for Novel Energy Harvesting and Sensing Structures

Chris Bowen

University of Bath, UK

The presentation will overview ferroelectric materials and their composites for sensing and harvesting via piezoelectric and pyroelectric effects. The role of forming porous and composite architectures will be examined to improve their interaction with mechanical and thermal loads for improved sensors and low power harvesting. Examples will include the harvesting of mechanical energy, thermal energy, and chemical energy using ferroelectric materials. The motivation of this work is the continuing need for reduced power requirements for small electronic components, such as wireless sensor networks. It will overview of piezoelectric harvesting system along with the closely related sub-classes of pyroelectrics and ferroelectrics. These properties are, in many cases, present in the same material, providing the intriguing prospect of a material that can harvest energy from multiple sources including vibration and thermal fluctuations. Examples of the modeling and manufacture of porous materials and pyroelectric harvesting are discussed and the potential of novel sandwich structures and porous materials are also described. Modelling of the frequency-dependent permittivity and conductivity of such materials are also overviewed using resistor-

capacitor networks, which can explain the 'giant' relative permittivity values that are sometimes reported in related materials.

Light-assisted ferroelectric domain switching for controlled modulation of current

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Opto-electric control of ferroelectric domains and polarization states offers advantages of reduced hysteresis and resistive Joule heating losses in contrast to the conventional electrical poling.¹ This work reports the combined effect of light and low-voltage electric bias for optoelectric control of ferroelectric domains in a semiconducting ceramic material - KNBNNO ((K_{0.5}Na_{0.5})NbO₃ doped with 2 mol.% Ba(Ni_{0.5}Nb_{0.5})O_{3-δ})²⁻⁴. The effect is utilized to achieve two orders of magnitude amplification in electrical response, asymmetric AC modulation and domain wall velocities of 30,000 nm s⁻¹ with ultra-large domain switching areas in fractions of a second. The light illumination on this material acts as a virtual electrode and results in the tuning of the material conductivity. Based on this mechanism, a proof of concept for a monolithic ferroelectric light-effect transistor with a source and drain as electrical contacts while light acting as a virtual gate is demonstrated. The same device is also demonstrated to work as a photodiode, a rectifier and an electrical output modulator. A fundamental understanding of the presented approach could potentially open new dimensions for next-generation photo-ferroelectric and neuromorphic computing devices.

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A detailed Study on Bulk Photovoltaic Effect in Ferroelectric System

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Ferroelectric systems exhibiting open-circuit voltage (V_{oc}) higher than the bandgap have gained tremendous importance as next-generation photovoltaic (PV) materials. Different studies have been carried out to understand the associate mechanisms and make use of these materials for real applications. In this presentation, I will discuss a detailed study of the ferroelectric PV effect from understanding the associated phenomenon to designing strategies for ferroelectric solar cell devices. In the first part of the work, a non-toxic Ba_{1-x}(Bi_{0.5}Li_{0.5})_xTiO₃ ferroelectric system is designed where the dopants influence the band topology in order to enhance the PV effect. In particular, at the optimal doping concentration ($x_{opt} \sim 0.125$) the sample reveals remarkably high photo-generated field $E_{oc} = 320$ V/cm ($V_{oc} = 16$ V). The band structure, examined through DFT calculations, suggests that the simultaneous effect of band topology and ferroelectric polarization plays an important role for large enhancement in PV effect in this family. Additionally, most of the phenomenological theory of the bulk photovoltaic effect (BPVE) is studied on single-phase ferroelectrics. We also have explored the detailed investigation of BPVE on Ba_{1-x}(Bi_{0.5}Li_{0.5})_xTiO₃ having co-existence of tetragonal and orthorhombic phases. The linear and sinusoidal photocurrent response as a function of light intensity and polarization-direction, respectively, elucidate the experimental evidence for the linear BPVE. As the efficiency of the measured PV effect is found to be less effective, in the second part of work, I will describe a few strategies to enhance the PV response in the ferroelectric system along with the result obtained so far. For instance, classic nanostructured ferroelectric materials (BaTiO₃, BiFeO₃ and LiNbO₃)

combined with organic PV materials, photo absorbers etc. could be the ideal system to study the BPVE in order to enhance its efficiency. Overall, in this presentation, I will discuss a general thought of the recent and future direction of ferroelectric PV effect.

Solution processable high – k polymer nanocomposite for low cost energy storage device applications

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Low cost, easily processable polymers are take considerably attention to the field of energy storage device application. However the lower dielectric constant of the polymeric materials limits its applications. We have used oxide nanoparticles as a filler materials inside the PVDF polymer matrix and systematically studied how the functionalization process of TiO_2 and BaTiO_3 improved the dielectric permittivity of PVDF composite material. It has been observed that after surface modification by silane can improve dielectric constant with a value of 23% and 74% at measured frequency of 1 kHz from the pristine composite materials, respectively. Further, a new concept has been introduced where hetero-phase fillers (TiO_2 and BaTiO_3) has been utilized to enhanced the dielectric permittivity of the composite materials. The measured dielectric constant for hetero phase filler where TiO_2 and BaTiO_3 nanoparticles mixed in a ratio of 1:3 is found to be 50.2 which is in comparable with the functionalized BaTiO_3 based composite. The highest dielectric constant we have achieved using the APTES modified hetero-phase based PVDF composite at a volume concentration of 5% is 76.1 measured at a frequency of 1 kHz. The corresponding tangent loss is found to be 0.01 at that condition.

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