

Functional Thin Films and Surfaces Room Pacific F-G - Session C1-1-WeM

Optical Materials and Thin Films I

Moderators: *Silvia Schwyn-Theony*, Evatec AG, Switzerland, *Juan Antonio Zapien*, City University of Hong Kong

8:00am **C1-1-WeM-1 Structural Colors and Flexible Transparent Conductors Based on Thin Film Technology**, *L. Jay Guo*, The University of Michigan, USA **INVITED**

This talk will describe two applications of thin film structures, which covers their design, optimization, and fabrication. The first one is related to structural colors, produced by the light interference phenomenon in a stack of thin films. It has shown great promise as an alternative for the existing colorant-based pigments owing to their noticeable advantages, such as high brightness, durability and stability, and environmental safety. They may find potential applications in energy-efficient displays, special effect coatings, and building-integrated photovoltaics. The structural colors can be produced by exploiting optical resonances in various resonators, which can be either 3D, 2D or 1D structures. To enable scalable manufacturing, layered structures can be designed, and can be easily made by additive PVD deposition, and more recently by solution deposition processes. Deep learning methods have been explored in the inverse design problems by searching for the suitable structures given a desired optical response. We recently benchmarked a few popular machine learning algorithms for the inverse design task against such criteria as accuracy, diversity and robustness. To facilitate and expedite the design of the structures that can satisfy the requirement and suitable for fabrication, we developed a reinforcement learning based approach for automatic design, which treats the optical coating design task as a sequence generation process and proved to be very effective in practical designs. Examples will be given to show the working and effectiveness of these AI-based design methodologies. The second example is a new type of flexible transparent conductor based on ultra-thin Ag film and dielectric films on plastic substrate such as PET. It can be designed to have even greater transparency than the PET substrate itself. Such transparent conductors could find wide range of applications in flexible displays, touch screens and transparent EM shielding, and can be manufactured in large area format by industrial roll sputtering processes.

8:40am **C1-1-WeM-3 High-Rate Deposition of Calcium Fluoride Coatings Using Radio-Frequency Magnetron Sputtering**, *Sharon Waichman, I. Zukerman, M. Buzaglo, S. Barzilai*, NRCN, Israel

Calcium fluoride (CaF_2) is a ceramic material that exhibits versatile properties such as broadband transparency, lubrication behavior at high temperatures (up to 900 °C), high mechanical stability, and hydrophobicity. CaF_2 can be utilized as an antireflective (AR) coating for different optical needs and applications. Due to its enhanced transmission, mainly in the UV and IR regime, a broader transmission range can be achieved, greater than the common oxide-based coatings (SiO_2 , TiO_2 , or their combination). Moreover, CaF_2 as a constituent in composite-based coatings can effectively reduce wear and friction in mechanical devices and working tools that are subjected to heavy loads and high temperatures. One of the challenges in CaF_2 sputtering concerns with the low stability of the target under high sputtering power (above $\sim 2 \text{ W/cm}^2$). Therefore, CaF_2 deposition is usually characterized by low deposition rates. In the current research, CaF_2 coatings were deposited on silicon (Si) substrates by means of a radio frequency (RF, 13.56 MHz) magnetron sputtering technique in an argon atmosphere, and fixed power of 4.4 W/cm^2 , whereas the substrate bias voltage (V_b) was varied from 0 to -150 V. The role of V_b is to enhance the adatoms' mobility and to increase their probability of residing in permanent binding sites. This lowers the roughness and increases the layer density. The coatings were characterized using SEM/EDS, XRD, and XPS. A spectrophotometer was utilized to study the optical properties of the deposited films. The residual stress was studied using the Stoney formula, while the adhesion was studied by a scotch-tape method. In this research, we report a high deposition rate of 8-25 nm/min, depending on the V_b value, with a maximum deposition rate at the low voltages applied, 0 and -30 V, as expected. To the best of our knowledge, this high deposition rate of CaF_2 coatings, using RF magnetron sputtering technique, is reported for the first time. The morphology of the surface was changed with increasing V_b , from a rough surface obtained at the low V_b regime to smoother at -50 to -75 V, which finally became rougher again at high V_b of -150 V, because of pits and holes that were generated by the excessive ion bombardment.

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Moreover, at -150 V, coatings were delaminated from the Si substrate immediately after deposition. Due to preferential sputtering of the light fluorine, the composition of the CaF_2 coatings turned fluorine deficient with increasing V_b .

9:00am **C1-1-WeM-4 Reactive Sputter Deposition of Nanoporous Black Zinc and White Zinc Oxide Coatings**, *Jakub Zawadzki*, Łukasiewicz Research Network - Institute of Microelectronics and Photonics, Faculty of Materials Science and Engineering - Warsaw University of Technology, Poland; *M. Borysiewicz*, Łukasiewicz Research Network - Institute of Microelectronics and Photonics, Poland

A group of metallic materials characterized by low reflectivity is known as black metals. The transition of the metal from being highly reflective to nearly black can be achieved by its preparation in a way providing a highly porous structure. This phenomenon has been widely applied to the synthesis of macroscopic black metal materials based on Ag [1], Au [2], Al [3], Cr [4], Ni [5], Mo [6], P [7], Zn [8] etc. The dominant applications of black metals are in solar cell collectors [9], optical sensing [10], electrochemical sensing [11] and decorative coatings [12]. On the other hand, white coatings can find applications where a high degree of specular light reflection is required – e.g. in cooling coatings [13] or decorative coatings [14]. Depending on the function, several properties must be considered, such as absorbance of electromagnetic radiation in specific wavelengths, thermal emittance, thermal stability and wear resistance.

Our study presents a way to make thin porous zinc and zinc oxide nanosystems obtained by reactive sputter deposition with additional postdeposition annealing as functional black coatings. We show that a wide array of morphologies from highly porous nanocoral and branched structures to densely-porous ones can be obtained by addition of oxygen to inert working gas [15,16]. The purely Zn films are characterized by a wide degree of black or off-black colours. They can be oxidized at an elevated temperature to form matt white ZnO nanoporous films. We show the strategy to increase the mechanical stability and the optical quality of the films, by applying additional protective overcoats. We show that the doping with Al increases the thermal stability of the Zn films for harsh applications. We investigate creating patterns in the black and white films. The influence of morphology, crystal structure and chemical composition on the optical and mechanical properties of the films is discussed.

The comprehensive approach allowed us to achieve highly functional black and white coatings based on zinc with the use of a solvent-free deposition technique widely spread in large-scale thin film industries.

References:

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9:20am **C1-1-WeM-5 High Hall Mobility W-Doped In_2O_3 Conductive Films with Thicknesses of Less Than 10 Nm Deposited on Glass Substrates**, *Tetsuya Yamamoto, R. Palani, H. Makino*, Kochi University of Technology, Japan

We have achieved polycrystalline W-doped In_2O_3 (p -IWO) films with thicknesses $\leq 10 \text{ nm}$ showing high Hall mobility $\geq 50 \text{ cm}^2/(\text{Vs})$. Amorphous IWO (α -IWO) films with thicknesses in the range from 5 to 50 nm were deposited on glass substrates without intentionally heating of the substrates by reactive plasma deposition with dc arc discharge (RPD). Then, the α -IWO were subjected to under-vacuum solid phase crystallization in the RPD chamber at a pressure of $5 \times 10^{-4} \text{ Pa}$ without any additional gas for

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30 min at 250 °C, to realize *p*-IWO films. Structural, electrical, and optical properties are characterized by X-ray diffraction and reflectivity (Rigaku SmartLab), Hall-effect combined with the van der Pauw geometry (Nanometrics HL5500PC), and spectrometer measurements (Hitachi U-4100), respectively. For obtaining high Hall mobility *p*-IWO films, flat surfaces of the IWO films and reduced roughness of film/substrate interface are essential. Resolutions to the above issue are follows: (1) control of the energy of flying particles such as positively charged indium (In⁺) ions of less than 25 eV; (2) to prevent the generation of low-melting-point In metals at the early growth stage of the films, optimization of the flow rates of oxygen (O₂) gasses introduced into the deposition chamber during the film growth. In this talk, we discuss thickness-dependent electrical and optical properties and elucidate the cause of the high carrier transport, on the basis of the analysis of the data obtained by high resolution Rutherford back scattering and hydrogen (H) forward-scattering spectroscopy measurements and combined with classical size effect theoretical models.

9:40am **C1-1-WeM-6 The Effects of Growth and Post-Annealing Temperatures on MoS₂ Thin Films Deposited by Magnetron Sputtering.** *C. Chao*, National Dong Hwa University, Taiwan; *P. Tsai*, National Chung-Shan Institute of Science & Technology, Taiwan; *P. Wu*, Stone & Resource Industry R&D Center, Taiwan; ***Ing-Song Yu***, National Dong Hwa University, Taiwan

Molybdenum disulfide (MoS₂), one of two-dimensional semiconductors, can be applied in various fields such as electronics, optoelectronics, energy storage, catalysis, etc. In our work, large-area and highly-continuous MoS₂ thin film was prepared by magnetron sputtering process at different growth parameters. Then, a post-deposition annealing process was conducted in ultra-high vacuum and in the nitrogen-plasma environments in order to improve the crystallinity and optical properties of the MoS₂ layers. After the deposition and annealing process, MoS₂ thin films were characterized by reflection high-energy electron diffraction, scanning electron microscopy, X-ray diffraction, X-ray photoelectron spectroscopy and Raman spectroscopy. The phase transformation of MoS₂ layers was studied by differential scanning calorimetry. The optical properties of MoS₂ thin films were further investigated by photoreflectance spectroscopy. From the observations, the crystallinity of MoS₂ films can be significantly improved after annealing at 300 °C for 45 minutes in ultra-high vacuum. The increase of the annealing temperature did not further improve the crystallinity, but the surface chemical composition ratio of sulfur and molybdenum decreased at higher temperatures. Moreover, the enhancement for crystallinity of MoS₂ can be also achieved by the deposition temperature of 150 °C.

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Optical Materials and Thin Films II

Moderators: *Silvia Schwyn-Theony*, Evatec AG, Switzerland, *Juan Antonio Zapien*, City University of Hong Kong

2:00pm **C1-2-WeA-1 Perovskite Stannate BaSnO₃ Films for Near- and Mid-Infrared Plasmonic Applications**, *Heungsoo Kim*, *A. Piqué*, Naval Research Laboratory, USA

Recently, ternary perovskite oxides have attracted great attention as alternative transparent conducting oxides (TCOs) because their structures are compatible with many other perovskite oxides that allow devices to be fabricated comprised entirely of perovskite oxides. Among these perovskite oxides, BaSnO₃ has gained considerable attention as a promising TCO because of its high mobility at room temperature ($\sim 320 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ in bulk single crystals and $\sim 100 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ in epitaxial thin films) and high temperature stability in oxygen atmospheres compared to other TCOs, such as Sn-doped In₂O₃, Al-doped ZnO, and F-doped SnO₂. We have grown epitaxial La-doped BaSnO₃ (LBSO) thin films on (001) SrTiO₃ and (001) MgO substrates by pulsed laser deposition and investigated their structural, electrical, and optical properties as a function of the oxygen pressure and substrate temperature during deposition. By adjusting the oxygen pressure and substrate temperature during deposition, we were able to control the film crystallinity and strain, which modified the electrical and optical properties. The LBSO films grown at the optimum conditions (780 °C and 100 mTorr of oxygen) show the highest conductivity ($3.6 \times 10^3 \text{ S cm}^{-1}$) with a carrier concentration of $3.5 \times 10^{20} \text{ cm}^{-3}$ and a carrier mobility of $65 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. This observed high conductivity corresponds to the film with the best crystallinity and the lowest strain state. The permittivity of the LBSO films can also be modified as a function of the oxygen pressure and temperature during deposition allowing tuning of their epsilon-near-zero (ENZ) wavelength from 2 μm to 6 μm . We will present details of the deposition conditions on the properties of LBSO films and the ability to tune the permittivity in infrared range.

This work was supported by the Office of Naval Research (ONR) through the Naval Research Laboratory basic research program.

2:20pm **C1-2-WeA-2 Studies on Sulfur Induced Binary Targets for the Formation of Cu₂ZnSnS₄ (CZTS) Absorber Layer Thin Films for the Fabrication of SLG/Mo/CZTS/CdS/GZO/Al Thin Film Solar Cells**, *Balaji Gururajan*, Yuan Ze University, Taiwan; *B. Rangasamy*, *P. Sankaran*, PSG College of Technology, India; *L. Wei-Sheng*, Yuan Ze University, Taiwan; *D. McIlroy*, Oklahoma State University, USA; *E. Echeverria*, The Center for Bright Beams, Cornell University, USA; *S. Kaliappan*, PSG College of Technology, India; *D. Velauthapillai*, Western Norway University of Applied Sciences, Norway

Copper Zinc Tin Sulfide (CZTS) thin films were prepared on soda lime glass substrates using RF Magnetron Sputtering technique. Sulfur induced Copper Sulfide (CuS), Zinc Sulfide (ZnS) and Tin Sulfide (SnS) sputtering targets were sequentially deposited at 300 °C. Primarily, the sequentially deposited CZTS thin films were studied for its properties and X-ray Diffraction revealed prominent SnS secondary phase in the thin films. So further post deposition sulfurization was performed at 350 °C in Hydrogen Sulfide atmosphere for 60 mins. X-ray diffraction revealed a prominent (112) CZTS kesterite phase, which was also confirmed using Raman Spectroscopy. X-ray Photoelectron spectroscopy studies were carried out on the sulfurized samples, and composition of the thin films were quantitatively estimated. Ultra-violet photoelectron spectroscopy and valance band spectra were used to study the electronic properties of the CZTS thin films. Ultra-violet spectroscopy studies were performed and CZTS thin films had an energy band gap of 1.45 eV. Hall-effect measurements were carried out which confirmed the P-type nature of the CZTS thin films and had a carrier concentration of $4.9 \times 10^{17} \text{ cm}^{-3}$. Finally, the CZTS absorber layer based thin film solar cell was fabricated only by vacuum based techniques as follows, Molybdenum (Mo) was used as the back contact and was deposited using RF Magnetron sputtering, Cadmium Sulfide (CdS) buffer layer thin films were deposited using Hot-wall deposition technique, Gallium doped Zinc Oxide (GZO) window layer was deposited using RF magnetron sputtering and finally Al top contact was deposited using RF magnetron sputtering. The SLG/Mo/CZTS/CdS/GZO/Al thin film solar cell was studied under 1.5AM and revealed low I_{sc} and V_{oc} of 2.6 nA and 0.6 V respectively.

2:40pm **C1-2-WeA-3 Hysteresis on Voltage-Current Characteristics and Optical Responses of PEDOT:PSS/ZnO Nanorods/ZnO:Ga Heterojunctions**, *Tomoaki Terasako*, Graduate School of Science and Engineering, Ehime University, Japan; *M. Yagi*, National Institute of Technology, Kagawa College, Japan; *T. Yamaoto*, Materials Design Center, Research Institute, Kochi University of Technology, Japan

Zinc oxide (ZnO) with a wide bandgap energy of $\sim 3.37 \text{ eV}$ is expected to be applied to ultraviolet (UV) detectors. In general, ZnO exhibits *n*-type conduction because unintentionally doped native defects and/or residual hydrogen (H) atoms act as donors. On the other hand, it is difficult to obtain *p*-type conduction with good reproducibility by intentional impurity doping. Therefore, we have fabricated the UV detectors composed of the heterojunctions between the ZnO nanorods (NRs) and poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS) instead of those of ZnO *pn* homojunctions. In this paper, generation mechanisms of both hysteresis on the voltage-current (V-I) curves and optical responses of the PEDOT:PSS/ZnO NRs/ZnO:Ga (GZO) heterojunctions will be discussed.

The GZO seed layers were deposited on alkali-free glass substrates by ion-plating (IP) with a DC arc discharge using a sintered ZnO pellet containing Ga₂O₃ powder of 4.0 wt.%. Preparations of ZnO NRs layers were done by chemical bath deposition (CBD) using the mixed aqueous solution of zinc nitrate hexahydrate and hexamethylenetetramine. The PEDOT:PSS layer was spin-coated on the surface of the ZnO NRs layer at 3000 rpm for 30 s, followed by thermal annealing in air at 80 °C.

The V-I curves of the PEDOT:PSS/ZnO NRs/GZO heterojunctions exhibited a rectification behavior with hysteresis loops both in forward and reverse voltage regions. Under the irradiation of the ultraviolet (UV) light of 360 nm, the hysteresis loop area in the forward voltage region decreased, but that in the reverse voltage region increased. Both the $\ln V$ vs. $\ln I$ plots and Fowler-Nordheim (F-N) plots, $1/V$ vs. $\ln(I/V^2)$ plots, for the voltage increase in the forward voltage region in a dark state can be clearly divided into three characteristic regions. The V-I curve showed an ohmic characteristic in the low voltage region, whereas the current was approximately proportional to the fourth power of the forward voltage in the high voltage region. Therefore, the possible transport mechanisms in the low and high voltage regions are direct tunneling and F-N tunneling, respectively. In the medium forward voltages, the current was approximately proportional to the square of the forward voltage, which is characteristic of space-charge-limited conduction. The increase in repetition number of V-I measurement under the forward voltage (0 \rightarrow 3 \rightarrow 0 V) in a dark state led to the increase in maximum forward current.

This work was supported by JSPS KAKENHI Grant Number JP22K04220.

3:00pm **C1-2-WeA-4 Effective Ways to Enhance the Performance of n-MoS₂/p-CuO Heterojunction Based Self-Powered Photodetectors**, *KRISHAN KUMAR*, *D. Kaur*, Indian Institute of Technology Roorkee, India

In the present study, two effective routes to improve the response time and the detection range have been investigated for the n-MoS₂/p-CuO (a conventional p-n heterojunction). Initially, an insulating Aluminium nitride (AlN) layer was inserted in between the Molybdenum disulfide (MoS₂) and Cupric Oxide (CuO) layer, which eventually converted the conventional p-n heterojunction to Semiconductor-Insulator-Semiconductor (SIS) with a superior carrier tunneling mechanism. Here, the n-MoS₂/p-CuO and n-MoS₂/AlN/p-CuO (SIS) heterojunctions have been fabricated using the dc magnetron sputtering technique. The responsivities of the n-MoS₂/p-CuO and n-MoS₂/AlN/p-CuO (SIS) heterojunction are found to be 3.88 mA/W and 20.22 mA/W for the visible radiations (532 nm) and 4.47 mA/W and 26.28 mA/W and NIR radiations (1064 nm), respectively. The response time (rise time and decay time) of the fabricated n-MoS₂/p-CuO heterojunction decreases from 93.35 ms and 102.68 ms to 11.31 ms and 12.73 ms with the insertion of ultrathin insulating AlN Layer. The higher current and ultrafast photoresponse in n-MoS₂/AlN/p-CuO (SIS) heterojunction can be ascribed to the carrier tunneling mechanism through the ultrathin insulating layer. Furthermore, the range of detection of the photodetection can be enhanced up to the UV region with the addition of a layer of MoS₂ quantum dots on the surface of the MoS₂ layer in the fabricated heterostructure. The fabricated n-MoS₂ QDs/n-MoS₂/AlN/p-CuO heterostructure shows photoresponse in a broad range from UV to NIR radiations. The recorded values of responsivity for the fabricated n-MoS₂ QDs/n-MoS₂/AlN/p-CuO heterostructure are 4.97 mA/W, 20.22 mA/W and 26.28 mA/W for the incident of UV (376 nm), visible (532 nm) and NIR

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(1064 nm) radiations, respectively. The obtained results demonstrate the n-MoS₂/AlN/p-CuO (SIS) heterostructure with the addition of MoS₂ QDs shows excellent potential for next-generation ultrafast optoelectronics applications.

3:20pm C1-2-WeA-5 Femtosecond Laser Ablation (FESLA) XPS – A Novel XPS Depth Profiling Technique for Optical/Electrical Thin Films and Multi-Layered Structures, Mark Baker, S. Bacon, S. Sweeney, University of Surrey, UK; A. Bushell, T. Nunney, R. White, Thermo Fisher Scientific, UK

XPS depth profiling is a widely employed analytical technique to determine the chemical composition of thin films, coatings and multi-layered structures, due to its ease of quantification, good sensitivity and chemical state information. Since the introduction of XPS as a surface analytical technique more than 50 years ago, depth profiles have been performed using ion beam sputtering. However, many organic and inorganic materials suffer from ion beam damage, resulting in incorrect chemical compositions to be recorded during the depth profile. This problem has been resolved for most polymers through the use of argon gas cluster ion beams (GCIBs), but the use of GCIBs does not solve the issue for inorganics. A prototype XPS depth profiling instrument has been constructed which employs a femtosecond laser rather than an ion beam for XPS depth profiling purposes. This novel technique has shown the capability of eradicating chemical damage during XPS depth profiling for all initial inorganic, compound semiconductor and organic materials examined. The technique is also capable of profiling to much greater depths (10s - 100s microns) and is much faster than sputter XPS sputter depth profiling. FESLA XPS results will be shown for selected bulk, thin film and multi-layered materials employed in optical and electrical applications.

3:40pm C1-2-WeA-6 2-Dimensional Growth of GaS_x Crystal by Low-Pressure Vapor Phase Deposition, Yijia Chen, National Dong Hwa University, Taiwan; C. Huang, National Dong Hwa University, Taiwan

GaS_x is a promising material with band gap of less than 3 eV, which could be used for novel 2-dimensional nano-electronics and nano-photoelectronics. We successfully prepared GaS_x thin film on silicon substrate by low-pressure vapor phase deposition. The examination of the microstructure reveals the evolution of the 2-dimensional GaS_x crystal growth. We found that the GaS_x platelets are just sprouting out of the substrate from those dots coincidentally aligned along some crystallographic direction of the Si substrate. The edges of the platelets show preferred directions, confirming the occurrence of preferred-orientation growth of GaS_x from the substrate. It is very interesting to find that the edges of the platelets are accumulated with the dots similar to those found further downstream on the substrate as the nucleation sites for GaS_x. Apparently, the dots not only signify the remnant for GaS_x nucleation, but also reveal their involvement of the lateral growth of GaS_x crystals. Recall that the dots are where GaS_x is abundant. It is most likely that sulfur is first dissolved in the Ga liquid, then reacts with Ga and precipitates as GaS_x on the existing edge of GaS_x platelets to expand the crystal size. These crystals were later exfoliated into GaS sheets, as evidenced by x-ray diffraction analysis. From this study, a major leap forward is provided toward the realization of 2-D GaS preparation.

Functional Thin Films and Surfaces Room Pacific D - Session C3-1-ThM

Thin Films and Novel Surfaces for Energy I

Moderators: *Clio Azina*, RWTH Aachen University, Germany, *Carlos Tavares*, University of Minho, Portugal

8:40am **C3-1-ThM-3 Tailoring Surface Reactivity of Perovskite Oxides for Water Oxidation**, *Kelsey Stoerzinger*, Oregon State University, USA INVITED
The intermittent nature of renewable energy sources requires a clean, scalable means of converting and storing energy. One Earth-abundant storage option is water electrolysis: storing energy in the bonds of O₂ and H₂, and later extracting electricity by the electrochemical reaction of gasses in a fuel cell. Nickel oxides are notably active for oxygen electrocatalysis in alkaline solutions, and the perovskite structure (ANiO₃) in particular. I will present studies of model oxide electrodes grown by pulsed laser deposition (PLD) and molecular beam epitaxy (MBE) on single crystal substrates that display a known crystallographic orientation, strain, surface area, and path for charge transport. Electrochemical measurements on these heterostructures can establish the intrinsic activity of oxide catalysts in a way that cannot be realized with polydisperse nanoparticle systems, and we use these findings to rationally design the nickelate composition and structure to maximize activity. Additional insight into the mechanism of the oxygen evolution reaction (OER) is obtained from spectroscopic probing of adsorbates with ambient pressure X-ray photoelectron spectroscopy (AP-XPS), pH dependence of activity, and measurements of oxygen isotope exchange. This fundamental understanding aids in the design of active, earth-abundant electrocatalysts for efficient conversion of renewable energy into chemical fuels.

9:20am **C3-1-ThM-5 The Influence of Sb Doping on the Local Structure and Disorder in Thermoelectric ZnO:Sb Thin Films**, *J. Ribeiro, F. Rodrigues, F. Correia*, University of Minho, Portugal; *A. Kuzmin*, University of Latvia; *E. Alves, N. Barradas*, University of Lisbon, Portugal; *O. Bondarchuk*, International Iberian Nanotechnology Laboratory, Portugal; *A. Welle*, Karlsruhe Institute of Technology (KIT), Portugal; *Carlos Jose Tavares*, University of Minho, Portugal

Thermoelectric transparent ZnO:Sb thin films were deposited by magnetron sputtering, with Sb content varying between 2-14 at.%. As evidenced from X-ray diffraction analysis, the films crystallize in the ZnO wurtzite structure for lower levels of Sb-doping, developing a degree of amorphization for higher levels of Sb-doping. Temperature-dependent (10-300 K) X-ray absorption spectroscopy studies of the produced thin films were performed at the Zn and Sb K-edges to shed light on the influence of Sb doping on the local atomic structure and disorder in the ZnO:Sb thin films. The analysis of the Zn K-edge EXAFS spectra by the reverse Monte Carlo method allowed to extract detailed and accurate structural information in terms of the radial and bond angle distribution functions. The obtained results suggest that the introduction of antimony to the ZnO matrix promotes static disorder, which leads to the partial amorphization with very small crystallites (~3 nm) for large (12-14 at.%) Sb content. Rutherford backscattering spectrometry (RBS) experiments enabled the determination of the in-depth atomic composition profiles of the films. The film composition at the surfaces determined by X-ray photoelectron spectroscopy (XPS) matches that of the bulk determined by RBS, except for higher Sb-doped ZnO films, where the concentration of oxygen determined by XPS is lower near the surface, possibly due to the formation of oxygen vacancies that lead to an increase in electrical conductivity. Traces of Sb-Sb metal bonds were found by XPS for the sample with the highest level of Sb-doping. Time-of-flight secondary ion mass spectrometry obtained an Sb/Zn ratio that follows that of the film bulk determined by RBS, although Sb is not always homogeneous, with samples with lower Sb content (2 and 4 at.% of Sb) showing a higher Sb content closer to the film/substrate interface. From the optical transmittance and reflectance curves, it was determined that the films with the lower amount of Sb doping have higher band-gaps, in the range of 2.9 – 3.2 eV, while the partially amorphous films with higher Sb content have lower band-gaps in the range of 1.6-2.1 eV. Albeit the short-range crystalline order (~3 nm), the films with 12 at.% of Sb have the highest Seebeck coefficient (~56 mV/K) and a thermoelectric power factor of ~0.2 mW·K⁻²·m⁻¹.

9:40am **C3-1-ThM-6 An Economic Experimental Approach to Optimize the Microstructure and Thermoelectric Performance of ZnSe Thin Films**, *Khalid Mahmood*, Department Of Physics, Government College University Faisalabad, Pakistan

Current study demonstrated the growth of good quality, smooth surface ZnSe thin films by an economic and safer thermal evaporation method. The quality of grown thin films was modulated by varying the source to substrate distance (SSD) from 5-15 cm with a step of 5 cm during the thermal evaporation process. XRD scans indicated the pure phase formation and SEM analysis has confirmed the uniformity and smooth surface growth of these deposited thin films. Crystallite size is found to be increased from 28.73-40.18 nm with the increase in source to substrate distance. Raman analysis has further confirmed the cubic phase crystalline nature of these films. Transport properties such as the electrical conductivity, Seebeck coefficient [https://www.sciencedirect.com/topics/physics-and-astronomy/seebeck-effect] and power factor were observed to be varied with source to substrate distance. The highest values of Seebeck coefficient, electrical conductivity and power factor (260 μV/C, 4.1 S/cm and 2.21344 x 10⁻⁶ W/mK² respectively) were achieved by varying SSD.

10:00am **C3-1-ThM-7 Designing Thin Film for Li-Solid State Batteries**, *Haemin Paik*, MIT, USA; *J. Rupp*, Technical University Munich, Germany INVITED

With an increasing demand in portable electronic devices and electric vehicles, breakthroughs in energy density, cycling rate, and shelf-life are necessary to advance energy storage technologies. Extensive research efforts have been focusing on reviving Li metal as an ultimate anode material due to its outstanding theoretical specific capacity and low electrochemical potential. Although using Li metal with conventional liquid electrolyte would achieve high energy density, safety concerns associated with material instabilities and heat release during Li dendrite growth through the electrolyte triggering internal short circuit of the battery move Li metal further away from practical applications. To suppress Li dendrite formation and improve overall battery safety, inorganic ceramic solid electrolytes are regarded as a key to mitigate the problem, and the interest of all solid state batteries (SSB) has grown rapidly over the last decade. In order to replace the conventional liquid Li-ion electrolyte, solid electrolytes have to be thin and uniformly dense for comparable Li-ion conductivity and reliability.

In this talk, recent developments on ceramic processing of Li-garnet solid electrolyte thin film from several hundreds of nanometers to micrometers thin with high Li ionic conductivity and at low synthesis temperatures will be introduced. The effect of these new fabrication methods on atomic structure and microstructure of the film will be discussed and the correlation of structure with Li ion transport kinetics as well as battery performance will be reported. Collectively, this will give new perspectives on advanced Li-ion solid-state electrolyte films, which can contribute to designing SSB architecture with more storage volumes, safer and longer cycle life at lowering synthesis temperatures and costs.

10:40am **C3-1-ThM-9 Hydrothermal-Based Synthesis of Piezo-Composite Thin Films and Their Applications**, *Thi Nghi Nhan Nguyen, K. Chang*, National Cheng Kung University, Taiwan

Piezoelectric materials have shown key characteristics for engineering and electrochemical applications, such as in sensors and actuators. The heterojunction has been investigated to prove the feasibility of the built-in electric field, which promotes the improvement of photocatalytic efficiency and the piezoelectric nanogenerator (PNGs) applications. In this research, detailed studies focused on piezo-composite thin films and their relative piezo-applications were reported. BiFeO₃ (BFO) microplates were grown on the ZnO nanorod array using the hydrothermal method, effectively reducing excess electrons in the ZnO layer. The BFO-ZnO composite act as a power generation source under a force from a motor stepper or external resources such as human hands and a pen. The output voltages and currents of the fabricated PNG were measured. The results clearly demonstrate the effectiveness of the BFO-ZnO heterostructure for realizing high performance, revealing the high durability and stability of devices. The piezo-photocatalytic application such as piezo-photodegradation and piezo-photoelectrochemical water splitting were studied under light irradiation and external stress. Moreover, the piezotronic effect on the performance of the BiOBr/Carbon fibers-based glucose sensor was systematically investigated under various glucose concentrations. The BiOBr/Carbon fibers exhibited a large surface area and low band gap, facilitating improved electrochemical reactivity toward glucose oxidation.

The piezo-electrochemical results indicated that the piezotronic effect significantly increased the sensitivity as well as improved the sensing resolution of the BIOBr/Carbon fibers-based glucose sensors. The working mechanism of coupling the piezoelectric effect and photoexcitation in composite has been proposed based on the analysis of the band energies in the heterojunctions. This work demonstrates a promising approach to improve the sensitivity and generally improve the performance of the PNGs, glucose sensors, and the photocatalytic activities of composites.

11:00am **C3-1-ThM-10 A Comparative Study of the Thermochromic Performances of VO₂ Films Obtained by Air Oxidation of V and VN Precursors**, D. Pilloud, A. Garcia-Wong, F. Capon, Jean-François Pierson, Institut Jean Lamour - Université de Lorraine, France

Thanks to its metal-to-insulator transition (MIT) temperature not so far from the ambient one, thermochromic VO₂ exhibits great potentiality in smart coatings devoted to energy purposes. Among the different methods to synthesize thermochromic VO₂ films, reactive sputter deposition remains one of the most suitable for cost-effective industrial-scale production. For a few decades, increasing attention has been paid to a fast and relatively simple approach based on two steps to produce thermochromic VO₂ thin films: synthesis of a vanadium thin film followed by its oxidation at moderate temperatures (often less than 500 °C). Recently, we have revealed that the oxidation of vanadium nitride (VN) thin films also give rise to the formation of thermochromic VO₂. This work aims to compare the thermochromic properties of VO₂ films achieved after thermal air oxidation of V and VN thin films prepared by sputtering deposition from an elemental vanadium target. For both precursors, short-time air annealing (from 60 to 300 seconds) was performed at 550 °C. X-ray diffraction and Raman spectroscopy depicted that the thermal oxidation domain allowing the formation of the thermochromic VO₂ phase was enlarged for VN compared to V precursor films. These results were confirmed by electrical measurements of the resulting oxides in the temperature region that encompass the MIT of thermochromic m-VO₂. Finally, a particular focus was made on the oxides of the VN series in terms of their infrared properties. The results indicate that the oxidation of VN precursor allows the synthesis of monoclinic VO₂ films with high purity displaying good thermochromic performances (IR contrast of 63%, electrical switch of ~ 2 decades) with narrower hysteresis widths (< 9.3 °C) as compared to those obtained by vanadium films oxidation, making VN precursor of particular interest for industrial upscaling.

11:20am **C3-1-ThM-11 Revolutionizing Concentrated Solar Thermal Power Technology: Developing Self-Cleaning Mirrors with TiO₂ Films**, Nafsika Mouti, V. Terziyska, N. Kostoglou, Montanuniversität Leoben, Austria; A. Kaidatzis, M. Arfanis, National Centre of Scientific Research "Demokritos", Greece; A. Eliades, K. Milidonis, The Cyprus Institute, Cyprus; K. Giannakopoulos, National Centre of Scientific Research "Demokritos", Greece; C. Mitterer, Montanuniversität Leoben, Austria

Concentrated Solar Thermal (CST) technologies constitute a promising approach for harnessing the power of the sun to drive high-temperature thermal processes of interest, such as a power block for electricity generation, high-temperature industrial process heat or high-temperature solar chemistry applications. The Light Collection and Concentration Subsystem (LCCS) of a CST plant is responsible for collecting the direct solar radiation and concentrating it onto the active surface of a receiver system, where the thermal energy conversion takes place. The LCCS use mirrors (usually of parabolic shape) to concentrate the sunlight onto the receiver, however, the accumulation of dust and other particles (e.g. mirror soiling) on the mirrors has a direct effect on the efficiency of the LCCS. To address this issue, the Nano4CSP project with partners from Austria, Greece and Cyprus focuses on the development of self-cleaning CST mirrors using TiO₂ films. TiO₂ is a well-known photocatalyst, capable of breaking down dirt and other contaminants upon exposure to sunlight, making it a suitable material for creating self-cleaning surfaces. Within this study, sputtering has been successfully used to deposit TiO₂ films on CST mirrors at room temperature under various growth conditions. The deposited films with thicknesses in the range of 10 to 1000 nm have been extensively characterized using scanning electron microscopy with energy-dispersive X-ray spectroscopy, X-ray diffraction, Raman spectroscopy, optical measurements, nanoindentation, nanoscratching and field tests. Films with a dominant anatase phase have been found to show self-cleaning properties, thus improving the mirrors efficiency. Additionally, doping of TiO₂ films with N₂ and Zr has been evaluated, to improve the transparency and photocatalytic properties and to maximize the performance of the mirrors. The obtained results indicate that sputter deposition represents a promising method for producing self-cleaning CST mirrors, since it allows a

precise control over the thickness and uniformity of the films, as well as the ability to apply them at room temperature, thus reducing costs and increasing efficiency for the necessary large area deposition on mirror surfaces. The findings of this research can have a significant impact on the further development of CST technology and its widespread implementation in larger scales, increasing the cost competitiveness of such plants by reducing the operational and maintenance costs and the associated water consumption for mirror cleaning.

11:40am **C3-1-ThM-12 The Influences of Plasmonic Resonance and Coupling Effect on Photocatalysis of MoS₂/Gold Hybrid Nanoparticles for Hydrogen Production**, Yi-Hsueh CHEN, J. Ruan, National Cheng Kung University (NCKU), Taiwan

Hydrogen energy is clean and more friendly to our environment, which drives scientists around the world to look for materials able to catalyze hydrogen production. MoS₂ has been recognized as the most efficient photocatalyst for hydrogen evolution among non-noble metals. In particular, MoS₂ nanosheets exposed lots of active sites for the attachment of proton and later reduction reactions, and the efficiency is better than other bulk materials. Unfortunately, the absorption wavelength of MoS₂ nanosheets is only within the UV region. Visible light accounts for 95% of sunlight and UV light occupies only 5%. It is vital for photocatalysts to be able to efficiently harvest visible light. The absorption of visible light is able to cause strong localized surface plasmon resonance (LSPR) of gold nanoparticles (AuNPs), which has been widely investigated able to promote the performance of MoS₂ as a photocatalyst. However, the desired dispersion patterns of AuNPs for the optimization of surface plasmon resonance are less achievable. As an approach to maximize the amount of energy absorbed from the sunlight, we aim to design and fabricate hybrid particles composed of AuNPs and MoS₂ nanosheets with the control of coupling effect of among AuNPs. Through the achieved adjustment of separation distances among AuNPs, we are able to clarify the required condition for the best effect LSPR to absorb visible light and thus to optimize the efficiency of electron transition from AuNPs to MoS₂ nanosheets, which largely enhances desired hydrogen production.

To prove the influence of local surface plasmonic resonance and coupling effect to achieve hot electron transfer and reflect in hydrogen production, we fabricated a plasmonic nanostructure with gold nanoparticles (Au NPs) / MoS₂ nanosheets. The distribution of Au NPs fabricated by a steric hindrance of PVP to avoid the characteristic that colloidal particles tend to agglomerate during precipitation. In the MoS₂-generated portion, the PVP segment originally attached to the gold surface is squeezed out by grafting the MoS₂ precursor thiourea, so that MoS₂ grows directly on the gold and maintains a specific spacing between the Au NPs on MoS₂ nanosheets.

12:00pm **C3-1-ThM-13 Self-Reconstruction of Sulfate-Containing High Entropy Sulfide for Exceptionally High-Performance Oxygen Evolution Reaction Electrocatalyst**, Thi Xuyen Nguyen, Y. Su, C. Lin, J. Ting, National Cheng Kung University (NCKU), Taiwan

Novel earth abundant metal sulfate-containing high entropy sulfides, FeNiCoCrX₂ (where X = Mn, Cu, Zn, or Al), have been synthesized via a two-step solvothermal method. We show that sulfate-containing FeNiCoCrMnS₂ exhibits superior OER activity with exceptionally low overpotential of 199, 246, 285, and 308 mV at current densities of 10, 100, 500 and 1000 mA cm⁻², respectively, and surpassing its unary-, binary-, ternary-, and quaternary-metal counterparts. The electrocatalyst yields exceptional stability after 12000 cycles and 55 h of durability even at a high current density of 500 mA cm⁻². Various *in-situ* and *ex-situ* analyses were used to investigate the self-reconstruction of the sulfides during the oxygen evolution reaction (OER) for the first time. The resulting metal (oxy)hydroxide is believed to be the true active center for OER. The remaining sulfate also contributes to the catalytic activity. Density function theory calculation is in a good agreement with the experimental result. The extraordinary OER performance of the high entropy sulfide brings a great opportunity for desirable catalyst design for practical applications.

Functional Thin Films and Surfaces Room Pacific F-G - Session C2-1-ThA

Thin Films for Electronic Devices I

Moderators: Julien Keraudy, Oerlikon Balzers, Oerlikon Surface Solution AG, Liechtenstein, Jörg Patscheider, Evatec AG, Switzerland

1:20pm **C2-1-ThA-1 An Imperfect High k Dielectric (O Vacancies, Contamination) Can Give a Perfect MIM Device, Christophe Vallee, N. Tokranova, K. Beckmann, SUNY College of Nanoscale Science and Engineering, USA; N. Cady, SUNY college of Nanoscale Science and Engineering, USA**

INVITED

The reduction of high k dielectric thicknesses below 10 nm gives rises to new potential application of these materials due to unexpected "resistance switching" which is a phenomenon by which some electrical insulators display a change of resistance upon application of a dc bias voltage. Increasing the voltage across an insulator inevitably leads to breakdown, which corresponds to an abrupt resistance decrease. In some circumstances, it is possible to restore a high value of resistance by applying a voltage of opposite polarity. Switching between a low and high resistance state can be repeated by voltage sweeping, leading to a hysteresis loop in the current-voltage (I-V) characteristic. Resistance switching is observed in a variety of metal oxides thin films. In recent years this phenomenon has attracted interest for the fabrication of memory devices ("resistive memory") and for the fabrication of voltage-controlled resistors ("memristors"). At present, physical mechanisms at the origin of resistance switching are based on field-enhanced electrode diffusion into the oxide (in that case the devices are named "conductive bridging memories" or CBRAM) and voltage-controlled creation and annihilation of oxygen vacancies (named "OXRAM" devices).

Within this presentation we will discuss the use of high k dielectrics and their fabrication for metal-insulator-metal (MIM) devices such as linear MIM capacitors, resistive memories, and MIM diodes. We will also discuss the use of resistive switching for neuromorphic computing and compute-in-memory applications, and the integration of resistive memory devices with CMOS. Whatever the device, due to the very small thickness of the high k dielectric, it seems obvious that the electrode materials as well as the interfaces between the insulator and the metal electrodes must be optimized. But more importantly, we will try to elucidate how impurities (like contamination from precursors used for the deposition process) and defects (O vacancies) can be intentionally added to improve the device properties. Ultimately, optimization of device performance is paramount for adoption of these devices for advanced, highly efficient computing hardware.

2:00pm **C2-1-ThA-3 Optoelectronic and Thermoelectric Properties of New Heterobilayers of Janus-Type Noble-Metal Chalcogenides Materials, Mourad Boujnah, CINVESTAV-Unidad Queretaro, Mexico**

Janus and non-Janus monolayers are one of the transformations of the 2D materials viz present an exceptional opportunity to control and manipulate their physical properties. Herein, we predict the two-dimensional of noble-Metal Chalcogenides (NMCs) materials A_2B ($A = Ag, Au$ and $B = S, Se$) heterobilayers (HtBLs) through first-principles calculations. The Ab initio molecular dynamics simulations demonstrate that these monolayers possess excellent dynamic and mechanical stabilities. According to that, a combination of Janus and non-Janus of NMCs monolayers (MLs) and HtBLs. High optical absorption of around $4.5 \times 10^5 \text{ cm}^{-1}$ and high anisotropic carrier mobility of $\sim 10^5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ is observed, which indicates that they may shine in the next generation of electronic and optoelectronic devices. All of these explorations not only enhance the types of 2D materials but also provide a structural reference for designing new MLs on the molecular level. The band gap values of α and β phases calculated at the HSE06 level are between 1.35 and 3.70 eV. The calculated lattice thermal conductivity of Ag_2S (about $0.85 \text{ W m}^{-1} \text{ K}^{-1}$) is low while the electrical conductivities and Seebeck coefficients are high at room temperature. Thus, the properties of these combinations show a high potential for thermoelectric applications.

2:20pm **C2-1-ThA-4 High-Entropy Ba(Ti,Zr,Ta,Hf,Mo)(on)₃ Gate Dielectric Films for Zn-Channel Thin Film Transistors, Van Dung Nguyen, Department of Materials Science and Engineering, National Cheng Kung University (NCKU), Taiwan, Viet Nam; K. Chang, Department of Materials Science and Engineering, National Cheng Kung University (NCKU), Taiwan**
In this study, high entropy Ba(Ti,Zr,Ta,Hf,Mo)(ON)₃ dielectric films were fabricated on an ITO/glass substrate via a combinatorial sputtering

technique. Favorable compositions were efficiently determined from a wide range of the Ba(Ti,Zr,Ta,Hf,Mo)(ON)₃ space. The high entropy oxynitride (HEON) was then incorporated in a metal-insulator-metal (MIM) stack and thin film transistor to investigate dielectric and electrical performance. HEON exhibits high dielectric constant and low dielectric loss, which is promising for MIM and TFT applications. HEON-based TFTs show excellent performance with a high on/off current ratio of 10^8 , low threshold voltage (V_T) of 0.14 V, a low subthreshold swing of $0.08 \text{ V} \times \text{dec}^{-1}$ and interfacial defect (D_{it}) of $7 \times 10^{10} \text{ eV} \times \text{cm}^{-2}$. Moreover, the devices exhibited stable performance with small V_T shifts and negligible changes in the maximum drain current under gate bias stress (GBS). HEON-based TFTs outperformed various reported thin film transistors, indicating the great promise of Ba(Ti,Zr,Ta,Hf,Mo)(ON)₃ as a dielectric layer in thin film transistors.

2:40pm **C2-1-ThA-5 Hydrothermal Fabrication of The Heterojunction of BaTiO₃ Nanorod Arrays with Ag₂O and their Applications, Yen-Lun Chiu, K. Chang, National Cheng Kung University (NCKU), Taiwan**

BaTiO₃ (BTO) is an attractive material because of its excellent piezoelectric characteristics. The property can be tailored through morphology control, specifically in the form of nanoparticles or one-dimensional nanorod and nanowire arrays. However, studies on this topic directly through hydrothermal processes for the fabrication of three-element compounds are still lacking. In this study, the nanocomposite, which combined well-aligned BTO nanorod arrays and Ag₂O nanoparticles, were synthesized through a single or two-step hydrothermal reaction by the addition of various surfactants e.g., polyethylene glycol-400 (PEG-400). To study the morphology effect on the junction, hydrothermal parameters, including concentrations of precursor solutions, reaction time, temperatures, different surfactants, and substrate positions in an autoclave, were manipulated. X-ray diffraction and scanning electron microscopy were employed to determine the phase and morphology of the resultant composite samples. In addition, an atomic force microscope was employed to determine the topography of the samples, and the amplitude of the piezoresponse (d_{33}) was measured through a piezoresponse force microscope. The morphology effect on the piezoelectricity and the optoelectronics of the samples was systematically studied and optimized for related applications.

3:00pm **C2-1-ThA-6 Toughening Mechanisms of Al Nanoparticles in Flexible Mo Thin Films Revealed by in-Situ Synchrotron Diffraction Experiments, Barbara Putz, T. Edwards, Empa, Swiss Federal Laboratories for Materials Science and Technology, Thun, Switzerland; P. Kreiml, Erich Schmid Institute of Materials Science, Austrian Academy of Sciences, Leoben, Austria; E. Huszar, L. Pethö, Empa, Swiss Federal Laboratories for Materials Science and Technology, Thun, Switzerland; D. Többsens, Helmholtz Zentrum Berlin, Germany; J. Michler, Empa, Swiss Federal Laboratories for Materials Science and Technology, Thun, Switzerland**

The combination of a nanoparticle gas phase condensation source with physical vapour deposition opens up plentiful potential for the fabrication of unique nanocomposite thin films, as it allows incorporation of nanoparticles with tailored size, shape, distribution and volume density without precipitation-related restrictions concerning phase diagrams and solubility. In this work, we use combined magnetron sputtering and nanoparticle (NP) deposition to fabricate Mo thin films (200 nm thickness) with and without incorporated Al NPs (diameter 50 nm) on flexible polymer substrates. Microstructure, NP size and concentrations of our nanocomposites were characterized by cross-sectional scanning (SEM) and transmission electron microscopy (TEM). Pure Mo layers are frequently used as part of the metallization of flexible thin film transistors; however they suffer from inherently brittle electro-mechanical behaviour, with through-thickness cracks forming at very low applied strains. To study deformation mechanisms as a function of Al nanoparticle concentration and test the hypothesis that ductile nanoparticles incorporated into the brittle matrix can introduce toughening effects such as crack deflection, arrest or bridging, the electro-mechanical behaviour of the nanocomposites and pure Mo reference films was studied with X-ray diffraction and four point probe resistance measurements during polymer-supported in-situ tensile experiments. Results indicate a clear toughening of Mo thin films with increasing amounts of incorporated Al nanoparticles, as revealed by the shape of the film stress curves recorded parallel and perpendicular to the loading direction. The less severe degradation of electrical conductivity during straining indicates that the nanoparticles limit the extend of mechanical failure of the Mo matrix. Combined with post-mortem focused ion beam (FIB) and SEM analyses of tensile induced cracks, this shed further light on the role of Al nanoparticles during crack

initiation and propagation. In summary, the novel manufacturing approach and resulting nanocomposites are promising candidates for multifunctional thin films in next-generation electronic devices.

3:20pm C2-1-ThA-7 Fabrication of 5g sub-6 Ghz Antennas on Polyimide Substrates Using Laser Thermo-Responsive Polymer Silver Nanocatalysts, *Y. Chen, J. You,* National Defense University, Republic of China; *M. Youh,* Ming Chi University of Technology, Taiwan, Republic of China; *C. Lee, T. Chiang, Chang-Pin Chang, M. Ger,* National Defense University, Republic of China

In the study, laser-activated nanosilver catalyst inks were used to fabricate copper-selective metal patterns for sub-5G-6 GHz antennas. The activity of the catalyst ink can be performed by laser activation treatment. Electroless plating was performed on catalytically active polyimide substrates used to form metallic antenna patterns. PEI-GA was synthesized from polyethyleneimine (PEI) and Glutaraldehyde (GA), followed by a heat-curable Ag composite ink composed of Ag salt and tert-butyl peroxybenzoate (TBPB) to enhance the adhesion between the Cu coating and the PI substrate. After laser activation of the lacquer ink and continuous copper chemical deposition on PI, a layer of copper with good adhesion will be formed on the PI substrate. Copper patterns can be easily prepared by combining laser direct patterning of lacquer inks and electroless copper deposition. This is a way to quickly obtain the best metallic wire patterns on polyimide substrates for fabricating sub-6 GHz antennas for 5G. FTIR-ATR analysis and characterization of monomer and polymer functional group changes before and after cross-linking, and verification of substrate metallization by SEM, EDS, and Cross-cut tester were performed. The laser direct writing process is fast and easy to apply and is expected to replace the existing printed circuit process in the future. The produced metallic wires can also be used in 5G antennas. There is great potential for development in the market. A convincing method and potential solution can be given in this study.

3:40pm C2-1-ThA-8 Metal-Semiconductor Amorphous Boron Carbide Contacts, *Vojislav Medic, N. Ianno,* University of Nebraska - Lincoln, USA

The development of amorphous hydrogenated boron carbide (BC) devices is an increasingly important research topic due to its application in radiation safety and deep space exploration. To improve the device performance research has been conducted in optimization of BC, its fabrication and through it, its electronic properties. However, to our knowledge, a study of metal-semiconductor amorphous boron carbide contacts has not been performed. In addition to understanding and quantifying the effect of contact resistance on device performance, it is important to differentiate metals that form Schottky or Ohmic contacts with BC. The spreading resistance model is used to quantify BC resistivity and metal-BC contact resistance. As the thickness of the deposited BC film is much smaller than the radius of the metal contact area, spreading resistance is linearly dependent on film thickness.

$$R_s = \rho * b / (\pi a^2)$$

b – film thickness, a – contact radius, ρ – resistivity of BC

Since the total resistance can be calculated from Ohmic measurements, the equation below shows the linear dependence of total resistance on BC thickness.

$$R = \rho * b / (\pi a^2) + 2 * R_c$$

R_c – contact resistance on each side of BC films

The linear equation for total resistance can be extrapolated to $b=0$ from the respective I-V curves of various thickness BC films with identical metal contacts, yielding twice the contact resistance. Once R_c is determined ρ can be calculated from any I-V curve. The I-V curves are collected from varying thickness BC films deposited via PECVD from orthocarborane/Ar or metacarborane/Ar gas mixtures, on a plane of metal with the constant size metal dot top contact deposited in a subsequent step. This effectively forms a BC resistor (Figure 1). Specific metals can be categorized as Schottky or Ohmic type contacts by observing the I-V curve measurements. Figure 2 shows the data collected for Cr contacts and Ti contacts on n-type BC. Both of those metals form a Schottky contact, which results in two diodes in series when a completed device is made. However, in Figure 3 Cr contacts on p-type BC structures show an Ohmic contact, and the resistance of this structure can be analyzed using the spreading resistance model previously discussed. As BC deposited from orthocarborane or metacarborane polymeric precursors is p-type or n-type respectively, we plan to study how different metals from the same groups in the periodic table interact with either p-type or n-type BC, whether they produce

Schottky contacts or Ohmic contacts, with the goal of quantifying losses that occur at the metal-BC junction due to contact resistance.

4:00pm C2-1-ThA-9 Bipolar Resistive Switching Characteristics in Functional Aln/Fsma Based Me Heterostructure for Highly Flexible Ram Application, *Pradeep Kumar,* Indian Institute of Technology, Roorkee, India; *D. Kaur,* Indian Institute of Technology Roorkee, Roorkee, India

Resistive random access memory (ReRAM) has been a promising future candidate for various applications such as nonvolatile data storage and neuromorphic devices. Moreover, multifunctional flexible electronics is the ongoing demand for fabricating wearable data storage and communication devices. The magnetoelectric (ME) heterostructure consisting of piezoelectric (AlN) and ferromagnetic magnetic shape memory alloy (FSMA (Ni-Mn-In)) was fabricated over flexible substrate for resistive random access memory application. The Cu/AlN/FSMA metal-insulator-metal (MIM) based memory cell displays bipolar resistive switching (RS) behavior. The formation of Cu metallic filament at a particular SET voltage leads the memory cell in a low resistance state (LRS) from its pristine high resistance state (HRS). The LRS and HRS are explained well by Ohmic and space charge limited conduction (SCLC) mechanisms, respectively. The fabricated memory cell displays excellent endurance (~ 5000 cycles) and data retention ($\sim 10^4$ seconds) capability with high OFF/ON ratio of $\sim 1.2 \times 10^3$. Furthermore, the multifunctionality of the ME heterostructure-based RAM was investigated by tuning the SET voltage with ambient temperature and external magnetic field remotely. A significant change in the SET voltage could be ascribed to the temperature and magnetic field-induced strain transferred to the AlN piezoelectric layer from magnetostrictive FSMA (Ni-Mn-In) bottom electrode. The residual Lorentz force explains the remotely controlled LRS and HRS in transverse magnetic field for multi-bit data storage applications. Moreover, the RS characteristics remain stable even after 800 bending cycles as well as with bending angle (0 to 180°). Hence, the present ME heterostructure integrated with flexible SS substrate can be a better choice for highly flexible, low-cost and multifunctional futuristic RAM application.

4:20pm C2-1-ThA-10 Modifying Oxidation State Distribution in Interfacial Layer of Ge Nmosfet with Pre- or Post- Remote Plasma Oxidation Treatment, *Pei-Hsiu Hsu,* National Tsing Hua University, Taiwan; *D. Ruan,* Fuzhou University, China; *K. Chang-Liao,* National Tsing Hua University, Taiwan

In this research, the effect of pre- or post- remote plasma oxidation treatment on interfacial layer (IL) of germanium (Ge) n-type metal oxide semiconductor field effect transistor (nMOSFET) has been discussed in detail. The pre-remote plasma oxidation treatment, which is similar with traditional post IL oxidation annealing, might reduce the unstable oxidation state in the IL. However, the remaining plasma damage may still enlarge surface roughness and induce high gate leakage current. On the other hand, the post-remote plasma oxidation treatment can effectively passivate the oxygen vacancy with well-bonded oxygen atom, instead of a post high-k deposition annealing process. Nevertheless, it seems that IL quality might not be further improved by the post high-k oxidation treatment. After analyzing X-ray photoelectron spectroscopy and electrical characteristics, it is found that the Ge nMOSFETS with pre-remote plasma oxidation treatment exhibits low subthreshold swing and high on-off current ratio. It may provide an important reference for high performance Ge device fabrication.

Functional Thin Films and Surfaces

Room Golden State Ballroom - Session CP-ThP

Functional Thin Films and Surfaces (Symposium C) Poster Session

CP-ThP-1 Structural and Compositional Analysis of Titanium-Based PVD Coatings, *Celia Rojo-Blanco*, Sheffield University, UK, Mexico; *J. Qi*, Sheffield University, UK, China; *G. Wu, L. Yang*, University of Leeds, UK, China; *S. Creasey-Gray, A. Leyland*, Sheffield University, UK

Novel Ti-Al-B and Ti-Al-B-N coatings have been deposited by plasma-assisted sputter PVD, with a view to constructing erosion-resistant metallic/ceramic multi-layered coatings for demanding environments of high humidity or temperature. Sample coatings of 1.2 to 1.6 μm thickness were produced on both AISI 316 and Si-wafer substrates, and were studied using optical profilometry, XRD, SEM, TEM and EPMA to analyse their (nano)structure and chemical/phase composition.

TEM samples were prepared in both plan-view and cross-section. It was found that the coatings were constituted primarily of an amorphous solid solution with embedded nanocrystals – of either intermetallic (in the Ti-Al-B films), or boride/nitride (in the Ti-Al-B-N films) compounds. Clear TEM Electron Diffraction Patterns were not formed, even though we could observe some XRD reflections, indicating some degree of crystallisation within the coatings deposited.

It was also observed that the coatings contained a significant amount of oxygen 'contamination', distributed primarily at nanograin boundaries.

CP-ThP-3 Study of Spatial Distribution of Sputtered Al-Doped Zinc Oxide for Optoelectronic Applications, *Eduard Llorens, E. Stamate*, DTU, Denmark

Transparent conducting oxides (TCOs) are needed for a wide range of applications in thin film devices such as thin film transistors, solar cells, and smart windows.

Tin doped indium oxide (ITO) is the most commonly used TCO material since it possesses high electrical conductivity and optical transmittance. Nevertheless, indium is scarce and expensive, hence, it would be desirable to find a cheaper and more abundant TCO material that could replace ITO. Aluminum doped zinc oxide (AZO) stands as one of the most suitable candidates due to its earth abundance, high stability, and non-toxicity. Despite its advantages, AZO suffers from low spatial uniformity when it is grown as a thin film using sputter deposition. This issue has been attributed to the bombardment of the film by high-energy negative oxygen ions generated at the sputter target and accelerated in the plasma region between the target and the substrate [1].

In this study, the spatial uniformity of the key properties of sputtered AZO films was studied as a function of the discharge type (DC, pulsed-DC, and RF), and sputtering parameters. A ceramic target containing 2 wt % of Al was used for all experiments. Usually, these kind of depositions are performed using rotation of the substrate to seek uniformity, in this study however, the rotation of the substrate was stopped for a better mechanism growth understanding, obtaining a thickness gradient.

Both pulsed-DC and RF discharges produced films possessing high electrical conductivity ($4 \times 10^{-4} \Omega \cdot \text{cm}$) and an average optical transmittance above 85% when the substrate temperature was above 100°C. All films deposited at room temperature presented lower electrical conductivity and lower transmittance. A DC discharge led to inferior film properties for TCO applications.

Films deposited at room temperature and using Pulsed-DC, and DC discharges showed an edge effect with an increase of the sheet resistance in the wafer region that is closer to the target, therefore, low distance from target to substrate (DTS). This undesirable characteristic was improved once the temperature of the substrate increased. Films deposited using RF discharge showed the best performance regarding optical and electrical properties.

[1] – K. Norrman, P. Norby and E. Stamate, *J. Mater. Chem. C* 10 (2022) 14353.

CP-ThP-4 Optical and Electrical Characterization of Thin NiO_x Films Obtained by R.F. Sputtering, *Francisco David Mateos-Anzaldo, R. Nedev, E. Osorio-Urquiza, M. Curiel-Alvarez, O. Perez-Landeros, J. Castillo-Saenz*, Universidad Autónoma de Baja California, Instituto de Ingeniería, Mexico; *A. Arias-Leon*, Universidad Autónoma de Baja California, Facultad de Ingeniería Mexicali, Mexico; *B. Valdez-Salas*, Universidad Autónoma de Baja California, Instituto de Ingeniería, Mexico; *N. Nedev*, Universidad Autónoma de Baja California, Instituto de Ingeniería, Mexico

Thin NiO_x films were deposited on crystalline Si (c-Si) and corning glass by r.f. magnetron sputtering at temperatures in the range of 25–250 °C and powers in the range of 40–80 W. After the deposition, the films were thermally annealed at 450 °C to form nanocrystals. Ellipsometry measurements were used to determine the film thicknesses and the optical constants. The size of nanocrystals in annealed layers was evaluated by XRD measurements, while the surface roughness of the as-deposited and annealed films was measured by AFM. Metal/NiO_x/n-Si heterostructures were prepared by deposition of thin NiO_x layers on n-type Si and evaporation of Au electrodes through a mask. The Au/NiO_x/c-Si structures were electrically characterized by current-voltage (I-V) and capacitance-voltage (C-V) measurements. The I-V dependences showed formation p-n heterojunction diodes with properties, which depend on the r.f. power, deposition temperature and annealing. The obtained results indicate that NiO_x films deposited at optimal conditions are promising for application in optical sensors.

CP-ThP-5 Deposition of Lanthanum-Doped Barium Stannate as Transparent Conducting Oxides, *C. Liu, Y. Yan, S. Chen, Yijia Chen, M. Wong*, National Dong Hwa University, Taiwan

The barium stannate (BaSnO₃, BSO) crystal has a perovskite structure, so as the lanthanum-doped barium stannate (LBSO). The LBSO thin film has high light transmittance and high electron mobility, and is considered potential candidate to replace the indium tin oxide (ITO) as conductive glass used for optoelectronics, because the indium in ITO is increasingly depleted in mineral resources and becomes ever expensive. We use the lanthanum metal target and the barium stannate target in the magnetron sputtering deposition system. The barium stannate is sputtered with 80W with radio frequency and lanthanum is sputtered with 10W DC. In the coated lanthanum-doped barium stannate film, a metal thin layer of pure lanthanum is intermittently deposited in between BSO layers to further improve the conductivity of the film. The transmittance of the coating shows periodical oscillation with the wavelength, which is typical of the consequence of interference phenomenon, signifying the flatness of the deposited film. After annealing at 600°C, crystallization occurs, as the x-ray diffraction analysis suggested. The concentration of La doping in the coating is about 10% of that of barium. According to Hall's measurement results, the multi-layered LBSO coating with inserted La metal layer has a carrier concentration of $1.3 \times 10^{22}/\text{cm}^3$ and a mobility of 177 $\text{cm}^2/\text{V s}$.

CP-ThP-6 Electrical Evaluation of Micro Water Droplets During Solidification Process Using Galvanic Array with Micro to Nano Gaps, *K. Hirayama*, Chiba Institute of Technology, Japan; *M. Mekawy, J. Kawakita*, NIMS, Japan; *Y. Sakamoto*, Chiba Institute of Technology, Japan

Frost damage can be classified into two types: white frost, which occurs when water vapor sublimates on the surface of an object to form ice, and water frost, which occurs when water droplets condense and solidify. Currently, there are no excellent sensors that can detect frost, and frost can only be observed visually, which makes field observations difficult and makes the actual state of frost damage unresolved. The presenters have developed a sensor that can detect water and adsorbed water molecules by measuring the electric current that flows spontaneously due to galvanic action when an aggregate of water or adsorbed water molecules comes into contact with adjacent arrays of alternating thin wires made of different metals at regular intervals (minimum value 100 nm) on an insulating substrate. Previous results have shown that the current peaks appear before and after the water undergoes a supercooled state and solidifies, indicating that frost formation detection may be possible, and furthermore, the current peaks and microscopic solidification process have been clarified. As a response to water solidification, the current response is decreasing despite the growth of water droplets. The authors believe that the temperature dependence of the conductivity of water is responsible for the decrease in the current response. However, although the temperature dependence of conductivity in water has been clarified, the temperature dependence of conductivity in the supercooled state of water has not been clarified.

In this study, we aim to clarify the temperature dependence of conductivity in supercooled water by cooling the sensor surface temperature step by step from 0°C and by clarifying the sensor response behavior.

CP-ThP-7 Engineered Ionic Diode Membranes Based on Subnanochannel Metal-Organic Frameworks with High Space Charges for Boosted Lithium Ion Transport and Unprecedented Osmotic Energy Conversion in Organic Solution, Amalia Rizki Fauziah, L. Yeh, National Taiwan University of Science and Technology, Taiwan

Harvesting the Gibbs free energy contained in the salinity gradient of waste organic solutions will not only relieve the environmental burden but also render a new clean energy resource to accomplish the never-ending energy demand. Taking inspiration from the electrocytes in the bioelectricity systems (e.g. electrical eel) which possess numerous subnanoscale rectified ion channels acting as an ion-selective filter allowing unidirectional ion transport, we sought a feasible strategy to design an ionic-diode membrane, ZIF-8/PSS@ANM, consisted of a continuous layer of the subnanochannel zeolitic imidazolate framework-8 (ZIF-8)/polystyrene sulfonate (PSS) and a highly ordered aluminum nanochannel membrane (ANM), as osmotic energy conversion generator. The SEM results indicate that a large-scale, continuous, defect-free ZIF-8/PSS membrane was successfully prepared. Furthermore, the BET result verifies that the as-synthesized MOF membrane possesses subnanoscale (~4.1 Å) channel windows with a high surface area (~1290 m²/g). The as-developed ZIF-8/PSS@ANM demonstrates a vivid diode-like ion current rectification effect even in methanol solutions with a ratio as high as ~5.57 in 1 mM LiCl (Fig. 1a), enabling the ion transport magnification at the subnanoscale confinement, due to the multiple broken symmetries (Fig. 1b). We, thereby, probe the application of this subnanoscale ionic-diode membrane in osmotic energy conversion, and exceptionally, an unprecedented osmotic power density of ~5.28 W/m² at 50-fold LiCl gradient in methanol was achieved (Fig. 2a), exceeding the bandgap of commercial benchmark value. The unbelievable osmotic power achieved can be plausibly elucidated to be associated with the space charges carried by PSS enhancing the ionic selectivity and accelerating ion migration, abundant ordered subnanoscale window-cavity channels of the ZIF-8 for screening dehydrated cations (Fig. 2b), and ionic-diode effect for amplifying the generated ionic current. The heterogeneous subnanochannel MOF membrane we designed will likely ignite valuable insight not only to help alleviate the environmental burden but also to open up a new avenue towards a new energy platform for meeting the need of the ever-growing energy demand.

CP-ThP-8 Designing Experimental Determination of Sheet Resistance of a Titanium Self-Aligned Silicide Formation, Jau-Shiung Fang, Y. Chang, Y. Kuo, National Formosa University, Taiwan

In recent years, metal silicides have been widely used in ultra-large scale integrated-circuit (ULSI). Shallow junction generation has always been a major challenge with ever-shrinking device dimensions, and size effects must be overcome for silicide generation, thermal stability, and electrical properties. Due to the size effects, evolution of metal silicides from TiSi₂, CoSi₂, to NiSi has been used as the source material for the formation of a low-resistivity form of metal silicide on top of the gate and source/drain for connecting the tungsten contact plug. However, characteristics of TiSi₂ have led to it having the potential to be a plug/interconnect material in nanoscale devices.

Because the TiSi₂ used in logic device is normally fabricated using a self-alignment silicide process, the process includes Ti/TiN deposition, first-step rapid thermal annealing (RTP-1), strip of unreacted Ti/TiN, second-step rapid thermal annealing (RTP-2), and final-step annealing for smoothing the top-cap dielectric. The process needs to be optimized using a design of experiment method. The influence of arsenic doping dosage, the thickness of titanium, the temperature of rapid thermal annealing on the sheet resistance of a polysilicon gate was experimentally analyzed. Experimental results revealed that thickness of titanium, the temperature of RTP-2, and the interaction between the thickness of titanium and the temperature of RTP-2 dominated the sheet resistance of TiSi₂. An optimum RTP-2 temperature was also required for reducing sheet resistance of TiSi₂. A low sheet resistance was yielded for titanium thickness = 32-35 nm, RTP-1 = 720-750°C for 75 sec, and RTP-2 = 860°C for 20 sec. Heavily doping of the polysilicon gate with arsenic suppressed the formation of C54-TiSi₂. The lowest sheet resistance of 3.91 Ω/sq. was obtained with an arsenic dosage of 1 × 10¹⁴ /cm². The characteristics of the TiSi₂ supports its capability as a contact material for next generation devices.

Keywords: Titanium self-aligned silicide, TiSi₂, Sheet resistance, Designing experimental

CP-ThP-9 Hybrid Structures of p-n junction for Improving Efficiency of Photovoltaic Devices, Paweł Jarka, T. Tański, Department of Engineering Materials and Biomaterials, Faculty of Mechanical Engineering, Silesian University of Technology, Poland; B. Hajduk, H. Bednarski, Centre of Polymer and Carbon Materials, Polish Academy of Sciences, Poland

The article describes the study of electrical properties investigations of solar cells based on organic p-n junction. Organic layer systems will be produced using spin-coating method with starting material in form of mixture of polymeric materials: Poly[2,6-(4,4-bis-(2-ethylhexyl)-4Hcyclopenta[2,1-b;3,4-b']dithiophene)-alt-4,7(2,1,3-benzothiadiazole)] - PCPDDTB and poly[2,5-bis(2-octyldodecyl)pyrrolo[3,4-c]pyrrole-1,4(2H,5H)-dione-3,6-diyl]-alt-(2,2';5',2'';5'',2'''-quaterthiophen-5,5''-diyl)] - DPP4T. The p-n heterojunction materials were selected on the basis of gap matching component acting as an electron donor to a wavelength of 700 nm, corresponding to the maximum photon density in the spectrum of sunlight (about 1.8 eV) to the acceptor material. Acceptor material was selected on the basis of the band gap width and the electron affinity energy ionization energies suitably greater than the electron affinity of the excited state of the donor, and ionization potential, ensuring dissociation of the exciton and transfer of the electron from the LUMO orbital of the donor into the LUMO orbital of the acceptor. The investigations presented in article included the production of organic solar cells (OSC) with bulk heterojunction (BHJ) and determination of the structure and morphology of the deposition layers, chemical composition analyzes, optical and electrical properties of the BHJ thin films and I-V characteristics of created OSC using PV Test Solar Cell I-V Tracer System and Keithley 2410 source meter under Standard Test Conditions (AM 1.5, 100 W/m²). The conducted basic research brings knowledge of controlling the structure and properties of the thin films of the semiconducting organic material (containing bulk heterojunctions). Analysis of the results of electrical properties testing will allow for a thorough examination mechanisms of electronic transitions, electron-electron and electron-phonon interactions in p-n heterojunctions combining organic materials.

CP-ThP-10 The Investigation of Electro-Optical Properties of Hybrid Organic-Inorganic Thin Films, Tomasz Tański, Department of Engineering Materials and Biomaterials, Faculty of Mechanical Engineering, Silesian University of Technology, Poland

The aim of research is to present influence of phase composition and manufacturing parameters on structure and electro-optical properties and surface morphology of hybrid nanocomposite thin films. The hybrid organic-inorganic material constitute thin layer of a nanocomposite with a polymer semiconductor matrix and reinforcement of semiconductor nanoparticles. The DPP4T polymer, Poly [2,5-bis (2-octyldodecyl) pyrrolo [3,4-c] pyrrole-1,4 (2H, 5H) -dione -3,6-diyl] -alt- was chosen as the matrix material (2,2';5',2'';5'',2'''-quaterthiophen-5,5''-diyl)] due to its narrow optical band gap and high charge-carrier mobility, as reinforcement were selected TiO₂ and ZnO nanoparticles (NPs) with a wider band gap, however ensuring high electrical stability of the inorganic semiconductor - polymer system. Such a hybrid combination of materials has a very perspective application in electronics, with particular emphasis on optoelectronics and photovoltaics. Thin films of DPP4T / NPs were deposited using spin-coating simple and fast method from solution. The spin-coating processes were carried out with the use of variable deposition conditions, from the point of view of the starting material and using different technological conditions.

In order to identify the structure (with particular emphasis on arrangement of the reinforcing phase) and surface morphology of thin films the Scanning Electron Microscope (SEM) and Atomic Force Microscope (AFM) were used. Chemical composition analysis was performed with use X-ray powder diffraction (XRD), energy-dispersive X-ray spectroscopy (EDS) and Fourier-Transform Infrared Spectroscopy (FTIR)

Opto-electrical properties have been performed by UV-Vis absorption spectroscopy and ellipsometry. Based on the analysis of the measurement results of produced nanomaterials, the energy band gap (E_g) for the materials, the refractive index, the extinction coefficient, the real and imaginary part of the dielectric constant will be determined. The equivalent electric model of the thin films was determined by impedance spectroscopy (EIS).

The results of the work indicate that the application of the developed thin layers may be a promising solution in optoelectronics and photovoltaics (especially in thin-film heterojunction systems) due to the optical and

electronic properties obtained, as well as the speed and simplicity of application.

CP-ThP-11 Multilayer Growth of 2D Layered Material Bi₂Se₃ Through Heteroatom-Assisted Step-Edge Barrier Reduction, Namdong Kim, Pohang Accelerator Laboratory, Republic of Korea

Various two-dimensional (2D) van der Waals systems including graphene, hBN, MoS₂, WS₂, and topological insulators form heterostructures with the high quality in atomic-layer scale. Understanding the growth kinetics of the layered heterostructure films is essential to control the atomic layer growth. We studied the growth kinetics of Bi₂Se₃ film on graphene by using AFM images and DFT calculations as well as by in-situ x-ray scattering.

During growth of 2D materials, abrupt growth of multilayers is practically unavoidable even under well-control. Delicate control of growth reaches its limits for complicated crystal structure. In epitaxial growth of Bi₂Se₃ thin film, we observe that the multilayer growth pattern deduced from in-situ x-ray diffraction requires nontrivial interlayer diffusion process. We expect that an intriguing diffusion process occurs at step edges where a slowly downward-diffusing Se adatom having a high step-edge barrier interacts with a Bi adatom pre-existing at step edges. The Se–Bi interaction lowers the high step-edge barrier of Se adatoms. This drastic reduction of the overall step-edge barrier and hence increased interlayer diffusion modifies the overall growth significantly. Thus, a step-edge-barrier reduction mechanism assisted by hetero adatom-adatom interaction could be widely utilized for multilayer growth of 2D heteroatomic materials.

KEYWORDS: heteroatom epitaxial growth, kinetic multilayer growth model, step-edge barrier

CP-ThP-12 Metallic Ground States of Strained Ti₂O₃ Thin Films, Heungsoo Kim, S. Mathews, E. Lock, J. Prestigiaco, Naval Research Laboratory, USA; M. Qazilbash, William and Mary University, USA; A. Piqué, Naval Research Laboratory, USA

Single crystal Ti₂O₃ with a trigonal corundum structure exhibits a metal to insulator transition (MIT) between 400K and 500K without a structural phase transition. Upon cooling Ti₂O₃ undergoes a transition from metallic state to a nonmagnetic insulating state showing a ultranarrow bandgap (~0.1eV). Compared to other MIT oxides such as V₂O₃ and VO₂ that undergoes structural phase transition during MIT, the Ti₂O₃ shows pure electronic MIT process without having a structural transition. This purely electronic MIT is unique and would be useful for many electronic and photonic devices. We have deposited epitaxial corundum structured Ti₂O₃ films on c-plane sapphire substrates using pulsed laser deposition and investigated their structural, electrical, and optical properties as a function of the film growth parameters. We have found that a MIT temperature is varied with a growth temperature and the MIT is suppressed when the films are grown at 480 °C, showing conducting behavior at all temperatures. This metallic ground states were further investigated by X-ray diffraction and spectroscopic ellipsometry measurements to provide crystal structure and broadband optical properties of Ti₂O₃ films. Results show that the electrical properties are governed by the lattice parameter ratio(c/a) of crystal structure and the imposed strain causes an increase in the c-axis length as the temperature is decreased, and thereby suppresses the MIT. We will present details of the deposition conditions on the structural, electronic, and optical properties of Ti₂O₃ films.

This work was supported by the Office of Naval Research (ONR) through the Naval Research Laboratory basic research program.

CP-ThP-14 Rapid Thermal Annealing and Structural Evolution of Sputter-Deposited AlScN Thin Films, Hongfei Liu, A. Yong, N. Gong, R. Karyappa, T. Meng, Institute of Materials Research and Engineering (IMRE), Singapore

The excellent chemical and mechanical stability, the excellent piezoelectrical properties, as well as the semiconductor compatibility, not only in thin film deposition but also in device processing, of AlN made it widely used in various piezoelectric applications. Recent studies revealed that incorporation of Sc with proper composition to substitutionally replacing Al in the lattice of AlN could dramatically increase its piezoelectric coefficient and electromechanical coupling. The presence of polarity inversion domains in III-nitride could negatively affect the piezoelectric coefficient. In fact, polar-controlled growth has long been studied in III-nitride thin films and heterostructures, e.g., to enhance carrier injections in GaInN/GaN multiple quantum well structures. Post-growth thermal annealing has also been found to reduce the density of polarity inversion domains in III-nitride thin films.

In this work, we have studied the effect of rapid thermal annealing (at T_{ann} = 600-900 °C) on surface chemical and structural evolutions of fiber-

textured AlScN thin films deposited on Si (111) substrate by magnetron-sputtering. The film thickness was controlled at 1.0 μm and the Sc composition was controlled at about 15%. By varying the annealing time t_{ann} in periodic cycles from 3 to 48 min, we found that the lattice constant along the (0001) direction tends to be increased, along with a peak splitting of the X-ray diffraction (XRD) peak around the (0002) atomic planes. In this post, we will be presenting the experimental results.

CP-ThP-15 Work Function Enhancement of WO₃ Filamentous Films Obtained by Resistive Heating Evaporation Technique, Fabien Sanchez, L. Marot, R. Antunes, R. Steiner, E. Meyer, University of Basel, Switzerland

Tungsten trioxide (WO₃) films are of great interest due to their electronic properties, which can be tuned by surface nanostructuring leading to enhanced efficiency for gas sensing, energy storage or electrochromic applications. Resistive heating of tungsten (W) filaments at pressures of few Pa in an oxygen O₂ atmosphere has already demonstrated its capability to form porous, micro/nano-structured, cheap and fast films making it suitable for industrial applications.

In this work, stoichiometric WO₃ films were produced by applying a current into a W filament in an O₂ atmosphere. The pressures were varied from 2 to 20 Pa. The increase of the pressure above 7.5 Pa led to amorphous WO₃ films with a filamentous morphology. As a function of the pressure, the film morphology and the work functions (W_F) were analyzed using Scanning Electron Microscopy (SEM) and in-situ Ultraviolet Photoelectron Spectroscopy (UPS). In addition to the high surface-to-volume ratio of the films, the W_F exhibited an increase from 5.8 eV, for a conventional WO₃ film, to a maximum of 8.7 eV at 20 Pa. This change corresponds to an increase of the W_F of about 50 %, making our films suitable for a large variety of applications.

CP-ThP-16 Impacts of Phase Interactions on Photocatalytic Hydrogen Evolution Reactions, Kun Ta Lin, J. Ruan, National Cheng Kung University (NCKU), Taiwan

In order to preserve nature environment from being further polluted by on-going human civilization development, pursuing affordable alternative green energy has emerged as global endeavor nowadays. Regarding the production of green hydrogen energy, the strategies able to facilitate water splitting reactions have been intensely studied. In recent research, piezoelectric materials have been widely incorporated with photocatalysts since the induced electric fields upon applied mechanical stress have been considered able to facilitate carrier generation, separation, transport, and recombination. However, the responsible mechanism has not been truly understood.

In this research work, instead of inorganic piezoelectric materials, evolution, stacking and assembly of PVDF-TrFE ferroelectric lamellar crystals have been systematically investigated as an approach to modify involved phase interactions and contribution of local electric fields around ferroelectric crystals. The feasibility of annealing-induced coalescence of PVDF-TrFE ferroelectric lamellar crystals has been unambiguously identified, which is critically subject to domain roughness, and able to significantly modify reachable crystal dimensions. As the polarization axis is along the b-axis of lattice packing, the thickness of lamellar crystals is surprisingly capable to serve as a critical factor for reachable magnitudes of crystal polarity and thus the strength of created local electric fields. Also, dissimilar to molecular interactions, phase interactions are subject to the number of involved interactions crystals, in addition to separation distance. Based on this collective feature, crystal engineering able to modify the evolution and dispersion of interacting crystals are capable to modify available phase interactions. With the absence of external forces, evolved monolayers of PVDF-TrFE ferroelectric/piezoelectric crystals are measured able to significantly promote the capability of P3HT/MoS₂ composite layers to catalyze hydrogen evolution reactions, which is also dependent on reached magnitude of crystal polarity. Upon Mott-Schottky plot and estimated binding energy of exciton, the electric fields of ferroelectric crystals not only lower the binding energy of photo-induced excitons and increase the concentration of charge carriers, but also modify band bending at solid/liquid interfaces. As a matter of fact, the Schottky contact at solid/liquid interfaces has been evaluated to switch to ohmic contact upon the presence of PVDF-TrFE ferroelectric lamellar crystals for the first time, which further unveils possible impacts of phase interactions.

CP-ThP-17 Controlled Thermal Conduction-based Detection of Dew Condensation on Target Solid Surface by Galvanic Arrays Sensor Chip, K. Iida, Chiba Institute of Technology, Japan; **M. Mekawy, N. Satoh, J. Kawakita,** NIMS (National Institute for Materials Science), Japan; **Y. Sakamoto,** Chiba Institute of Technology, Japan

Reliable early detection of dew condensation is considered a bottleneck in surface protection against numerous negative effects such as surface fogging and corrosion. To detect the early stage of dew condensation, we fabricated a thin film-based sensor chip composed of a confined silica surface between two adjacent interdigitated gold and aluminum metal arrays that are arranged alternately at regular intervals varied between 0.5 to 10 mm. Whenever a tiny droplet is bridging between these arrays, a passage of galvanic current could be reliably detected. Imitating the surface condition for dew condensation was carried out in a temperature-controlled scheme employing thermal conduction heat transfer between the solid surfaces of the sensor chip and its contacting heat conductor. However, the effect of the geometrical shape of the heat conductor on the thermal heat conduction rate is yet to be emphasized. In this study, three different fabricated geometrical shapes of aluminum heat conductors (Fig. 1(a)) were attached to the back side of sensor chip and used for a step-wise (static) or direct (dynamic) temperature-cooling heat transfer mechanisms (Fig. 1(b)). The experimental results revealed that the sensor response current (as a measure of dew condensation detection) increased when the sensor surface temperature was dropped below the dew point. Moreover, the rate of thermal conduction was larger at direct temperature-cooling mechanism than at step-wise mechanism as shown in Fig.2. In addition, the temperature difference between the sensor surface and each heat conductor was found to follow the order of $DT_I > DT_{II} > DT_{III}$. Furthermore, the rate of thermal conduction between the thermally contacted surfaces of sensor and each heat conductor was found to follow the order of $I < II < III$. This could be attributed to the thermal resistivity of each examined heat conductor which followed the order of $R_I > R_{II} > R_{III}$ (listed in table 1). The results were in an agreement with further simulation-based investigations that were also performed to correlate the geometrical shape of contacting heat conductor with its heat transfer to the sensor surface. These results demonstrated that controlling the temperature of the sensor surface depends on the geometrical shape and the temperature change of its contacting heat conductor. Therefore, it can be concluded that our developed sensor can be beneficially used for the enhanced early detection of dew condensation at the solid substrate surfaces of interest employing the thermal conduction heat transfer mechanism.

CP-ThP-18 Polyimide-Based Gate Dielectrics for High-Performance Organic Thin Film Transistors, Yan-Ting Chen, Y. Yu, Ming Chi University of Technology, Taiwan

In this study, TiO₂-SiO₂ nanoparticles with OH group on their surface will be prepared by sol-gel process from TEOS and titanium ethoxide. The particle size and morphology, crystal phase, crystallinity, and the corresponding dielectric constants are investigated. The prepared ST colloids will further react with the fluorine-containing soluble polyimide with side OH group chains to form the PI/ST hybrid thick films on glass and flexible plastic substrate. The prepared high dielectric PI/ST films will be applied to fabricate the high capacitance memory devices with structure Al/PI-ST/ITO-Glass or Al/PI-ST/ITO-PET. The PI/BT films will further be applied to fabricate the OFETs with structure Al/PffBT4T-2OD/PI-ST/Si on the silicon substrate with the PI/ST hybrid film as the gate insulator. We have systematically investigated the effects of properties of PI/ST films such as dielectric constant, surface roughness, and thickness on capacitance, field-effect charge mobility, on/off ratio, threshold voltage, and leakage current.

CP-ThP-19 Epitaxially Grown Gold (100) Surfaces for Oxygen Reduction Reactions, Katharina Kohlmann, D. Guay, Institut national de la recherche scientifique, Canada; **A. Sarkissian,** Plasmionique Inc., Canada; **C. Schindler,** Munich University of Applied Sciences, Germany; **A. Rüdiger,** Institut national de la recherche scientifique, Canada

Noble metals have long been known to be an excellent basis for electrocatalysts. While the effectivity of catalysts depends on the reaction they are used for, studies have shown that for the oxygen reduction reaction (ORR), Au (100) is the most active face of Au in alkaline media. This work investigates magnetron sputtered epitaxial Au-films on MgO (100) for electrocatalysis. We show that the deposition parameters and their effect on the surface morphology are a key factor to optimize catalytic activity. We further explore various surface treatment methods to improve the adhesion of Au as well as its surface morphology without the use of a transition metal seed layer. The samples are characterized by atomic force

microscopy, X-ray diffraction and cyclic voltammetry to establish a correlation between the surface topography and electrocatalytic activity.

CP-ThP-20 Ion-Selective Capacitive Deionization of Saltwater Using Functionalized Graphene Thin-Film Coated Electrodes, H. Cheng, National Cheng Kung University, Taiwan; **J. Wang,** Stanford University, USA; **Hong Paul Wang,** National Cheng Kung University, Taiwan

Drinking water shortage is getting worse in recent decades. Desalination of saltwater by capacitive deionization (CDI) with the advantages of relatively low energy consumption and environmental friendly is of increasing importance. To improve the desalination performances, by introducing ion exchange membranes (IEM) on the surfaces of CDI electrodes for weakening co-ion repulsion effect, membrane CDI (MCDI) cell architectures have been constructed. Nevertheless, IEM may suffer from high cost and interfacial resistance. It would be economically attractive to use IEM for selectively moving relatively ions to electrodes for better desalination performances and higher feed rates. Thus, in the present work, sulfonated- and poly(diallyldimethylammonium chloride)-functionalized graphene oxide (SGO and PGO) serves as hydrophilic cation- and anion-exchange membrane (CEM and AEM), respectively to enhance CDI efficiencies. The positively charged PGO thin-film coated on the activated carbon (AC) can selectively transport anions to positive electrodes in the CDI process. The SGO and PGO coated AC electrode pair (AC/SGO || PGO/AC) for CDI of saltwater ([NaCl]=200-500 ppm) under +1.2 V for 1 h reaches a high optimized salt removal (200 mg/g-day) and electrosorption capacity (9 mg/g). In the reverse voltage desorption operation mode, effective desorption of anions for regeneration with the thin-films can also be achieved. This work presents the feasibility using the high-efficiency, low-cost and facile SGO and PGO ion-selective thin-film coated on AC electrodes to enhance desalination performances.

CP-ThP-21 Research on the Application of the Double-layer Hole Transport Layer of Novel Functional Organic Small Molecule Materials in High-efficiency Inverted-Perovskite Solar Cells, Wei-En Wu, Y. Yu, Ming Chi University of Technology, Taiwan

This research is divided into two parts. Both use the hydrophobic small molecule as a double-layer hole transport material provided by Professor Yung-Chung Chen from Kaohsiung University of Science and Technology. In the first part, we investigate the effects of the three p-type small molecules (CL-1~CL3) with tetraphenylethylene as the core and different aromatic rings attached to the side chain. The side chains are benzene, naphthalene, and pyrene. The tetraphenylethylene core has highly distorted nature, even without alkyl solubilizing groups, it can still have good solubility, and then through the modification of side chain groups, the energy level and hole mobility can be fine-tuned. Under the condition of AM1.5, NiOx/CL-3 double-layer hole transport layer has the best power conversion efficiency of 20.15% in the trans-structured perovskite solar cell.

CP-ThP-22 High-Performance non-Fullerene Systems for Organic Solar Cells, Chun-Chieh Lee, Y. Yu, Ming Chi University of Technology, Taiwan

For the organic photovoltaics (OPVs), the choice of solvent affects the morphology of the active layer blend as well as the device performance and potential commercial applications. In this study, two different solvents, chloroform (CF) and chlorobenzene (CB) with optimal process parameters, were applied to prepare the OPVs with the PM6:BTP-eC9 as the active layer. Atomic force microscopy and grazing-incidence wide-angle X-ray scattering were used to evaluate the blend morphologies of the OPVs, and also examined the optoelectronic properties of the blend films and devices. The power conversion efficiencies could reach up to 17.82% when using CB as the solvent, without any additives. Compared with the CF-based device, the optimized CB-derived OPV exhibited a more suitable phase-segregated domain size with stronger face-on molecular stacking, leading to more efficient carrier transport. Thus, by optimizing the fabrication conditions and selecting a suitable solvent that could improve the structure of the PM6:BTP-eC9 blend films and thus also improve the OPV performance.

CP-ThP-23 Vanadium Doped ZnO Nanorod Array Piezoelectric Pressure Sensor, Shu-Yu Lin, J. Huang, S. Brahma, National Cheng Kung University (NCKU), Taiwan

ZnO has semi-conductivity and piezoelectricity at the same time that makes it a promising material for piezotronics. Zinc Oxide (ZnO) nanorod array was grown on silicon substrates by a hydrothermal method. From SEM top-view image, well aligned ZnO nanorods were deposited on the silicon substrate. The XRD patterns showed that the nanorods behaved highly (002) oriented. Resonant Raman spectroscopy revealed that the degree of (002) orientation was decreasing with raising the vanadium concentration

Thursday Afternoon, May 25, 2023

of growth solution. The Photoluminescence spectrum showed typical ZnO UV emission and the 6% sample has an obvious red shift.

Functional Thin Films and Surfaces Room Pacific F-G - Session C2-2-FrM

Thin Films for Electronic Devices II

Moderators: Julien Keraudy, Oerlikon Balzers, Oerlikon Surface Solution AG, Liechtenstein, Jörg Patscheider, Evatec AG, Switzerland

8:00am **C2-2-FrM-1 3D Device Integration Technology for AI Computing**, S. Chang, Powerchip Semiconductor, Taiwan; **Shou-Zen Chang**, Powerchip Semiconductor Manufacturing Corporation, Taiwan **INVITED**

To cope with the massive computation requirement of semiconductor chips in 5G and AI, 3D device integration incorporating with new system architectures is developed, to mitigate the computational von Neumann memory wall via minimized data movement. Both the 3D Memory-Logic chip stacking and monolithic SoC integration are developed for near-memory and in-memory analog computing, to perform high bandwidth and efficient power performance, leading AI computing toward a green technology era.

3D Memory-Logic Chip Stacking

We focus on wafer-on-wafer Memory-Logic 3D stack technologies, including (1). Oxide bond with paired TSV (Through Silicon Via), (2). Hybrid bond with TSV middle embedded in DRAM, and (3). Multi-DRAMs stack on Logic. The interconnect density is among $1E4$ - $1E6/mm^2$ range.

For intensive data-movement computation applications, hybrid bond with 2 - $3\mu m$ Cu bond pitch is developed to provide $1E6/mm^2$ high interconnect density and $< 0.5\Omega$ low interconnect (Via-pad-bond-pad-Via) resistance. After implementing dispersive DRAM I/O and skipping interface PHY circuit in the system design, 55nm-node Logic chip and 38nm-node customized 6GB DRAM chip are bonded together to demonstrate 3-9X excellence in system performance, better than the products using advanced Logic chip while choosing commercial DRAM with standard I/O interface.

For AI image recognition applications, oxide bond with paired TSV is implemented for Logic-DRAM stacking, which a $10\mu m/15\mu m$ dual TSV with $20\mu m$ pitch is used for interconnection, providing $< 0.2\Omega$ (pad-TSV-Cu line-TSV-pad) resistance and $1E4/mm^2$ interconnect density. The AI inference (# of image recognition/s) performance is expected to improve $> 50\%$, as the I/O bandwidth enlarges 8X to 51.2GB/s. These 3D schemes can minimize energy consumption to $< 1pJ/bit$ during data movement.

3D Monolithic SoC for Analog in-Memory Computing (AiMC)

AiMC eliminates data movement during computation, offering low-power multiply-accumulate (MAC) operations in AI computing. To improve the poor power consumption and worse temperature variation reported in some emerging memory AiMC, monolithic 3D Si/CAAC (c-axis-aligned crystalline)-IGZO 3T1C AiMC chips with ultra-low operation cell currents (< 1 nA/cell), multiple analog states (8 and 64), and high computing efficiency (143-210 TOPS/W) are demonstrated [1,2]. We further implement Ferroelectric material into the MIM capacitor to keep the MNIST inference accuracy $> 90\%$ even at $125^\circ C$ high temperature operation, while retention time > 50 hrs. We believe it is a good candidate for the next-gen AI computing.

Ref. [1,2] IEDM 2021/2022

8:40am **C2-2-FrM-3 Magnetic Nanolaminates Deposited by Magnetron Sputtering for Next Generation Electronic Devices**, Claudiu V. Falub, M. Bless, J. Richter, X. Zhao, H. Rohrmann, M. Tschirky, M. Padrun, Evatec AG, Switzerland

The emerging Internet-of-Things (IoT) and artificial intelligence (AI) applications, combined with the ongoing miniaturization of mobile devices, has led to an increasing demand for heterogeneous systems that need to be integrated with the silicon complementary metal-oxide-semiconductor (Si-CMOS) platform, such as smart power management units based on integrated voltage regulators (IVR) and ultrahigh density non-volatile memory (NVM) cells. Besides numerous challenging surface engineering processes and corresponding equipment concepts for high volume manufacturing, the realization of these 3D monolithic architectures often requires advanced magnetic thin-film materials with precisely designed magnetic properties and thickness ranging from just a few tenths of nanometers up to several micrometers. In this talk we will illustrate some of these challenges using two examples where nanostructured magnetic thin film systems enable the requested functions for new generation electronic devices: (i) soft magnetic nanolaminates $n \times [m \times (\text{FeCoB}/\text{CoTaZr})/\text{Al}_2\text{O}_3]$ with tunable in-plane magnetic anisotropy that form the inductor cores for high-frequency RF filters and on-chip IVR; (ii) enhanced permeability dielectrics $n \times (\text{FeCo}/\text{Al}_2\text{O}_3)$ for single-layer Magnetoresistive Random Access Memory (MRAM) and programmable logic systems. These novel composite magnetic multilayers can be economically deposited on Si-CMOS wafers up to 300 mm in diameter using the high-throughput, multi-source, dynamic sputter systems in our portfolio, by simultaneously using two or more cathodes, and by carefully adjusting the thickness of individual nanolayers, i.e., by changing the cage rotation speed and sputter power of the individual stations [1]. Thus, due to the continuous rotation of the substrate cage, such that the substrates face different targets alternatively, the obtained thin films exhibited a nanolayered structure with very sharp interfaces in the case of soft magnetic nanolaminates [2], or a discontinuous structure with magnetic nanoparticles smaller than the superparamagnetic (SP) limit embedded in an insulating matrix [3]. We will discuss the interdependence of structure and magnetic properties in these thin films, and we will show how the latter can easily be tuned by choosing the sequence of magnetic and non-magnetic layers, and the thickness ratio of individual nanolayers.

[1] M. Bless, C.V. Falub, WO 2018/197305 A2.

[2] C.V. Falub et al., AIP Advances 8, 048002 (2018).

[3] C.V. Falub et al., AIP Advances 9, 035243 (2019).

9:00am **C2-2-FrM-4 Tungsten-Based Thin Film Metallic Glass as Diffusion Barrier between Copper and Silicon**, Pei-Yu Chen, J. You, C. Hsueh, National Taiwan University, Taiwan

A qualified diffusion barrier layer in integrated circuits (IC) industry is essential to prevent the degradation of devices due to the rapid inter-diffusion of copper (Cu) and silicon (Si) for Cu metallization. In recent years, thin film metallic glass (TFMG) barrier materials are getting considerable attentions due to the advantages over the others by its thermal stability, low resistivity and excellent mechanical properties for some compositions. In this work, the performance of amorphous W-Ni-B TFMG as a diffusion barrier between silicon and copper layers is reported. The Cu (150 nm)/W-Ni-B (10 nm)/Si multilayered structures were fabricated by direct-current (DC) magnetron sputtering and annealing at 700 - $950^\circ C$ for 30 min in vacuum. The nanomechanical properties and thermal characteristics of the W-Ni-B TFMG was evaluated by nanoindentation and differential scanning calorimeter, respectively. The interfaces and microstructures of Cu/W-Ni-B/Si multilayered structures were characterized by a field-emission gun scanning electron microscope, X-ray diffractometer and transmission electron microscope. The findings indicated that W-Ni-B TFMG showed extremely high hardness of 20 GPa and high reduced modulus of 217 GPa. In addition, W-Ni-B TFMG effectively blocked the intermixing of Cu and Si atoms at $800^\circ C$. However, the failure of W-Ni-B TFMG barrier against Cu and Si inter-diffusion was observed after annealing at $950^\circ C$. Based on its unique combination of excellent barrier performance and high hardness, W-Ni-B TFMG could be regarded as a robust diffusion barrier for Cu interconnect technology.

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9:20am **C2-2-FrM-5 Investigation of Properties and Microstructures of Ag-Cu Alloy Thin Films by Co-sputtering and First-principles Calculations**, *Yu-Chieh Wang, C. Chen, F. Ouyang, H. Chen*, National Tsing Hua University, Taiwan

As the demands of electronic devices continue to be high performance and small transistor size, the current density in electronic devices will significantly increase and electromigration (EM) is expected to be a critical reliability issue. Thus, developing a high EM-resistant interconnect will be desired in the electronic industry. Ag interconnects have drawn many eyes because they possess extreme thermal and electrical conductivity. In addition, Ag has low stacking fault energy (SFE), which is easy to form a twinned structure that exhibits unique properties, including low electrical resistivity, high EM resistance, high mechanical properties, and high thermal stability. [1] Alloying is a common way to enhance the EM resistance, but it would simultaneously change SFE with the doping elements and concentration, further impacting the formation of nanotwin structure in thin films. In this study, we first investigated how the doping concentration of Cu affects SFE of Ag-Cu alloy thin films by first-principles calculations. Their intrinsic stacking fault energy (γ_{ISF}) and unstable stacking fault energy (γ_{USF}) were calculated. In addition, the wide-angle powder X-ray diffraction was conducted to measure the SFE of pure Cu and Ag-Cu alloys. Then, Ag-Cu alloy thin films with different doped Cu concentrations were fabricated by co-sputtering system and their properties and microstructures were studied. The results show all Ag-Cu alloy thin films exhibited highly (111) orientation. The results show that low Cu concentration in Ag films exhibits a higher density of nanotwin structure with good properties than pure Ag, demonstrating that adding Cu into Ag films can effectively lower stacking fault energy and facilitate the formation of nanotwin structure.

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9:40am **C2-2-FrM-6 Multi-Step Method for the Fabrication of High-Performance Continuous Ultra-Thin Silver Films for Energy Applications**, *Phillip Rumsby, B. Baloukas, O. Zabeida, L. Martinu*, Polytechnique Montréal, Canada

While Ag films offer significant advantages over other transparent conductors, their high surface energy gives rise to a Volmer-Weber, or island growth mode. In order to achieve films which are both thin enough to be highly transparent and continuous as so to ensure their conductivity, various methods are used to suppress the Volmer-Weber growth mechanism. Typically, one will decrease Ag adatom mobility via doping, lower Ag surface energy using nitrogen as a surfactant, or increase the surface energy of the substrate.

In this work we propose a novel method for producing continuous ultra-thin silver (Ag) films in different architectures and with different microstructural characteristics which exploits rather than avoids the high surface energy of Ag. Specifically, we deposit continuous films at an arbitrary thickness, and we then etch them using RF plasma until a desired thickness and performance are achieved. In this way, the surface energy minimization constraints which drive the agglomeration of adatoms into islands during film growth now help maintain the film morphology, as re-separation would increase the Ag surface area. Thus, continuous films can be produced at thicknesses below the threshold of continuous film formation during deposition.

The formation or breakdown of continuous Ag films can then be resolved using a facile *in-situ* ellipsometry approach. After depositing a fully continuous Ag film onto a well-characterized substrate, the thickness and optical properties of the continuous Ag film are modeled. Then, using the obtained Ag optical properties and only the thickness of the layer is fitted, for each data acquisition. The mean square error (MSE) obtained on the fit then reflects divergence of the optical properties of the Ag layer from those previously modeled. Thus, as the film goes from continuous to discontinuous, plasmonic effects alter the optical properties and the MSE increases, and conversely for a discontinuous film achieving a continuous morphology during deposition, an example of which is shown in **Figure 1**.

We have validated the methods described above using *in-situ* sheet resistance measurements and cross-section transmission electron microscopy. Furthermore, we have demonstrated that this method enables the deposition of transparent, conductive, continuous thin films in atypical conditions, such as on a silica seed layer and at elevated temperatures, and can additionally be used to supplement the performance of Ag films in typical configurations. Finally, we propose additional processing steps

between deposition and etching which may further improve coating performance.

10:00am **C2-2-FrM-7 Structural, Electrical, and Thermal Properties of Ge-Rich $\text{Ge}_2\text{Sb}_2\text{Te}_5$ Alloys**, *Matias Kalaswad, A. Jarzembki, P. Kotula*, Sandia National Laboratories, USA; *T. Beechem*, Purdue University, USA; *M. King, D. Adams*, Sandia National Laboratories, USA

Materials which undergo vast and rapid transformations in optical, electrical, and thermal properties due to a phase change (e.g. amorphous to crystalline) at, or near, room temperature have long been utilized as key components in memory devices. Among these materials, germanium-antimony-telluride alloys, particularly $\text{Ge}_2\text{Sb}_2\text{Te}_5$ (GST), are especially prevalent due to their fast switching and large resistivity difference between the amorphous and crystalline states. Although chalcogenide phase change materials have been implemented in various non-volatile memories, efforts to improve phase stability and data retention in high-temperature (i.e. >100 C) environments (e.g. automotive) are still ongoing. Nearly a decade ago, a "golden composition" of a Ge-rich GST alloy exhibited a crystallization temperature of up to 250 C while still maintaining relatively fast (~80 ns) switching speed. Recent computational ab-initio studies have predicted that increasing Ge content in Ge-rich GST alloys leads to increased crystallization temperature, at the expense of phase stability in the form of segregation into regions of pure Ge and GST.

In this work, we present experimental results of structural, electrical, and thermal properties of three Ge-rich GST alloys: $\text{Ge}_2\text{Sb}_1\text{Te}_2$, $\text{Ge}_3\text{Sb}_2\text{Te}_4$, and $\text{Ge}_3\text{Sb}_1\text{Te}_2$. These are also compared to a benchmark GST film. From in-situ X-ray diffraction experiments, the Ge-rich GST alloys are observed to crystallize into cubic GST at temperatures between 250 C and 260 C, which is consistent with previous reports of similar alloys. Raman measurements generally support the XRD results, with cubic Sb-Te bonds forming around 200 C. Interestingly, Ge-Ge bonds form at 250 C, which is evidence of phase segregation as suggested by previous computational studies. In-situ measurements of the electrical resistivity show a decrease in resistivity beginning around 175 C for GST-212 and 230 C for GST-524 and GST-312, compared to 150 C for GST. The resistivity of all three Ge-rich GST alloys decrease by nearly four orders of magnitude, which is comparable to that of the benchmark GST alloy, albeit not as drastically (i.e. over a greater range of temperatures). Lastly, thermal conductivity measurements show an abrupt transition around 200 C from 0.2 W/mK to 0.7 W/mK for all three Ge-rich GST, which is nearly 50 C more than the transition temperature of GST.

10:20am **C2-2-FrM-8 Preparation and Electrical Properties of Tantalum Silicate Thin Films**, *You-Sheng Lu, C. Chen, C. Huang, S. Chen, Y. Liu*, Ming Chi University of Technology, Taiwan; *W. Huang*, Chien Hwa Coating Technology Inc., Taiwan; *W. Yang*, General Research Institute for Nonferrous Metals, China

The electric resistivity and temperature coefficient of resistivity (TCR) of a material are important parameters when developing thin-film resistors for advanced electronics and electricity consumption. There are many materials that can be applied to thin film resistors, among which the tantalum silicate (Ta-Si-O) thin film material has attracted much attention, which is due to the excellent thermal stability and adjustable TCR of this material. However, the electrical properties of such thin film materials are highly correlated with the composition control and structure of the material. How to develop an efficient process method to more precisely control the composition and structure so as to obtain the required resistivity and TCR has always been an important issue. In the past, there were very few academic reports on the manufacturing process of these materials, so the related electrical research has been very limited. In this work, we prepared two targets with different target compositions, such as $\text{Ta}_{65}(\text{SiO}_2)_{35}$ and $\text{Ta}_{80}(\text{SiO}_2)_{20}$, to study the effects of target composition and sputtering power on the material structure and electrical behavior of the film. Our series of experimental results indicate that a Ta-Si-O film with a high Ta content can be produced by using a target material with a high Ta content, and a film with low resistivity and low TCR can be successfully produced with an appropriate sputtering power. According to material analysis, the excellent electrical properties of the film are highly related to the formation of Ta_5Si_3 as the main structural phase in the film.

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10:40am **C2-2-FrM-9 Enhanced Reliability Characteristic Oftri-Gatepoly-Ge Charge Trapping Flash Memory with Ultra-Thin Tunneling Layer Engineering**, *Che-Wei Lin*, National Tsing Hua University, Taiwan; *D. Ruan*, Fuzhou University, China; *K. Chang-Liao*, National Tsing Hua University, Taiwan

In this report, a high performance FinFET based junction-less charge trapping flash memory with poly-germanium (Poly-Ge) channel was successfully fabricated. Due to the high carrier mobility of Ge material, the operation speed of flash memory device can be improved by using a low temperature Poly-Ge channel. However, the reliability of poly-Ge flash device fabricated under harsh thermal budget limitation may be degraded after inevitable thermal process. The reason may be Ge out-diffusion phenomenon and narrow energy band gap of Ge material. After stacking tunneling layer and plasma treatment, the memory device with aluminium oxynitride (AlON) tunneling layer may exhibit high programming speed, high erasing speed, long data retention time, and excellent endurance cycle. Firstly, those improvements can be attributed to better thermal stability and interface quality of AlON. Besides, the trap charge level of AlON might be much shallower than that of traditional silicon nitride trapping layer. It means that the operation speed can be further enhanced without sacrificing data retention time and endurance cycle.

Functional Thin Films and Surfaces

Room Town & Country B - Session C3-2-FrM

Thin Films and Novel Surfaces for Energy II

Moderators: *Clio Azina*, RWTH Aachen University, Germany, *Carlos Tavares*, University of Minho, Portugal

8:40am **C3-2-FrM-3 Survey for Ferroelectric/Antiferroelectric Films for Energy Storage**, *Mitsuru Itoh*, *H. Takashima*, National Institute of Advanced Industrial Science and Technology/Tokyo Institute of Technology, Japan
INVITED

Ferroelectric (FE) materials such as BaTiO₃, KNbO₃, and Pb(Zr,Ti)O₃ have dipole moments aligned to one direction. FEs have been already industrially applied as capacitor, piezoelectric actuator, filters, optical devices, and so on. However, antiferroelectric (AFE), such as PbZrO₃, AgNbO₃, and NaNbO₃, has two sublattices of the dipole moments, and each sublattice have dipole moments with opposite directions. As a whole, AFE does not have a net polarization and is structurally centrosymmetric. Under a smaller external electric field (*E*), AFE shows a linear response of electric displacement (*D*) against *E* like a paraelectric. Applying larger *E* causes flipping of dipole moments in one sublattice parallel to *E*. This constrained aligned dipole moments in one direction are comparable to a FE state that has a net polarization. However, once *E* is decreased, dipole moments in one sublattice flip to the original direction and consequently the structure returns to an original centrosymmetric and non-polar state. Utilizing such a characteristic of AFEs under *E*, a field-induced FE, they can be applied for the energy storage especially for the power devices, utilizing two step *D-E* responses of a linear one near the origin followed by a hysteresis loop at larger *E*.

Design of FE is easier compared to AFE because of the rich accumulated scientific knowledge of FE both in the structure and property during last 100 years. However, chemical design of AFE is still difficult due to the reason that the number of AFEs is limited compared to that of FEs. This study is going to try giving a comprehensive explanation on the phase stability of complex oxide materials including perovskites in the bulk and thin film including metastable state. Candidate compounds of AFE will be discussed for various phases in A₂O₃ and ABO₃.

9:20am **C3-2-FrM-5 First Attempt to Describe the Effect of the Substrate Temperature on the Depth Concentration Profile of Reactively Sputtered ZnGeN₂ Thin Films**, *A. Virfeu*, *F. Alnjiman*, *A. Borroto*, *S. Migot*, *J. Ghanbaja*, *D. Mangin*, *D. Pilloud*, *Jean-Francois Pierson*, Institut Jean Lamour - Université de Lorraine, France

The reactive sputtering process is widely used at the academic and industrial scales to deposit thin films of various nitrides, oxides, oxynitrides... For some materials, the crystallization of the film can occur even when the deposition temperature is fixed at room temperature (Cu₂O, ZnO, TiN, ...). Nevertheless, other materials require to heat the substrates during the deposition to obtain a crystallized film (VO₂, LaCoO₃, Si₃N₄, ...).

Zinc germanium nitride (ZnGeN₂) exhibit optoelectronics properties that makes this abundant material an interesting compound to replace the

indium and gallium nitrides for LEDs applications. As previously mentioned by several groups, the deposition of crystalline ZnGeN₂ using sputtering processes can only be achieved by heating the substrate during the growth.

A new approach to describe the mechanism of growth induced by the temperature during reactive co-sputtered ZnGeN₂ deposition is reported. The complex crystallization of this system requires the deposition of films at high temperature involving difficulties in controlling the stoichiometry of the films due to the evaporation of zinc atoms relative to germanium at these temperatures. The range of temperature considered in this work is between 70°C to 280°C. The study of this material is done at different scales and a model is proposed to bring answers to the observations made. X-ray/electron diffraction, transmission electron microscopy combined with in-depth SIMS analyses reveal the origin of two different zones of composition and crystallization. At a temperature above 130°C, the first step of growth is described by an under-stoichiometry of Zn entailing an amorphous phase. The second zone appears after an increase of the zinc content makes it possible to obtain a stoichiometric crystallized material. Hypotheses are announced, supported by calculations of sputtered zinc atom energy and zinc vaporization temperature in the present conditions of growth. The model provides a good agreement with the observation of the difference of composition for films deposited at temperatures above 130°C in our sputtering process. In this paper, an original work on the phenomena taking place at different stages of the growth of ZnGeN₂ thin films is described and gives new elements of understanding to the previous studies in the literature.

9:40am **C3-2-FrM-6 Effect of Oxygen Flow Rate on the Hydrogenation Resistance of ZrN_xO_y Thin Films on Zircaloy-4**, *Yen-Ting Chen*, *K. Lan*, National Tsing Hua University, Taiwan; *H. Tung*, Institute of Nuclear Energy Research, Taiwan

The purpose of this study was to investigate the effect of oxygen flow rate on the hydrogenation resistance of ZrN_xO_y thin films prepared by physical vapor deposition on the zirconium alloy Zircaloy-4. Hydrogen absorption of used nuclear fuel claddings made of zirconium alloys in light water nuclear power plants, which may lead to an increased risk of delayed hydrogen cracking during interim dry storage. Reducing the amount of hydrogen absorption of fuel cladding materials by the improved material processing combined with advanced surface technologies is one of crucial target of research and development in recent years. Considering about the compatibility between surface coating and zirconium alloy, the zirconium nitride film has high hardness, high corrosion resistance and wear resistance, which has been used in medical equipment, cutting and decoration industries. Recently, the positive correlation between the oxygen content and the hardness of ZrN thin film on superalloy has been exhibited. Therefore, this study aimed to investigate the relationship of the hydrogenation resistance of the different compositions of ZrN_xO_y on Zircaloy-4. ZrN_xO_y thin films were deposited on Zircaloy-4 by Hollow cathode deposition ion-plating systems (HCD-IP), the oxygen flow rate was a controlled variable in this study and varied from 0, 2, 3, to 10 sccm. Then, the cathodic hydrogen charging method was charging for sample with different flow rate. After charging, the thickness of all specimens was measured by scanning electron microscope (SEM). X-ray diffraction (XRD) was used to characterize the structure and the texture. Besides, the hydrogen content of the specimens was measured to verify the Hydrogenation Resistance of ZrN_xO_y Thin Films on Zircaloy-4.

10:00am **C3-2-FrM-7 Engineered Metal-Organic Framework-Based Heterogeneous Membranes with High Ionic Rectification for Ultra-high Osmotic Power Generation from Organic Solutions**, *Amalia Rizki Fauziah*, *L. Yeh*, National Taiwan University of Science and Technology, Taiwan

Taking inspiration from the electrocytes in the electric eel, consisting of numerous "subnanometer-scale" rectified ion channels allowing unidirectional and amplified ion transport, assorted artificial solid-state ion channel membranes have been developed over these past few years. Nevertheless, the conventional membrane designs were only limited to either one structure homogenous or two structures heterogeneous membranes. Herein, we report a feasible yet versatile strategy to fabricate the breakthrough membrane design that has never been reported before. An engineered metal-organic framework (MOF)-based ionic diode membrane (termed as ZIF-8/PSS@BANM), composed of a continuous zeolitic imidazolate framework-8 (ZIF-8) membrane incorporated with negatively space-charged polystyrene sulfonate (PSS) and branch-type alumina nanochannel membrane (BANM), was successfully developed (Fig. 1). Results obtained confirm that the heterogeneous membrane is with high geometry gradient from sub-micro-scale to sub-nano-scale and high

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space charge property. We show that the engineered MOF-based heterogeneous sub-nano-channel membrane can vividly rectify ion transport even in LiCl-methanol solution with a ratio as high as ~ 13 (Fig. 2a), capable of amplifying ionic current uni-directionally at the subnanometer-scale confinement, due to the broken symmetries in channel sizes, charges, and wettabilities (Fig. 2b). We thereby probe the use of this subnanometer-scale MOF-based ionic-diode membrane in harvesting osmotic energy from organic solutions. Captivatingly, a record power output of up to ~ 9.58 W/m² at a 50-fold LiCl gradient in methanol can be harnessed (Fig. 3a), majorly because of its unique geometry gradient channel structure (Fig. 3b), outperforming the bandgap of the commercial benchmark value (5 W/m²) and the other conventional membranes structures. The continuous MOF-based heterogeneous membrane we developed here is with high novelty in membrane technology and the design strategy provides a promising approach for constructing new multifunctional biomimetic membranes towards advanced high-performance energy conversion devices.

10:20am **C3-2-FrM-8 CVD Process Development of Thin Film Triniobium-Tin on Copper SRF Cavities**, *Mohamed A. Cheikh, S. McNeal, V. Arrieta, Ultramet, USA*

Innovative fabrication technologies for cost-effective high quality factor (high-Q), high-field superconducting radio frequency (SRF) components are needed for the economic viability of future accelerator facilities. The worldwide particle accelerator community continues to investigate alternatives and performance-enhancing modifications to bulk niobium accelerator components via the application of superconducting films. The Department of Energy (DOE) is interested in development of advanced process technologies to deposit superconducting materials such as triniobium-tin (Nb₃Sn), which has the potential to exceed the performance capabilities of bulk niobium when formed on the interior surface of existing bulk niobium, or less costly copper, accelerator component structures, enabling substantial fabrication and operating cost reductions for continuous wave and high-gradient accelerators.

In recent research for DOE, Ultramet has developed chemical vapor deposition (CVD) techniques to create well-bonded layers of Nb₃Sn on copper, niobium, and molybdenum substrates. Multiple copper single-cell 1.3-GHz SRF test cavities of the International Linear Collider (ILC) design were fabricated using the new CVD Nb₃Sn process. The CVD Nb₃Sn-on-copper fabrication methodology was scaled up and used to produce testable flanged Nb₃Sn-lined copper cavities and copper/Nb₃Sn cavities with an interlayer of CVD niobium formed using a thick film CVD process technology developed in concurrent research for DOE. RF performance capabilities and survivability at cryogenic temperatures were characterized by RF testing of the cavities at Cornell University.

Ultramet's thick film niobium and thin film CVD Nb₃Sn processes are uniquely well-suited for coating complex SRF accelerator component geometries because the virtually 100% dense coatings are formed on the substrate at the molecular level, and purity levels in excess of 99.99% are achievable. The CVD coating process exhibits the greatest throwing power, or ability to uniformly deposit materials onto/into intricately shaped or textured substrates.

Completion of this research was a necessary step toward the eventual commercial and scientific application of advanced accelerator component-forming technologies. A significant technical milestone was reached by the advances made in developing reliable fabrication techniques for reproducible CVD Nb₃Sn- and niobium-lined copper accelerator components that offer high-gradient operation and substantial cost reduction for SRF applications worldwide.

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