Advanced Characterization, Modelling and Data Science for Coatings and Thin Films
Room Town & Country C - Session CM4-2-MoA

Simulations, Machine Learning and Data Science for Materials Design and Discovery II

Moderators: Dr. Davide G. Sangiovanni, Linköping University, Sweden, **Dr. Ferenc Tasnadi**, Linköping University, Sweden

1:40pm **CM4-2-MoA-1 Computational Approach to Probing Hydrogen in Atomic Layer-Deposited Barrier Coatings**, ***Vladyslav Turlo [vladyslav.turlo@empa.ch]***, Empa, Swiss Federal Laboratories for Materials Science and Technology, Switzerland **INVITED**

The energy transition of our society requires an improved fundamental understanding of the chemical interaction of H with oxide materials, such as oxide membranes for H-purification, oxides for photocatalytic water splitting, and passivated oxides on steel. In particular, the effect of H impurities on the barrier properties of oxide layers grown by Atomic Layer Deposition (ALD) is of great scientific and technological interest, since hydrogen permeation barriers fabricated by ALD are broadly applied to address specific challenges for transport, handling, and storage of H, as well as for electronics, catalysis and gas sensing. However, resolving tiny changes in local chemical bonding states and structure of e.g. amorphous alumina oxides, as induced by H impurities originating from the ALD deposition process, still poses huge challenges for modern analytical tools up to date.

Here, the effect of hydrogen on the local chemical bonding states and structures of amorphous ALD alumina films is disclosed by predicting Auger parameter shifts, as measured by XPS/HAXPES, using a combination of atomistic and electrostatic modeling. First of all, it is demonstrated that a conventional melt quenching simulation procedure is not applicable for generating representative amorphous oxide structures with different H contents and densities, as observed in the experiment. Instead, a novel approach is proposed for simulating amorphous H-containing oxide structures by annealing reconstructed, highly defective crystalline hydroxide structures using a universal machine learning interatomic potential. As such, excellent agreement between the density, structure, and H-content was obtained between theory and experiment. Moreover, measured Auger parameter shifts for Al as a function of the H-content were accurately predicted by assuming all H atoms to be present in the form of hydroxyl ligands in the randomly interconnected 4-fold, 5-fold, and 6-fold nearest-coordination spheres of Al (by O). Combined atomistic and electrostatic modeling shows in detail how measured Auger shifts depend on the complex correlations between local coordination, bond lengths, bond angles, and ligand type(s) around the core-ionized atom, which equally applies to amorphous and crystalline compounds.

This work enables the computational design of new barrier coatings for hydrogen economy, providing a comprehensive computational characterization framework able to interpret even the tiniest Auger parameter chemical shifts obtained from experimental XPS/HAXPES techniques.

2:20pm **CM4-2-MoA-3 Conditions for the Preparation of Maximum-Quality Crystalline ZnO by Molecular Dynamics Simulations of the Atom-by-Atom Film Growth**, ***Jiri Houska [jhouska@kfy.zcu.cz]***, *Kamila Hantova,* University of West Bohemia, Czechia

Crystalline zinc oxide thin films are important due to a combination of optical transparency, electrical conductivity and piezoelectric and pyroelectric properties. These functional properties largely depend on perfection of the crystalline structure. Reproducing the growth of thin films by molecular-dynamics (MD) simulations is very useful for the disentanglement of processes and phenomena which take place in parallel in the experiment and can yield a lot of atomic-scale information which is difficult to access experimentally. After introducing MD simulations in general, classical MD based on a reactive force field is used to study the atom-by-atom growth of ZnO_x films on a crystalline template. The effect of kinetic energy of fast atoms (E_{fast}) and fraction of fast atoms (f_{fast}) at varied elemental ratio (x = [O]/[Zn]) is analyzed in a wide range. Following the visual inspection, the crystallinity is quantified in terms of network ring statistics.

Simulations with fixed f_{fast} = 100% revealed that the highest crystal quality was obtained at x = 1.03 (not at the intuitive ratio x = 1.00) and E_{fast} = 3-12 eV with a maximum at E_{fast} = 10 eV. When only a low E_{fast} = 1 eV is available, even higher x = 1.10 leads to the relatively best results. Simulations with varied f_{fast} and fixed E_{fast} = 10 eV revealed that the crystallinity at f_{fast} ≥ 50% is saturated. The ratio x = 1.03 is optimum at all these f_{fast} values and it is followed by x = 1.05 which also leads to higher crystal quality than x = 1.00. However, the ratio x = 1.10 is for the present energy distribution functions too high, not only in terms of growth rate which decreases with increasing x, but also in terms of crystal quality.

First, the results provide a quantitative insight into the role of individual deposition parameters. Second, the results explain available experimental data (for example, the dependence of the mobility of free charge carriers on the pulse-averaged target power density expresses the same character as the dependence of network ring statistics on E_{fast}). Third, the results facilitate a further improvement of the film properties. For example, it is important that the recommended E_{fast} is comparable to achievable positions of maxima of energy distribution functions during reactive HiPIMS.

2:40pm **CM4-2-MoA-4 Effect of the Presence of Oxygen on Hydrogen Adsorption on BCC Fe Surface: A Density Functional Theory Study Combined with Molecular Dynamics Simulations**, *Zixiong Wei, Fei Shuang,* ***Poulumi Dey [p.dey@tudelft.nl]***, Delft University of Technology, Netherlands

Hydrogen is one of the most promising candidates for the replacement of current carbon-based energy sources. It is one of the most potential candidates of sustainable energy produced in an eco-friendly manner. However, the use of hydrogen as an energy source is severely restricted by its damaging effect on mechanical properties of materials widely known as Hydrogen Embrittlement (HE). It is, thus, urgently needed to develop new HE resistant alloys or re-design the existing alloys for safe and efficient hydrogen storage and transportation. In this regard, Density Functional Theory (DFT) based approach is particularly important for obtaining atomistic insights into hydrogen interaction with surfaces to ensure less uptake of hydrogen by the material e.g. steels. Within this study, DFT is employed to obtain atomistic insights into hydrogen adsorption on different surfaces of bcc-Fe in the presence of oxygen. At the initial stage, we investigate the adsorption of oxygen on different surfaces of bcc-Fe using DFT following which we study the adsorption of hydrogen on Fe surface in the presence of the oxygen. At the next stage, we employed Molecular Dynamics (MD) simulations to study hydrogen diffusion into the bulk of bcc Fe from the surface in the presence of oxygen atoms on the surface. Our combined DFT-MD study thus offers atomistic insights into how surface oxygen atoms influence hydrogen intake into bcc Fe.

3:00pm **CM4-2-MoA-5 Machine Learning Prediction of Work Functions for No, No_2, Co, Co_2, and H_2S Gas Molecules Adsorbed on $Znga_2O_4$(111) Surfaces**, ***Po-Liang Liu [pliu@dragon.nchu.edu.tw]***, *Hsiang-Yu Hsieh, Chao-Cheng Shen,* National Chung Hsing University, Taiwan

Zinc gallium oxide is a metal oxide gas sensing layer with exceptional thermal and chemical stability, capable of detecting gases such as NO, NO_2, CO, CO_2, and H_2S. The work function of Zinc gallium oxide can be assessed through first-principles calculations based on Density Functional Theory, which allows for the prediction of the sensor's sensitivity. Although Density Functional Theory provides accurate computational results, its high computational cost and time requirements limit its applicability for large-scale surface screening. This study used a database based on a density functional theory-based zinc gallium oxide sensor model. We developed an automated workflow using Python programming to extract crystal structure features as input for the machine learning model. The processed and filtered input features were employed to predict the work function of the sensor model, achieving a mean absolute percentage error below 6% in the prediction results. This study presents a trained machine-learning model interface that allows users to input crystal structure files for the rapid and accurate evaluation of the work function of Zinc gallium oxide sensors.

3:20pm **CM4-2-MoA-6 Ml-Assisted Atomistic Modeling of Transition Metal Diborides: Mechanical Response and Phase-Dependent Phenomena**, ***Shuyao Lin [shuyao.lin@tuwien.ac.at]***, TU Wien, Institute of Materials Science and Technology, Austria*; Davide Sangiovanni, Lars Hultman,* Linköping Univ., IFM, Thin Film Physics Div., Sweden*; Paul Mayrhofer, Nikola Koutna,* TU Wien, Institute of Materials Science and Technology, Austria

Transition metal diborides (TMB_2) represent materials with ultra-high hardness and melting points but limited resistance to crack propagation. Understanding the thermodynamic stability of typical TMB_2 polymorph structures (α, ω, and γ) at finite temperatures as well as the phase-dependence of mechanical and fracture properties has been challenging due to non-trivial synthesis and structural similarity of the phase polymorphs, complicating their detection. This work presents uniform shear and tensile strain simulations of defect-free Group IV-VII TMB_2

ceramics using *ab initio* molecular dynamics as well as molecular dynamics powered by machine-learning interatomic potentials (MLIP), trained in the moment tensor potential framework. Studied materials include TiB_2 (Group IV), TaB_2 (Group V), WB_2 (Group VI), and ReB_2 (Group VII), covering the α, ω, and γ polymorphs. Among our main results is a robust workflow for training transferable MLIPs. These MLIPs are suitable for atomic-to-nanoscale MD simulations, allowing to understand deformation and fracture mechanisms of each TMB_2, and extending the insights into phase transformation mechanisms under shear deformation. By demonstrating the outstanding mechanical performance of TMB_2:s in extreme environments, our predictions clearly underpin their huge application potential in protective coatings and high-temperature engineering. To deepen our understanding of fracture behavior, we additionally perform Mode-I fracture simulations allowing to quantitatively assess fracture toughness (K_{IC}) using pre-cracked models. The results are discussed in light of relevant experimental data, including high-resolution transmission electron microscopy analysis of nanoindentation experiments on TiB_2 thin films.

4:00pm **CM4-2-MoA-8 Computational Modeling of Nanoelectronics and Emerging Materials**, ***Chao-Cheng Kaun [kauncc@gate.sinica.edu.tw]***, Academia Sinica, Taiwan **INVITED**

Using first-principles calculations, we investigate electronic transport through carbon-, oxide- and transition metal dichalcogenide (TMD)-based nanojunctions for nanoelectronic applications. Effects of biasing, defecting, contacting and quantum interfering are addressed. We study noncollinear interlayer exchange coupling in magnetic trilayers for spintronic application. Effects of spacing are uncovered. We also study the efficiencies of hybrid-protected perovskite quantum dot films for LED backlighting and hydrogen evolution reaction in oxides for sustainable-energy applications. Effects of polymer-adsorbing and material-configuring are highlighted. Moreover, we explore the plasmonic properties of complex transition metal nitrides for photonic applications.

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