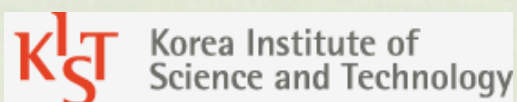# Nano Korea 2018, Satellite Session

July 13, 2018, 9:00 – 18:00
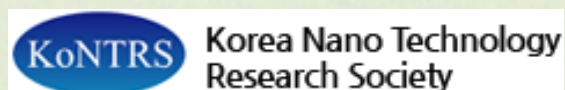KINTEX Exhibition Center II   Room 308,
Ilsan, Korea

Program & Abstracts

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SCIENTIFIC PROGRAM

09:00-09:30 Registration

09:30-10:40 (Chair: Kwang-Ryeol Lee)
09:30-09:40 Opening Remarks (Kwang-Ryeol Lee, KIST, Korea)
09:40-10:00 (Invited-1) Yoshiyuki Kawazoe (Tohoku Univ., Japan) "Materials Informatics to Realize Penta-graphene based on Reliable Materials Database"
10:00-10:20 (Invited-2) Jisoon Ihm (POSTECH, Korea) "First-principles study on Exfoliation Energies of Layered Materials"
10:20-10:40 (Invited-3) Yong-Hoon Kim (KAIST, Korea) "Toward the first-principles design of next-generation nanodevices"

10:40-11:00 Break

11:00-12:00 (Chair: Seungchul Kim)
11:00-11:20 (Invited-4) Isao Tanaka (Kyoto Univ., Japan) "First principles phonon calculations for structural materials"
11:20-11:40 (Invited-5) Kersti Hermansson (Uppsala Univ., Sweden) "Materials modelling to interpret experimental spectra – a REAL help!"
11:40-12:00 (Invited-6) Aloysius Soon (Yonsei Univ., Korea) "*Ab initio* thermodynamics of surface oxide structures under controlled growth conditions"

12:00-13:30 Lunch at KINTEX (to be announced), Group Photo

13:30-14:30 (Chair: Sang Uck Lee)
13:30-13:50 (Invited-7) Hiroshi Mizuseki (KIST, Korea) "New 2D and 3D Allotropes of Carbon"
13:50-14:10 (Invited-8) Junichiro Shiomi (U. Tokyo, Japan) "Geometric effects in carbon nanotube thermoelectrics"
14:10-14:30 (Invited-9) Kahyun Hur (KIST, Korea) "Controlling Wave Transport in Periodic and Quasiperiodic Structures"

14:30-15:00 Break

15:00-16:20 (Chair: Yong-Hoon Kim)
15:00-15:20 (Invited-10) C.-K. Andy Siu (City Univ. Hong Kong, Hong Kong) "Self-Assembled Binuclear $Cu(II)$ − L-Histidine Complex in the Gas Phase for Chiral Differentiation of Naproxen Drug"
15:20-15:40 (Invited-11) Byung-Hyun Kim (KIER, Korea) "Multiscale Modelling of the Oxygen Chemistry of CeO_2 Nanoparticles"
15:40-16:00 (Invited-12) Sang Uck Lee (Hanyang Univ., Korea) "Theoretical Study on Electrocatalytic Activity"
16:00-16:20 (Invited-13) Sang Soo Han (KIST, Korea) "First-Principles Guided Discovery of Novel Bimetallic Catalysts for H_2O_2 Direct Synthesis"

16:20-16:40 Break

16:40-18:00 Suggestion and Discussion for ACCMS-PRO (moderated by Kwang-Ryeol Lee, KIST, Korea)
19:00-21:00 Banquet (to be announced, near KINTEX)

Materials Informatics to Realize Penta-graphene based on Reliable Materials Database

Yoshiyuki Kawazoe
Industry Creation Hatchery Center, Tohoku University Sendai, Japan
Physics and Nanotechnology, SRM Institute of Science and Technology, Chennai, India

Based on the recent rapid progress in AI technology, a number of researchers have already started to introduce this new tool into their research fields to try making a fundamental improvement. It has been very successful in games (AI chess player is better than the human champion), operation, driving, trading, etc., all based on the "deep learning" technology. In our research field of materials science, recently MI=Materials Informatics is becoming a popular keyword. However, compared to other engineering fields above which treat "human made objects", our subject is directly connected to "nature" and fundamentally different from others; only God knows the materials.

There is a long history of materials database, studied, created, and used with a lot of effort all over the world. From 1995 I have been working to create and distribute the non-equilibrium phase diagram data for metallic amorphous materials and others as one of the Landolt-Boernstein Series from Springer. By this project, I have experienced how important to evaluate data before register to the database, since simply accumulating existing data sometimes makes serious problems for applications.

Experimental materials data are very costly to be accumulated to be able to reach a level of "big data" useful in MI. On the contrary, computer generated materials data are seemed to be cheaper based on the rapid increment in computer power, but are still not good enough in quality; for example resent standard method where a number of DFT calculations are conducted to construct a band gap database to be used for "band gap engineering" is theoretically not acceptable, since DFT is a ground state theory. We should concentrate on overcoming these problems. Actually this level of study we have already conducted more than 10 years ago on fluorides for UV laser applications. We have implemented GW approximation in our original all electron *ab initio* code TOMBO to be able to estimate absolute energy levels, which has a power of prediction of band gap values of unknown materials.

Concerning penta-graphene, since it is a new material, no experimental data have been accumulated, and MI is essentially important. We have studied penta-graphene for various applications including energy related subjects. We have started to apply MI technology to study penta-graphene to accelerate searching speed. In the future, combining a high level reliable materials database based on our effort and sophisticated deep learning technology, we could make a good contribution in MI.

First-principles study on Exfoliation Energies of Layered Materials

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The exfoliation energy, the energy required to peel off an atomic layer from the surface of a bulk material, is of fundamental importance in the science and engineering of layered materials. Traditionally, the exfoliation energy has been obtained from first-principles by calculating the difference in the energy between (i) a slab of N layers ($N \gg 1$) and (ii) a slab of $N - 1$ layers plus a layer separated from the slab. In this talk, we prove that the exfoliation energy can be obtained *exactly* as the difference in the energy between a bulk material (per atomic layer) and a single isolated layer [1]. The proposed method is (i) tremendously lower in computational cost than the traditional approach because it does not require calculations on thick slabs, (ii) still valid even if there is a surface reconstruction of any kind, (iii) capable of taking into account the relaxation of the single exfoliated layer, and (iv) easily combined with many-body computational methods. As a proof of principles, we calculate the exfoliation energies of graphene, hexagonal boron nitride, MoS_2, and phosphorene using density-functional theory.

Reference
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Toward the first-principles design of next-generation nanodevices

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In the effort to develop advanced electronic, optoelectronic, energy, and bio devices based on emerging low-dimensional materials, first-principles or *ab initio* simulations are playing an increasingly important role by providing atomistic information that are not easily obtained from experiments. For this purpose, I envision that a key ingredient that should be still matured and additionally developed is the capability to treat nonequilbirum junction systems under finite bias in a first-principles manner. In this talk, I will first apply the existing machinery that combins density functional theory (DFT) and nonequilibrium Green's function (NEGF) formalisms and predict that ultrahigh negative differential resistance (NDR) can be obtained from the nanowires derived from recently synthesized one-dimensional halide perovskites. Next, I will discuss the limitations of DFT-NEGF in simulating the finite-bias nonequilibrium electronic structure of nanoscale junctions and introduce the novel multi-space constrained search DFT (MS-DFT) formalism that we have recently developed. The MS-DFT formulation goes beyond the standard DFT-NEGF in several aspects, and as an application example I will consider graphene-based two-dimensional vertical heterosturcture tunneling transistors, which show NDR and are a promising platform to realize next-generation "More Moore" and "More than Moore" devices.

First principles phonon calculations for structural materials

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We have been working on first-principles phonon calculations to elucidate thermal properties, phase stability/transitions, deformation twinning, and others in a number of metallic and insulating systems. The central software is PHONOPY [1] developed by Togo, which is an open source package of phonon calculation based on the supercell approach. The SPGLIB code by Togo [2], another open source C library for finding and handling crystal symmetries, has also been successfully adopted. In order to investigate paramagnetic systems and/or disordered alloys, cluster expansion method is very powerful. A multi-purpose cluster expansion code, CLUPAN [3] by Seko, has been used for such purposes.

An example is the study of the continuous evolution of crystal structures in metallic elements based upon a large set of lattice dynamics calculations. Crystal structures are often treated as dense packing of atomic spheres. Face-centred cubic (FCC) and hexagonal close-packed (HCP) structures are favoured in many metals. We have developed a simple algorithm for automated searching of the phase-transition pathway based upon first-principles calculations [4]. Eigenvalue problems of the dynamical matrices of the supercell force-constant matrix at commensurate q points are solved to find dynamical instability. The method is applied to systematically pursue the continuous evolution of crystal structures. Following the algorithm, dynamical stability and interrelationships of different structures generated from a simple cubic (SC) structure are revealed for seven metallic elements, i.e., Na, Mg, Al, Cu, Ti, Zr and Hf. Structures generated from the SC structure are simple hexagonal (SH), body centered cubic (BCC), omega, FCC, HCP and five different kinds of long period structures, such as 6H, 9R and 12R. Effects of pressure are examined as well. The powerfulness of the automated method to investigate the nature of the phase transition and to predict as-yet-unknown metastable structures is demonstrated.

Investigation of atomic shuffling associated with deformation twinning is another example for the use of first principles phonon calculations. Results will be shown mainly on the $\{10\bar{1}2\}$ twin of HCP-metals (Be, Mg, Ti, Zr and Hf). When shear strain corresponding to the $\{10\bar{1}2\}$ twin is applied, one of the acoustic phonon-mode is significantly softened at an M-point of the HCP lattice. With the increase of the shear strain the M-point frequency becomes imaginary implying the spontaneous collective motion of atoms. This should correspond to the atomic shuffling associated with deformation twinning.

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Materials modelling to interpret experimental spectra – a REAL help!

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One class of applications where nanomaterials modelling play an undeniable role is in the generation of images and spectra that mimic those produced by experiment. This is an area where experiment and modelling meet in a very natural fashion.

In fact, modelling is sometimes the only way forward for experimentalists to be able to assign observed spectral features to the proper underlying structural feature. This may require a post-processing step after the initial generation of atomic trajectories other modelling data. Other times the image or spectral generation needs to be integrated with the simulation stage itself. In either case, this type of modelling is seldom trivial as the underlying physical processes are not trivial to describe.

For nanostructured materials, vibrational spectroscopies (IR, IRRAS, Raman, SFG,...) sail up as *the* most important methods for structure determination. I will discuss some examples of the use of modelling to predict vibrational frequencies of a high quality and to identify structure-property relations among them. My example systems are interfaces between waster and ionic materials.

Ab initio thermodynamics of surface oxide structures under controlled growth conditions

Taehun Lee[1], Yonghyuk Lee[1], Simone Piccinin[2], and Aloysius Soon[1]*

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Having a robust and predictive *ab initio* thermodynamic model to examine and describe the interplay of the oxygen gas and evaporated metal atoms on another metal substrate may prove to be very helpful in understanding the surface phase diagrams of these oxygen/metal systems. In this work, we examine the O/Cu/Au(111) system and provide a refined atomistic thermodynamic model which takes different definitions of the chemical potential of the less abundant metal, Cu into account. We argue that the latter highly depends on the various surface structures (overlayers and alloys) that forms on the metal substrate under growth conditions. We demonstrate that our improved thermodynamic model rationalizes new experimentally observed oxide structures and may pave a systematic way to predict new surface structures of reduced stoichiometries, which would otherwise be missed by the common practice of taking only the bulk limits.

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New 2D and 3D Allotropes of Carbon

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The search for new carbon structures have been very intriguing in physics, chemistry and materials science because of their scientific as well as technological importance. This fact has triggered research initiatives towards the development of various experimental and theoretical tools for predicting the desired properties of materials.

In this presentation, we will introduce a new methodology for the prediction of new carbon-based structures using combination of multi-start local search approach and atomistic simulations [1] by Tersoff potential and first-principles calculation. Next, we will discuss this approach by giving an example of carbon allotrope and checking its efficiency for the large system as well.

Using this approach, we have been able to predict new carbon allotropes named C_{568} (2D) [2] and H_{48} (3D) [3]. Interestingly, these allotropes are semiconducting in nature and possess some interesting mechanical, electronic and optical properties, which might find their applications in nanoelectronic devices. We further confirm their thermal and dynamical stabilities by performing phonon and *ab initio* molecular dynamics simulations.

References
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2. Babu Ram and Hiroshi Mizuseki, submitted
3. Babu Ram and Hiroshi Mizuseki, under preparation

Geometric effects in nanocarbon thermoelectrics

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Thermal properties of nanocarbons such as carbon nanotubes and graphenes have been widely studied aiming to use them for high thermal conductivity materials such as thermal interface materials and nanofluids in forms of films and composites. There, thermal conductivity of nanocarbons was found to be sensitive to geometrical effects, such as defects and mechanical deformation, which were a drawback. However, as nanocarbon thermoelectrics have caught attention as a flexible energy harvesting device, the geometrical effects have become a merit since thermoelctrics requires thermal conductivity to be low. Here, geometrical effects also influence electrical properties, in general in unfavorable way, and thus the major game is to identify and use geometrical effects that reduce thermal conductivity more than electrical conductivity. This also implies that the uncontrolled geometrical effects in nanocarbons such as defects that are omnipresent in usual as synthesized nanocarbon materials are important for the material performance. This motivates the studies on the response of thermal and electrical conduction to geometrical variation of nanocarbon, where numerical simulation is naturally useful. In this talk, we will introduce some works in this course, covering case studies with defects [1,2], bending/buckling [3.4], encapsulation [5,6].

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Controlling Wave Transport in Periodic and Quasiperiodic Structures

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Controlling the wave transport properties of materials provides opportunities for improving material properties for a variety of applications including photonics, thermoelectrics, acoustics, and electronics. In particular, translational symmetry of materials strongly governs transport properties of every form of waves such as electron, photon, and phonon since the waves having a wavelength commensurate with crystal's periodicity can transmit without losses. In this talk, I will discuss photonic and phononic properties of periodically structured nanomaterials derived from bottom-up type block copolymer self-assembly. Peculiar wave transport behaviors of quasiperiodic structures, ordered and well-defined structures without any translation order, will be further discussed.

Self-Assembled Binuclear Cu(II)–*L*-Histidine Complex in the Gas Phase for Chiral Differentiation of Naproxen Drug

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Naproxen is one of the most consumed nonsteroidal anti-inflammatory drugs and marketed as *S*-naproxen as its *R*-enantiomer is hepatotoxic [1]. Developments of efficient and reliable methods for chiral differentiation and quantification of naproxen are thus essential [2]. In this work [3], we discovered a self-assembled binuclear Cu(II) diastereomeric complex ion in the gas phase, $[(Cu^{II}L\text{-His})_2(S/R\text{-naproxen})-3H]^+$, that can be applied to effectively determine the absolute configuration of naproxen and quantify the enantiomeric excess of *S/R*-naproxen mixtures using collision-induced dissociation with a tandem mass spectrometer. The key candidate structure of this gas-phase diastereomeric complex ion has an unusual self-assembled, compact geometry with the two Cu(II) ions bridged closely together by the carboxylate groups of the two histidines, proposed using density functional theory, and supported by ion-mobility spectrometry and collision cross section modeling. The difference in dissociation efficiency of the two diastereomers is attributed to the σ-π interaction between the NH_2 group of one histidine and the naphthyl ring of naproxen in the complex. The present report observes and characterizes the diastereomeric complexes of $[(Cu^{II})(L\text{-His})_2]$ with aromatic acid, which could contribute to the recognition of chiral aromatic acids, design of catalysts based on binuclear copper-bound complex, as well as the better understanding of metal-ion complexation by His or His-containing ligands.

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Multiscale Modelling of the Oxygen Chemistry of CeO₂ Nanoparticles

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Ceria (CeO₂, cerium oxide) is a reducible metal oxide with a unique oxygen chemistry which makes use of various different technological applications, including a three-way catalyst, a ceramic electrolyte in solid oxide fuel cells and chemical sensors [1]. These applications primarily exploit the oxygen storage capacity of CeO₂, which can be interpreted as a redox capability enabled by which oxygen vacancies may be formed, healed and diffuse in the crystalline lattice.

Synthesized ceria is often found in the form of agglomerates of nano-crystallites [2-4]. The agglomeration of nanoparticles (NPs) results in the formation of specific interface, which obviously affects the catalytic activity in ceria. In this work, we address that stable ceria-ceria interface structures formed between agglomerated ceria NPs will show unique oxygen redox properties, which are very different from those of either the bare surface or the bulk material, in particular those related to the increased oxygen storage capacity found for small ceria nanoparticles (with d < 5nm). [5]

Here, we applied a multiscale simulation approach combining three different levels of theory in the typical simulation hierarchy: The reactive force field (ReaxFF) [6], the approximate density functional method, (self-consistent charge density functional based tight binding, SCC-DFTB) [7], and finally Hubbard augmented density functional theory (DFT+*U*). We will present the stable interface structures and the effect of interface formation on the oxygen chemistry.

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Theoretical Study on Electrocatalytic Activity

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We introduce the density functional theory (DFT) based computational approaches to the study of various electrochemical reactions (hydrogen evolution reaction (HER), oxygen evolution reaction (OER), oxygen reduction reaction (ORR), Iodion reduction reaction (IRR) occurring on heterogeneous catalysis surfaces. A detailed computational approach to the theoretical interpretation of electrochemical reactions and structure-catalytic activity relationships for graphene-based catalysts will be discussed. The electrocatalytic activity of catalysis can be theoretically evaluated by overpotential value determined from free energy diagram (FED) of electrochemical reactions. By comparing electrocatalytic activity of systematically designed graphene-based catalysts, we will discuss the structure-catalytic activity relationships, especially the electronic and geometrical effects of heteroatom dopants.

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First-Principles Guided Discovery of Novel Bimetallic Catalysts for H$_2$O$_2$ Direct Synthesis

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The direct synthesis of hydrogen peroxide (H$_2$O$_2$) from H$_2$ and O$_2$ represents a potentially atom-efficient alternative to the current industrial indirect process. And, Pd has been currently used as an efficient catalyst for direct H$_2$O$_2$ synthesis, although it has not been commercialized yet due to its expensive cost and still unsatisfied catalytic performances. In this regard, it is imperative to discover an inexpensive alternative with a superior catalytic property. For this purpose, we have designed several bimetallic catalysts replacing Pd through first-principles calculations, in particular, a high-throughput approach combining a calculation automation. Moreover, the designed catalysts were experimentally confirmed.

Electronic structure of a material is one of key features to determine catalytic properties as like *d*-band center theory. In the extension line of the theory, we screened 4350 bimetallic alloys by first-principles calculation and calculated similarities of their electronic densities of states (DOS) by comparing with that of Pd. Then, we filtered 10 potential catalysts (thermodynamically miscible) with the highest DOS similarities. And then, we synthesized the 10 candidates and experimentally measured their catalytic properties for H$_2$O$_2$ direct synthesis. After the experimental confirmation, it is found that, of the 10 candidates, three bimetallic alloys indeed show similar or superior catalytic properties to Pd. In addition, we suggest a new design scheme using "immiscible" bimetallic systems based on first-principles calculation, and the scheme is also successfully confirmed by an experiment. In this talk, I will also discuss the scheme.