

초청연사

강영호 (재료연구소 소재데이터센터)

김동훈 (한국과학기술연구원 계산과학연구센터)

김범현 (고등과학원 계산과학부)

김봉재 (군산대학교 물리학과)

김용성 (한국표준과학연구원 양자기술연구소)

박경화 (버지니아공대 물리학과)

박노정 (울산과학기술원 물리학과)

박세영 (기초과학연구원 강상관계 물질 연구단)

박창원 (기초과학연구원 원자제어 저차원 전자계 연구단)

이인호 (한국표준과학연구원 양자기술연구소)

이재찬 (성균관대학교 신소재공학부)

서호성 (아주대학교 물리학과)

장기주 (한국과학기술원 물리학과)

정성철 (부경대학교 물리학과)

조영미 (삼성디스플레이)

채기성 (고등과학원 계산과학부)

자문위원회

김용현 (한국과학기술원), 김용훈 (한국과학기술원),
권영균 (경희대학교), 박철홍 (부산대학교),
심지훈 (포항공과대학교), 유재준 (서울대학교),
이승미 (한국표준과학연구원), 조준형 (한양대학교),
정유성 (한국과학기술원), 최형준 (연세대학교),
한상수 (한국과학기술연구원), 한승우 (서울대학교)

조직위원

손영우 (고등과학원)
김세중 (UST)

담당자 : 김혜미 (고등과학원)

전화 : 02-958-2638

이메일 : hmkim@kias.re.kr

홈페이지 : <http://events.kias.re.kr/h/ESCW2018/?pageNo=3182>

제14회 고등과학원

전자구조 계산학회

일시 : 2018년 6월 21일 (목) - 6월 22일 (금)

장소 : 고등과학원 1호관 대강당

제 14회 고등과학원 전자구조계산학회 학회

6월 21일 (목)		6월 22일 (금)	
8:30 – 9:20 등록		8:30 – 9:30 등록	
9:20 – 9:30 환영인사			
Session I: 물질설계 (Materials Design) 사회 김한철 (숙명여대)	09:30 - 10:10 장기주 (한국과학기술원) Prediction of new superconducting silicon allotropes at ambient pressure using AMADEUS	Session VII: 결함과 뭉치 (Defects and Clusters) 사회: 이승미 (한국표준과학연구원)	9:30 - 10:10 김용성 (한국표준과학연구원) Magic Clusters of MoS2 by Edge S2 Inter-Dimer Spacing Modulation
	10:10 - 10:50 이인호 (KRISS) Materials design using global optimization and artificial intelligence		10:10 - 10:50 서호성 (아주대) First-principles theory of defect-based quantum bits in semiconductors
	10:50 - 11:30 채기성 (고등과학원) SANDWICH for Two-Dimensional Materials Design	10:50 – 11:20 휴식	
11:30 – 12:00 휴식		Session VIII: 상호작용 현상 2 (Correlated Phenomena 2) 사회 고아라 (기초과학연구원)	11:20 - 12:00 박세영 (기초과학연구원) First-principles study of charge-order-induced ferroelectricity in LaVO3/SrVO3 superlattices
Session II: 상호작용 현상 1 (Correlated Phenomena 1) 사회 심지훈 (포항공대)	12:00 - 12:40 이재찬 (성균관대) Defects in correlated oxides		12:00 - 12:40 김봉재 (군산대) Anisotropic magnetic interactions in transition metal oxides: the cases for Sr2RuO4 and Sr2VO4
	12:40 - 13:20 김범현 (고등과학원) Electronic and excitation properties of 4d/5d transition-metal compounds with a honeycomb lattice	12:40 – 14:00 점심식사	
13:20 – 14:30 점심식사		Session IX: 위상학적 성질 (Topological Properties) 사회 김영국 (성균관대)	14:00 - 14:40 박노정 (울산과학기술원) Ab initio TDDFT study of band topology and phonon-dressed spin state of spin-orbit coupled semiconductors
Session III : 에너지응용 물질 (Energy Materials) 사회 김용훈 (한국과학기술원)	14:30 - 15:10 정성철 (부경대) First-Principles Study of Energy Storage Materials		14:40 - 15:20 박경화 (버지니아공대) Evolution of Fermi Arcs in a Magnetic-Field Induced Weyl Semimetal from Wannier-Function-based Tight-Binding Model
	15:10 - 15:50 김동훈 (한국과학기술연구원) Novel Materials for Ferroelectric Photovoltaics	15:20 - 15:40 폐회 및 우수 포스터 시상식	
15:50 – 16:10 휴식			
Session IV: 전자-소리알 상호작용 (Electron-Phonon Interactions) 사회 민승규 (울산과학기술원)	16:10 - 16:50 박창원 (기초과학연구원) Two-dimensional charge density wave as a ground state of interacting soft phonons		
	16:50 - 17:30 강영호 (재료연구소) Electron-phonon interactions and their influence on transport property in polar semiconductors		
Session V: 산업체 응용 (Industrial Application) 사회 손영우 (고등과학원)	17:30 - 18:10 조영미 (삼성디스플레이) Materials modeling and design for OLED display application		
Session VI: 포스터 발표 사회 김세중 (과학기술연합대학원대학교)	18:10 - 20:00 포스터 발표 및 저녁식사		

Prediction of new superconducting silicon allotropes at ambient pressure using AMADEUS

Kee Joo Chang,¹ Ha-Jun Sung,¹ Woo Hyun Han,¹ and In-Ho Lee²

¹*Department of Physics, Korea Advanced Institute of Science and Technology, Daejeon, Korea*

²*Korea Research Institute of Standards and Science, Daejeon, Korea*

Recently, we have developed a protocol for computational materials design, called AMADEUS (*Ab initio* MAterials DEsign Using cSa), in which the conformational space annealing (CSA) algorithm for global optimization is combined with first-principles density functional calculations [1]. The AMADEUS has been successfully used to predict new materials, such as Si and C allotropes with optically allowed direct band gaps, a new carbon allotrope belonging to the class of topological nodal line semimetals, non-icosahedral B allotropes formed as an intermediate phase on the pressure-induced transition pathway from α -B to γ -B, a new phosphorus allotrope called green phosphorus with a layered structure and tunable direct band gaps, and a two-dimensional triangular Kagome lattice of boron that exhibits the exotic electronic properties [1-8]. Here we will show the successful application of AMADEUS to predict a novel metallic clathrate structure of silicon at ambient pressure [9]. The new Si₆ clathrate can be obtained by removing Na atoms from a chemical precursor NaSi₆ in the *P6/m* space group that is discovered using AMADEUS at high pressure. We find that both the metallic Si₆ and NaSi₆ clathrates are stable and superconducting with the critical temperatures of about 12 and 13 K at zero pressure, respectively.

References

1. I.-H. Lee, Y. J. Oh, S. Kim, J. Lee, and K. J. Chang, *Comp. Phys. Commun.* 203, 110 (2016).
2. I.-H. Lee, J. Lee, Y.J. Oh, S. Kim, and K.J. Chang, *Phys. Rev. B* 90, 115209 (2014).
3. Y. J. Oh, I.-H. Lee, S. Kim, J. Lee, and K. J. Chang, *Sci. Rep.* 5, 18086 (2015).
4. I.-H. Lee, J. Lee, Y. J. Oh, S. Kim, and K. J. Chang, *Phys. Rev. B* 93, 085201 (2016).
5. H.-J. Sung, S. Kim, I.-H. Lee, and K. J. Chang, *NPG Asia Materials* 9, e361 (2017).
6. W. H. Han, Y. J. Oh, D.-H. Choe, S. Kim, I.-H. Lee, and K. J. Chang, *NPG Asia Materials* 9, e400 (2017).
7. S. Kim, W. H. Han, I.-H. Lee, and K. J. Chang, *Sci. Rep.* 7, 7279 (2017).
8. W. H. Han, S. Kim, I.-H. Lee, and K. J. Chang, *J. Phys. Chem. Lett.* 8, 4627 (2017).
9. H.-J. Sung, W. H. Han, I.-H. Lee, and K. J. Chang, *Phys. Rev. Lett.* 120, 157001 (2018).

Materials design using global optimization and artificial intelligence

In-Ho Lee¹, W. H. Han², H.-J. Sung², J. Lee³, and K. J. Chang²

¹*Korea Research Institute of Standards and Science, Daejeon 34113, Korea*

²*Department of Physics, Korea Advanced Institute of Science and Technology, Daejeon 34141, Korea*

³*Korea Institute for Advanced Study, Seoul 02455, Korea*

Condensed matter theorists have been successful to explain some of the observed properties of materials. Recent advances in computational capabilities have revolutionized materials design/engineering. Several inverse methods, in which a specific material property is initially assigned and target materials are subsequently searched for, have been proposed and used for a wide range of applications in materials design and discovery. In this approach, exploring and identifying materials with their synthesis routes remains a challenge. The objective of this talk is to present a set of methods that can efficiently explore and produce both multiple materials and multiple reaction pathways. We combine the global optimization method of conformational space annealing (CSA) with first-principles electronic structure calculations. We introduce a neural network approach using Gaussian radial basis functions to fit machine learning interatomic potentials. The CSA algorithm is used to find optimal parameters in potential fitting. We show that the proposed Gaussian radial basis function neural network combined with the global optimization approach is highly efficient in training potentials, allowing for reliable atomistic simulations without the loss of numerical accuracy. Our scheme not only provides a new perspective for the inverse problem of materials design but also may serve as a new tool for the computational design of a wide range of materials. Global searching for reaction pathways is a long-standing challenge in computational physics, chemistry, and biology. We also present a computational approach, called Action-CSA, to find multiple reaction pathways connecting fixed initial and final states through the global optimization of the Onsager–Machlup action using the CSA algorithm. Action-CSA successfully overcomes large energy barriers via crossovers and mutations of pathways and finds all possible pathways of small systems without any initial guess on pathways. The results show that Action-CSA is an efficient and robust computational approach to study multiple pathways in the problems of complex reactions and large-scale conformational changes. The utilization of global optimization and artificial intelligence in first-principles electronic structure calculations and the kinetic theory will accelerate solving the materials search problem.

Reference URLs:

<http://webzine.kps.or.kr/contents/data/webzine/webzine/15088273131.pdf>

<http://webzine.kps.or.kr/contents/data/webzine/webzine/15211841581.pdf>

<http://webzine.kps.or.kr/contents/data/webzine/webzine/14762092704.pdf>

SANDWICH for Two-Dimensional Materials Design

Kisung Chae

School of Computational Sciences, Korea Institute for Advanced Study, Korea

Quasi two-dimensional (2D) layered materials with a finite thickness may offer great promises in various applications owing to their intriguing properties. Furthermore, the property-spectrum of these materials can become even more extensive when they make heterostructures. To meet various demands, it is requested to have a fruitful database for properties of 2D materials, which can be achieved by designing novel 2D materials. Interestingly, it has recently been found that most of the 2D materials can be categorized by their space group [1]. This indicates that materials- and property-spectrum can be extended by searching for new 2D prototypical crystal structures. In this talk, I will present a novel structure-searching method (SANDWICH*) [2], by which conformational space for finding stable 2D crystalline materials can be significantly reduced. Specifically, 2D materials are constructed by “sandwiching” bridge atoms between top and bottom surface layers with a specific space group. Note that the bridge atoms can sit on a subset Wyckoff positions corresponding to the space group determined by the surface layers. By using this method, a new family of quasi-2D silicon crystals is proposed, which shows varying thickness and electronic structures, ranging from semimetal to indirect semiconductor with a band gap up to ~ 0.5 eV [2]. In addition, novel 2D T₃X materials (T=C, Si, Ge, Sn; X=O, S, Se, Te) are also proposed with a variety of electronic structures, ranging from indirect/direct band gap semiconductor, to quantum spin Hall insulator [3].

[1] Mounet *et al.*, Nature Nanotechnology **13**, 246 (2018)

[2] **Chae et al.**, 2D Materials **5**, 025013 (2018); arXiv:1709.01675 [3] **Chae et al.**, *in preparation*

* Search by *Ab initio* Novel Design via Wyckoff positions Iteration in Conformational Hypersurface

Defects in correlated oxides

Kitae Eom¹, Minsu Choi¹, Euiyoung Choi¹, Kyung Song², Si-Young Choi², Daesu Lee³, Jung-Woo Lee³, Hua Zhou⁴, Chang-Beom Eom³, and Jaichan Lee^{*1}

¹*School of Advanced Materials Science and Engineering, Sungkyunkwan University (SKKU), Suwon, 16419, Republic of Korea*

²*Department of Materials Modeling and Characterization, Korea Institute of Materials Science, Changwon, 51508, Republic of Korea*

³*Department of Materials Science and Engineering, University of Wisconsin-Madison, Madison, Wisconsin, 53792, United States*

⁴*Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois 60439, United States of America*

Defects introduced into a material in an intrinsic or extrinsic way result in a wide variety of functionality in the material. We have studied an intrinsic defect such as oxygen vacancy, and extrinsic defects La in a transition metal oxide, SrTiO₃. It is well known from several studies that the oxygen vacancies can be singly ionized. However, the first-principles study shows that the oxygen vacancies cluster in a linear way, accompanied by electron localization, which successfully explains several phenomena including the decrease in the carrier density and photoluminescence in oxygen deficient SrTiO₃. Electron doping was also studied in La:SrTiO₃ thin films where the film thickness was varied down to a few unit cells. Large electron-lattice coupling is obtained along with anomalously large tetragonal distortion, which is sensitively influenced by the in-plane strain imposed to the thin films. Here, we design and demonstrate a strongly enhanced electron-lattice coupling in electron-doped SrTiO₃, that is, the t_{2g} electron system. In ultrathin films of electron-doped SrTiO₃ [i.e., (La_{0.25}Sr_{0.75})TiO₃], we reveal the strong electron-lattice-orbital coupling, which is manifested by extremely increased tetragonality and the corresponding metal-to-insulator transition. Our findings open the way of an active tuning of the charge-lattice-orbital coupling to obtain new functionalities relevant to emerging nanoelectronic devices.

Electronic and excitational properties of 4d/5d transition-metal compounds with a honeycomb lattice

Beom Hyun Kim

School of Computational Sciences, Korea Institute for Advanced Study, Korea

Electronic and excitational properties of 4d/5d transition-metal (TM) compounds are strongly attributed to the mutual interplay among electronic kinetics, electron-electron correlation, and strong spin-orbit coupling (SOC). In honeycomb systems like Na_2IrO_3 and $\square\text{-RuCl}_3$, edge-shared geometry gives rise to strong directional hopping between adjacent TMs, which can play a role in the formation of quasimolecular orbitals (QMOs) in weak coupling limit [1] and the realization of Kitaev-type interaction in strong coupling limit [2]. Because the strength of electron-electron correlation is in a similar energy scale of both hopping integral and SOC, their insulating nature has been controversial. For this end, two contradictory scenarios, i.e., Slater picture based on QMOs vs. Mott picture based on relativistic j_{eff} orbitals, has been proposed.

In this talk, we will explore their electronic characteristics based on minimal microscopic model which can capture both extreme insulating phases on an equal footing [3]. We will also present the theoretical excitational features measured with the optical conductivity, Raman spectroscopy, and resonant inelastic x-ray scattering. Based on our result, we will clarify the insulating nature of both Na_2IrO_3 and $\square\text{-RuCl}_3$.

[1] I. I. Mazin, H. O. Jeschke, K. Foyevtsova, R. Valentí, and D. I. Khomskii, Phys. Rev. Lett. **109**, 197201 (2012)

[2] G. Jackeli and G. Khaliullin, Phys. Rev. Lett. **102**, 017205 (2009)

[3] B. H. Kim, T. Shirakawa, and S. Yunoki, Phys. Rev. Lett. **117**, 187201 (2016)

First-Principles Study of Energy Storage Materials

Sung Chul Jung

Department of Physics, Pukyong National University

Computational materials science is important to understand the fundamental mechanism of energy storage materials and to improve their electrochemical performance. By combining density functional theory calculations and ab initio molecular dynamics simulations, we elucidate the physical and chemical properties of battery electrode materials and correlate them with electrochemical properties. We present the calculation results of several electrode and coating materials for Li-ion and Na-ion batteries such as Si, P, SiO, Sn₄P₃, graphite, and Al₂O₃. The calculation results reveal that capacity, rate, and cycle performance are closely associated with thermodynamic stability, ionic/electronic conductivity, and mechanical property of electrode material, respectively, and provide atomic-level insight into the design of efficient electrode materials.

Talk No. 07

Novel Materials for Ferroelectric Photovoltaics

Donghun Kim

Computational Science Research Center, Korea Institute of Science and Technology

Despite their potential to exceed the theoretical Shockley-Queisser limit, ferroelectric photovoltaics (FPVs) have performed inefficiently due to their extremely low photocurrents. In this talk, I will suggest novel ferroelectric materials that are likely to overcome the challenge, as a result of density functional theory calculations. These works indicate that the large band-gap and inefficient e-h separations of active layer materials are the most important issues to be addressed for FPVs to compete with conventional devices.

Talk No. 08

Two-dimensional charge density wave as a ground state of interacting soft phonons

Changwon Park

Center for Artificial Low Dimensional electronic Systems, Institute for Basic Science, Pohang

Many transition metal dichalcogenides (TMDCs) such as 2H-NbSe₂ and 1T-TaS₂ is unstable to periodic lattice distortion leading to a charge density wave. Their distortion patterns depend on temperature, pressure, alloying or intercalation and are studied extensively from 1970's. Until now, the phase transitions are explained in the framework of phenomenological Landau theory that the link between parameters in the Landau theory and first-principle calculations is missing. In this talk, I will show interatomic potential model can explain all the essential features of phase transitions and replace the Landau theory. From the model, charge density wave can be viewed as a ground state of soft phonon mode interacting via short-ranged anharmonic potential. The agreement between theory and experimentally observed distortion patterns are superior to the traditional Landau theory. The procedure that can extract anharmonic potentials from first-principles calculations will be suggested.

Electron-phonon interactions and their influence on transport property in polar semiconductors

Youngho Kang¹

¹Korea Institute of Materials Science

Electron-phonon interactions (EPIs) are ubiquitous in solids and responsible for diverse materials physics. For instance, they provide intrinsic scattering channel of carriers in semiconductors as well as enable optical absorption in indirect-gap insulators. Therefore, EPIs have been the subject of intensive researches in condensed matter physics.

In this work, we investigate EPIs in polar semiconductors in which constituent elements possess different valence states and their impacts on the transport properties of semiconductors. We develop a framework to compute the carrier mobility based on the electronic band structure, vibrational spectra, as well as EPIs, which are obtained in an unempirical way using first-principles calculations. In our calculations, the long-range EPIs due to the polar longitudinal optical (LO) modes are properly accounted for. We apply this method to study the carrier mobility of various semiconductors such as ABX₃ perovskite, Ga₂O₃, and TiO₂. Based on our results, we address several issues: (1) conditions for achieving high mobility, (2) anisotropy in the electrical conductivity, and (3) influence of renormalization of a band structure on the transport properties.

Materials modeling and design for OLED display application

Youngmi Cho¹

¹CAE Team, Samsung Display, Youngin, Korea

Recently, OLED display technology has attracted interest due to the rapidly expanding market in mobile, VR and automotive area. High density resolution, high mobility and multi-function are challenges in market and they are related with efficiency, color, lifetime of OLED, electrical stability of TFT and mechanical reliability of stack structure. In this presentation, I will introduce technical issue in OLED display including material, process, device, stack architecture and application of materials modeling for OLED, TFT, and Polymer. In OLED applications, bulk transport in organic solid, optical properties, photo-chemical reaction are investigated by MD and TD-DFT simulation, in TFT applications, defect dynamics are investigated by DFT, kMC and in Polymer applications, polymerization process and mechanical properties are investigated by TD-DFT, MD simulation. Finally, recent approach and limitation using multi-scale simulation and machine-learning will be reported.

Magic Clusters of MoS₂ by Edge S₂ Inter-Dimer Spacing Modulation

Junga Ryou and Yong-Sung Kim

Korea Research Institute of Standards and Science

MoS₂ has been used as a hydro-desulfurization reaction catalysis and recently attracted great attention for hydrogen evolution reaction catalysis. The edges of MoS₂, which are manifested in nanoclusters, have been known to play the central role in the catalytic activity and their detailed atomic and electronic structures have been intensively studied. Direct chemical vapor deposition growth of MoS₂ in typical S-rich condition yields as precursors S-saturated Mo-edge MoS₂ triangular nanoclusters. The MoS₂ nanoclusters have also been used in field-effect transistors, and the edge electronic states have been found to contribute significantly. A long-term puzzling aspect on the MoS₂ nanoclusters is that there are particularly abundant sizes, so-called the magic clusters. In this study, we investigate the atomic and electronic structures of the S-saturated Mo-edge triangular MoS₂ nanoclusters through density-functional theory calculations. It is shown that the edge S₂ dimers in the S₂-Mo-edge MoS₂ triangular nanoclusters prefer to undergo $\sqrt{5}$ lateral spacing modulation. It is a consequence of the Peierls instability of the edge S₂-p_xp_x * (S₂-□_x) electrons interacting with the Mo-d_{xy} sub-edge electrons. The $\sqrt{5}$ modulation affects the edge energies of the finite-size nanoclusters, and the especially stable sizes are found to be $N=5n-2$ and $5n$, where N is the number of S₂ dimers at each edge and n is a positive integer. The S₂-Mo-edge becomes S₂-□_x semiconducting and sub-edge Mo-d_{xy} metallic for large N . The obtained stable sizes and the edge modulation structures are found to be in good agreement with the scanning tunneling microscopy experiments.

First-principles theory of defect-based quantum bits in semiconductors

Hosung Seo

*Department of Physics & Department of Energy Systems Research
Ajou University, Suwon, Republic of Korea*

In recent years, remarkable advances have been reported in the development of defect spin quantum bits (qubits) in semiconductors for solid-state quantum information science and technology. Promising spin qubits include the nitrogen-vacancy center in diamond, dopants in silicon, and the silicon vacancy and divacancy spins in silicon carbide. In this talk, I will highlight some of our recent efforts devoted to defect spin qubits in wide-gap semiconductors [1-4]. In the first part of the talk, I will explain basic concepts of using point defects in semiconductors as qubits and their applications. Then, I will describe our recent combined theoretical and experimental study on remarkably robust quantum coherence found in the divacancy qubits in silicon carbide [2]. I will also discuss progress and challenges in computational design of new spin defects for use as qubits in piezoelectric crystals such as AlN and SiC, including a new defect design concept using large metal ion - vacancy complexes [1]. I will conclude my talk by discussing a perspective of combining density functional theory and cluster correlation expansion calculations to predict not only the thermodynamic and electronic properties of defect qubits, but their coherent quantum properties from first-principles.

- [1] H. Seo, H. Ma, M. Govoni, and G. Galli, *Physical Review Materials* **1**, 075002 (2017).
- [2] H. Seo, A. L. Falk, P. V. Klimov, K. C. Miao, G. Galli, and D. D. Awschalom, *Nature Communications* **7**, 12935 (2016).
- [3] H. Seo, M. Govoni, and G. Galli, *Scientific Reports* **6**, 20803 (2016).
- [4] W. F. Koehl, H. Seo, G. Galli and D. D. Awschalom, *MRS Bulletin* **40**, 1146 (2015).

First-principles study of charge-order-induced ferroelectricity in LaVO₃/SrVO₃ superlattices

Se Young Park^{1,2}, Anil Kumar³, and Karin M. Rabe⁴

¹*Center for Correlated Electron Systems, Institute for Basic Science (IBS), Seoul 08826, Republic of Korea*

²*Department of Physics and Astronomy, Seoul National University (SNU), Seoul 08826, Republic of Korea*

³*Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*

⁴*Department of Physics & Astronomy, Rutgers University, Piscataway, NJ 08854, USA*

The identification of novel mechanisms for ferroelectricity and discovery of their materials realizations has attracted much interest in recent years. One approach, proposed by Khomskii* is to combine two charge orderings, neither of which separately lift inversion symmetry, to generate a switchable polar structure. By generalizing this idea, I will introduce a new design principle utilizing (A,A')BO₃ perovskite oxide superlattices in which the superlattice layering of A-site cations combined with a charge ordering induces switchable polarization. As a candidate material, I will present the structure and properties of 1:1 superlattice composed of LaVO₃ and SrVO₃ using a first-principles density-functional-theory-plus-*U* (DFT+*U*) method and show that layered charge ordering combines with the layered La/Sr cation ordering to produce a polarization comparable to BaTiO₃ by electron transfer between the V³⁺ and V⁴⁺ layers. The vanishing energy difference between polar and non-polar states with small tensile strain suggests that the polar state could be induced by applied electric field, and, depending on the switching process, a ferroelectric hysteresis loop could be observed.

References:

*D. V. Efremov, J. van den Brink, and D. I. Khomskii, *Nat. Mater.* **3**, 853 (2004).

Se Young Park, Anil Kumar, and Karin M. Rabe, *Phys. Rev. Lett.* **118**, 087602 (2017).

Anisotropic magnetic interactions in transition metal oxides: the cases for Sr_2RuO_4 and Sr_2VO_4

Bongjae Kim

Dept. of Physics, Kunsan National University

Anisotropic magnetic interactions are found to play key roles in explaining many exotic properties in various types of transition metal oxides. In this talk, I will discuss two quasi-square net transition metal oxide systems: Sr_2RuO_4 and Sr_2VO_4 .

For Sr_2RuO_4 , a candidate of chiral p-wave superconductor, we first access the possibility of proposed order parameter rotation in an external magnetic field of 200 Oe, and conclude that the spin-orbit interaction in this material is several orders of magnitude stronger than this hypothesis implies. Thus, the observed invariance of the Knight shift across T_c has no plausible explanation, and casts doubt on using the Knight shift as an ultimate litmus paper for the pairing symmetry. Then, quantitative double-exchange-like model, which combines itinerant fermions with magnetic interaction is proposed. This is complementary to the Hubbard-model-based calculations published so far, and forms an alternative framework for exploring superconducting symmetry in Sr_2RuO_4 .

For Sr_2VO_4 , I will talk about highly competing exchange interactions in the system, which places the system at the crossover between XY- and Heisenberg model regime by involving non-negligible interlayer couplings. Interestingly, two-site exchange anisotropy is found to be strong which is relieved by the orthorhombic distortion induced by the spin-stripe order.

Bongjae Kim, Sergii Khmelevskiy, I. I. Mazin, D. F. Agterberg, and Cesare Franchini, npj Quantum Materials **2** 37 (2017).

Bongjae Kim, Sergii Khmelevskiy, Peter Mohn, and Cesare Franchini, Phys. Rev. B **96** 180405(R) (2017).

Ab initio TDDFT study of band topology and phonon-dressed spin state of spin-orbit coupled semiconductors

Noejung Park

Department of Physics, Ulsan National Institute of Science and Technology, Ulsan, 689-798 Korea

Topological states have commonly been cited as a new classification of materials, and global properties immune to local perturbations have been suggested as topological non-trivial attributes. For real materials actual computational results of the topological quantities have been obtained through the theories of linear responses over the static ground electronic structure. Here, we propose an alternative way by considering the time-evolution of the Hamiltonian, which lets the pumping parameter run periodically through the geometric space of the Hamiltonian. As test examples of this method, we present a trivial insulator, a spin-frozen valley-Hall system, a spin-frozen Haldane-Chern insulator, and a quantum spin-Hall insulators. In later part, we also demonstrate the spin precession dynamics of MoS₂, and other semiconductor with strong SOC, in which the spin is strongly coupled to the optical phonon. This dynamical spin state can be resolved into discrete Floquet-phononic spectra, and once the phonon is pumped so as to break time-reversal symmetry, the resulting spin-Floquet structures induce net out-of-plane magnetizations in the otherwise non-magnetic 2D material.

Evolution of Fermi Arcs in a Magnetic-Field Induced Weyl Semimetal from Wannier-Function-based Tight-Binding Model.

Kyungwha Park

Department of Physics, Virginia Tech, Blacksburg VA 24061, USA

Weyl semimetals (WSMs) have a three-dimensional (3D) bulk band structure in which the conduction and valence bands meet at discrete points, i.e. Weyl points. Projections of Weyl points with opposite chirality are connected by Fermi arcs at a surface. Topological Dirac semimetals (DSMs) have 3D Dirac points which can be viewed as superimposed copies of Weyl points stabilized by rotational symmetry. When an external magnetic field is applied to a DSM, Dirac points can be separated into multiple Weyl points and so a WSM phase can be driven. DSMs and WSMs have received a lot of attention because they exhibit the chiral anomaly and novel magneto-transport signatures. We develop a tight-binding model based on Wannier functions directly from density functional theory (DFT) calculations for a topological DSM. We add spin-orbit coupling and Zeeman terms in the tight-binding model. Upon an external magnetic field along the rotational axis, we find that each Dirac node splits into two single Weyl points and two double Weyl points with opposite chirality. Our calculations also reveal interesting evolution of Fermi-arc surface states and other topological surface states as a function of chemical potential in the presence of the external magnetic field.

제 14회 고등과학원 전자구조계산 학회 Poster Session

* 포스터세션은 Banquet 장소(고등과학원 대강당 로비)에서 진행됩니다.

강기재	1-POS	Board 1
권정안	2-POS	Board 2
김도훈	3-POS	Board 3
김범섭	4-POS	Board 4
김영재	5-POS	Board 5
Thekkepat, Krishnamohan	6-POS	Board 6
Liu Kai	7-POS	Board 7
박가람	8-POS	Board 8
박선호	9-POS	Board 9
박은원	10-POS	Board 10
박재홍	11-POS	Board 11
박환열	12-POS	Board 12
변성재	13-POS	Board 13
성하준	14-POS	Board 14
송도형	15-POS	Board 15
심재훈	16-POS	Board 16
여인원	17-POS	Board 17
여현우	18-POS	Board 18
Okyay, Mahmut Sait	19-POS	Board 19
Wang, Chongze	20-POS	Board 20
유동선	21-POS	Board 21
윤영채	22-POS	Board 22
윤원석	23-POS	Board 23
윤홍기	24-POS	Board 24
이경미	25-POS	Board 25
이경풍	26-POS	Board 26
이동현	27-POS	Board 27
이미소	28-POS	Board 28
이성우	29-POS	Board 29
이승준	30-POS	Board 30
이형근	31-POS	Board 31
장보규	32-POS	Board 32
장승우	33-POS	Board 33
장승훈	34-POS	Board 34
정민용	35-POS	Board 35
정상민	36-POS	Board 36
정원석	37-POS	Board 37
진태원	38-POS	Board 38
최이철	39-POS	Board 39
한규승	40-POS	Board 40
한우현	41-POS	Board 41
홍지숙	42-POS	Board 42
강성우	43-POS	Board 43

First-principles study on K center in nitrogen-deficient amorphous Si₃N₄

Gijae Kang, Dongheon Lee, Kyeongpung Lee and Seungwu Han

Department of Materials Science and Engineering, Seoul National University, Seoul 151-744, Korea

As the consumer market for the flash memory has grown rapidly over the last decades, the needs to compensate the scaling limitation and the reliability issue of the floating gate memory have been increased significantly. One of the most promising solutions to these problems related to the charge-loss is the charge trap flash (CTF). In CTF, amorphous silicon nitride (a-Si₃N₄) is often utilized as the charge trapping layer. The localized nature of mid-gap states in a-Si₃N₄ renders the charge carriers immobile, reducing the leakage current and hence providing high performance.

It is known that the key variable governing the device performance of the charge trap memory is the trap states in the Si₃N₄ layer. The trap states of Si₃N₄ have been investigated for decades in several experimental and theoretical works, and yet the atomistic origin of the trap states in a-Si₃N₄ is still elusive. The intrinsic defects concerning the crystalline Si₃N₄ were studied comprehensively, and the relevant levels are contributed to the dangling bond or Si-Si antibonding orbital. On the other hand, the amorphous Si₃N₄ is relatively less studied, especially in the view of the characterization of the trap states.

To resolve these issues, we carry out DFT calculations on stoichiometric a-Si₃N₄ and non-stoichiometric nitrogen-deficient a-Si₃N₄ (a-Si₃N_{4-x}). Amorphous ensembles are generated using DFT molecular dynamics (MD), and the atomic configuration, electronic properties of Si₃N₄ is discussed. The charge trap states in the a-Si₃N_{4-x} is analyzed using ab-initio simulation based on DFT and hybrid functional. The majority of amorphous samples generated by melt-quench procedure has the silicon dangling bond called K center which can serve as trap states in electron or hole charging process. We found that the K center in a-Si₃N₄ gives various U including positive value, and yet the fluctuation in local environment of the K center in amorphous matrix yields seemingly negative- U behavior. Finally, the charge injection level distribution is calculated using the thermodynamic formation energy of each charged trap.

Density functional theory study on durable titanium nitride (TiN) electrocatalyst supports

Jeong An Kwon^a, Min-Su Kim^a, Dong Yun Shin^a, and Dong-Hee Lim^{*,a}

*^aDepartment of Environmental Engineering, Chungbuk National University, Cheongju, Chungbuk 28644,
Republic of Korea*

Transition metal nitrides possessing superior resistance to oxidation and corrosion have been described as good substitutes for carbon support materials in proton exchange membrane fuel cell (PEMFC) operation. In the current study, the stability and electronic properties of titanium nitride (TiN)-supported Pt in comparison with carbon-supported Pt (using graphite and graphene) has been conducted using density functional theory (DFT) calculations. A single Pt atom adsorbed more strongly to the TiN surface than to both graphite and graphene. (The adsorption energies (E_{ads}): $-3.72 \sim -3.76$ eV, $-1.50 \sim -2.20$ eV, $-0.82 \sim -1.57$ eV, respectively). By visualizing the charge transfer, it was observed that the better stability of Pt on TiN is not due to the interaction between Pt and N atoms, but to the stronger interaction between Pt and Ti atoms. The d-band centers of Pt adsorbed on TiN (-1.91 eV) were more downshifted from the Fermi level than those of Pt adsorbed on graphene (-0.97 eV), indicating that the TiN support is more durable and has more potential as a catalyst in the oxygen reduction reaction (by moderately weakening oxygen adsorption) than traditional carbon materials.

Branching ratio calculation based on first principles for $5d$, $4d$ and $3d$ transition metals

Do Hoon Kim, Jae-Hoon Sim, Hongkee Yoon and Myung Joon Han*

Department of Physics, Korean Advanced Institute of Science and Technology (KAIST), Daejeon 34141, Korea

The branching ratio is generally used to estimate spin-orbit coupling strength in X-ray absorption spectroscopy. We present the calculation results of spin-orbit coupling and branching ratio for a variety of $5d$, $4d$, and $3d$ transition metal materials based on first principles. The calculated branching ratios of $5d$ and $4d$ transition metal materials are in good agreement with the experimental data. For $3d$ transition metals, the difference between the calculation and experiments can be understood by core-valence interaction whose energy scale is competing to spin-orbit coupling of p orbitals. These results support a theoretical understanding the relation between branching ratio and spin-orbit coupling.

An investigation of the shift current using time-dependent density functional theory

Bumseop Kim¹, Dongbin Shin¹, Jeongwoo Kim¹, Mahmut Sait Okay¹, Noejung Park^{1,2}

¹Department of Physics, Ulsan National Institute of Science and Technology, Ulsan, 689-798 Korea

²Max Planck Institute for the Structure and Dynamics of Matter, Luruper Chaussee 149, 22761 Hamburg, Germany

The bulk photovoltaic effect (BPVE) characterized by the generation of a steady photocurrent without the aid of external p-n junction has attracted a lot of attention due to its novel physics and high performance as a solar cell device. In noncentrosymmetric materials, the asymmetric wave function enables the spontaneous charge separation so called the shift current. In this presentation, we study the underlying physics of the shift current developed in ferroelectric materials using two different methodologies; 1) conventional density function theory (DFT) calculations and 2) real time propagation time dependent density functional theory (rtp-TDDFT) calculations. Using the Su-Schrieffer-Heeger model, we show the validity of TDDFT study for the shift current calculation in comparison with the well-established DFT scheme. We also discuss the possibility of the application of TDDFT in practical examples.

Subfemtosecond optical processing with the wide-gap semiconductor band engineering

Youngjae Kim and J.D. Lee*

Department of Emerging Materials Science, DGIST, Daegu 42988, Korea

**Corresponding author: jdlee@dgist.ac.kr*

It is theoretically indicated from the first-principles and model calculations that, in a wide-gap semiconductor AlN plate, the petahertz (PHz, 10^{15} Hz) current induced at a rate of the light frequency of the applied few-cycle optical waveform can be precisely controlled depending on the band distortion engineered by the strain along the [0001] direction of wurzite AlN. With the findings, we propose a prototype of the solid-state petahertz device enabling the subfemtosecond volatile or nonvolatile signal processing with the band engineering.

Towards a Stable Rare-earth Free Permanent Magnet: The stability of Fe₁₆N₂

Krishnamohan Thekkepat^{1,2}, Satadeep Bhattacharjee³, Mahesh Chandran³, Pil-Ryung Cha⁴,
Jung-Hae Choi¹ and Seung-Cheol Lee^{1, 2, 3*}

¹ *Center for Electronic Materials, Post-Silicon Semiconductor Institute, Korea Institute of Science and Technology, Seoul 02792, Republic of Korea*

² *Division of Nano & Information Technology, KIST school, Korea University of Science and Technology, Seoul 02792, Republic of Korea*

³ *Indo-Korea Science and Technology Center, Jakkur, Bangalore 560065, India*

⁴ *School of Advanced Materials Engineering, Kookmin University, Seoul 02707, Republic of Korea*

Permanent magnets are ubiquitous in information storage and energy conversion applications. Currently, the most widely used permanent magnets are Nd-Fe-B and Sm-Co alloys, which rely mostly on rare-earth elements^[1,2]. Less reliance on rare-earth based permanent magnets is important due to strategic concerns based on cost and supply of rare-earth elements. Fe₁₆N₂, a promising alternative to rare-earth based permanent magnets, has a very limited applicability due to its poor thermal stability^[3,4,5]. In this work, using density functional theory, we investigate the effect of alloying Fe₁₆N₂ with 3d and 4d transition group elements on its formation energy and magnetic properties. Using a systematic screening procedure, we propose V as an excellent alloying element that improves both the thermal stability and magnetocrystalline anisotropy energy of Fe₁₆N₂. Our work demonstrates that alloying Fe₁₆N₂ with V improves its MAE by 20 % in addition to making it suitable for high temperature applications. Synergistic improvement in both these performance parameters has not been reported so far.

References:

- [1]. Gutfleisch, O., Willard, M.A., Brück, E., Chen, C.H., Sankar, S.G. and Liu, J.P., *Advanced materials* 23(7), 821 (2011).
- [2]. Coey, J.M.D., *Scripta Materialia* 67(6), 524 (2012).
- [3]. Kim, T.K. and Takahashi, M., *Applied Physics Letters* 20(12), 492 (1972).
- [4]. Coey, J.M.D., *Journal of Applied Physics* 76(10), 6632 (1994).
- [5]. Sims, H., Butler, W.H., Richter, M., Koepf, K., Şaşıoğlu, E., Friedrich, C. and Blügel, S., *Physical Review B* 86(17), 174422 (2012).

Oxygen adsorption on Ge [001] nanowire

Kai Liu^{1,2}, Eunjung Ko¹, Cheol Seong Hwang², and Jung-Hae Choi^{1*}

¹ *Center for Electronic Materials, Korea Institute of Science and Technology, Seoul 02792, Korea*

²*Department of Materials Science and Engineering, and Inter-university Semiconductor Research Center, Seoul National University, Seoul 151-744, Korea*

E-mail: choijh@kist.re.kr

In recent years, with silicon-based field-effect transistor (FET) technology approaching its physical limits, semiconductor manufacturers have been searching for the non-silicon based materials that have high mobility charge carriers to replace the current silicon-based transistor channel [1]. Ge nanowire has been attracting a lot of research interest due to its potential for high carrier mobility [2]. The improvements on the Ge NW-based FET will be critically important for application of Ge in new generation transistors [3,4].

In this study, we perform ab-initio calculation on the effects of oxygen adatoms on Ge nanowire with various diameter along the [100] orientation. The atomic structures and electronic properties of Ge nanowire with the adsorbed oxygen atoms are characterized. The bonding configurations of oxygen adsorption on Ge nanowire surface are revealed, and differences in electronic structure, such as local density of states (LDOS) and dynamics of oxygen atom & molecule adsorption process, are specifically discussed. A detailed knowledge of the reaction of oxygen with Ge nanowire may provide the way toward a more effective next-generation switching device surface configuration.

- [1] Nature, Vol. 479, 2011:324–328.
- [2] J. Appl. Phys. 118, 2015:225302.
- [3] J. Appl. Phys. 108, 2010:084108.
- [4] Applied Physics Letters 102(10), 2013: 102106.

Van der Waals approximation: interaction between surface and molecule

Karam Park, Sukmin Jeong

Department of Physics and Research Institute of Physics and Chemistry, Chonbuk National University, Jeonju 54896, Korea

Van der Waals (vdW) interaction in the density functional theory (DFT) is relatively well described by the repulsion force (Pauli term) of Lennard-Jones potential, but attraction force (vdW term) has errors that are not well represented even with some hybrid functionals.

Various type of attraction terms are added into the exchange-correlation functionals are found to work effectively for several molecule.

In understanding two-dimensional material growth precise description of both molecular-molecular and surface-molecular interaction is very important however the use of the initial form of the attractive terms causes large errors.

We study relative validity of the vdW approximation for the interaction between the Pt(111) surface and the hexagonal molecules (benzene, borazine) used in 2D material growth.

We investigate the energetics and structures using local density approximation (LDA) and generalized gradient approximation functional (PBE) including van der Waals (vdW) interactions (D2 ^[1], D3 ^[2], dDsC ^[3], TS ^[4]).

As a result, in the LDA case, large errors occur in describing adsorption structures and energies. In case of normal PBE, the adsorption energy clearly varies for each type of vdW interaction, but the atomic structures and molecular orbitals are similar for all vdW interactions.

In this paper, we present our analysis for the different behaviors of the vdW approximations.

[1] S. Grimme., J. Comp. Chem. **27**, 1787 (2006).

[2] S. Grimme., J. Chem. Phys. **132**, 154104 (2010).

[3] S. N. Steinmann., J. Chem. Phys. **134**, 044117 (2011).

[4] A. Tkatchenko., Phys. Rev. Lett. **102**, 073005 (2009).

First-Principle Study on Triazine and Heptazine-based Graphitic Carbon Nitride Structures

Sunho Park, Hosin Song, Min-Gwan Kim, Seoung-Hun Kang and Young-Kyun Kwon

Department of Physics and Research Institute for Basic Sciences, Kyung Hee University

We investigate the structural and electronic properties of graphitic carbon nitrides (g-CNs) in sheet and tube forms using a first-principles calculations based on density functional theory. Graphitic carbon nitride in the composition of C_3N_4 can form not only a porous 2D layer in a honeycomb lattice, but also carbon nitride nanotubes(CNNTs) made by rolling its single layer. We considered triazine and heptazine structures of g- C_3N_4 , which naturally contains pores or vacancies. The triazine structure has three C atoms cross-linked by four N atoms in a unit cell, and the heptazine counterpart is formed by a doubled unit cell. We found that the g- C_3N_4 is stabilized by forming a curved sheet with local buckling from its planar form, and further stabilized by forming a CNNT specified with a certain chirality. Such an interesting structural deformation can be elucidated by an electron lone pair belonging to each two-coordinated nitrogen atom existing near vacancies. These g-CN based materials, which are semiconducting with a direct band gap, can be a good adsorbent for various molecules or chemicals easily attached to near such vacant sites. Thus they can be a potential candidate for electronic, optical, and sensing devices for future nanotechnology.

Intrinsic and Extrinsic Defects (Au, O) in MoTe₂ Few Layers

Eun-Won Park¹, Jung-Min Hyun¹, Miyoung Kim, and Hanchul Kim^{1*}

¹ *Department of Physics, Sookmyung Women's University, Seoul, Korea*

*hanchul@sookmyung.ac.kr

Transition metal dichalcogenides (TMDCs) are layered compounds made up of X-TM-X trilayers, where TM and X refer to transition metal atom and chalcogen atom, respectively. MoTe₂ is one of the representative TMDCs and has been studied extensively due to its high current on/off ratio and appropriate band gap for field-effect transistors and integrated circuits [1-5]. In a previous study, intrinsic defects in MoTe₂ monolayer were investigated systematically and the Te adatom on top of a Te atom was found to be the most stable defect [6]. In this work, we studied intrinsic and extrinsic defects in MoTe₂ few layers. We performed density functional theory (DFT) calculations by using Vienna ab-initio simulation package. We used the PBE exchange-correlation functional in conjunction with the DFT-D3 method to describe the van der Waals interaction between the trilayers. We used the 5×5×3 and 5×5×2 supercells to simulate isolated intrinsic defects and extrinsic defects, respectively. For intrinsic defects, we investigated vacancies (V_{Mo} , V_{Te}), adatoms (Mo_{ad} , Te_{ad}), interstitials (Mo_{I} , Te_{I}), intercalations (Mo_{ic} , Te_{ic}), and antisites (Mo_{Te} , Te_{Mo}). The most stable structures are V_{Mo} in Te-rich condition (3.26 eV) and Mo_{I} in Mo-rich condition (2.70 eV) for Mo defects and Te_{ad} on top of a Te atom in both Te-rich and Mo-rich conditions (1.06 eV and 1.35 eV) among Te defects. For extrinsic defects, we took Au and O related defects and examined adatoms (Au_{ad} , O_{ad}), interstitials (Au_{I} , O_{I}), intercalations (Au_{ic} , O_{ic}), and substitutions (Au_{Mo} , Au_{Te} , O_{Mo} , and O_{Te}). The most stable structures are Au_{ic} at hexagonal site in Te-rich condition (1.18 eV) and Au_{Te} facing the van der Waals gap in Mo-rich condition (1.17 eV) for Au defects and O_{Te} at the in both Te- and Mo-rich conditions (1.78 eV and 2.04 eV) for O defects.

Reference

1. M. Xu *et al.*, Chem. Rev. **113**, 3766 (2013)
2. D. H. Keum *et al.*, Nat. Phys. **11**, 482 (2015)
3. M. Chhowalla *et al.*, MRS Bulletin **40**, 585 (2015)
4. R. Kappera *et al.*, Nat. Mater. **13**, 1128 (2014)
5. S. Cho *et al.*, Science **349**, 625 (2015)
6. S. Haldar *et al.*, Phys. Rev. B **92**, 235408 (2015)

Intrinsic electric polarization and its switching mechanism in 2D transition metal dichalcogenides having polar symmetry

Jaehong Park^{1,2}, Cheol Seong Hwang^{2,3}, and Jung-Hae Choi^{1*}

¹Center for Electronic Materials, Korea Institute of Science and Technology, Seoul 02792, Korea

²Department of Materials Science and Engineering, Seoul National University, Seoul 08826, Korea

³Inter-university Semiconductor Research Center, Seoul National University, Seoul 08826, Korea

Spontaneous electric polarization is an important physical property associated with various phenomena such as pyro-, piezo- and ferroelectricities. In contrast to the bulk materials of 3D crystalline structures, the intrinsic polarization is rarely reported in the 2D crystals[1]. In this presentation, we investigate the spontaneous polarization of various 2D transition metal dichalcogenides MX_2 ($\text{M}=\text{Mo}, \text{W}$, $\text{X}=\text{S}, \text{Se}, \text{Te}$) based on density functional theory calculations. We show the dependence of the lattice structures and magnitude of polarizations on various exchange-correlation functionals from the conventional LDA to the state-of-art metaGGA in conjunction with van der Waals corrections. Further, we investigate the possibility of polarization reversal by certain zone-center phonon mode resulting in the interlayer sliding based on Landau theory.

[1] Nat. commun. 8, 14956 (2017).

Ab initio study of the reactivity of B₂H₆ molecule on TiN Surfaces for Tungsten Atomic Layer Deposition

Hwanyeol Park¹, Sungwoo Lee¹, Ho Jun Kim², Daekwang Woo³, Euijoon Yoon^{*1}, Gun-Do Lee^{*1}

¹*Department of Materials Science and Engineering, Seoul National University, Korea*

²*Department of Mechanical Engineering, Dong-A University, Busan 49315, South Korea*

³*Memory Thin Film Technology Team, Giheung Hwaseong Complex, Samsung Electronics, 445-701, South Korea.*

In the fabrication process of memory devices, a void-free tungsten (W) gate process with good conformability is very important for improving the conductivity of the W gate, leading to enhancement of device performance. As the downscaling continues to progress, void-free W deposition becomes more difficult due to the experimental limitations of conformal film deposition even with atomic layer deposition (ALD) W processes. In ALD W processes, it is known that the B₂H₆ dosing process plays a key role in deposition of the ALD W layer with low resistivity and in removal of residual fluorine (F) atoms. To comprehend the detailed ALD W process, we have investigated the dissociation reaction of B₂H₆ on three different TiN surfaces, TiN (001), Ti-terminated TiN (111), and N-terminated TiN (111), using first-principles density functional theory (DFT) calculations. N-terminated TiN (111) shows the lowest overall reaction energy for B₂H₆. These results imply that severe problems, such as a seam or void, in filling the W metal gate for memory devices could be attributed to the difference in the deposition rate of W films on TiN surfaces. From this study, it was found that the control of the texture of the TiN film is essential for improving the subsequent W nucleation.

[1] Hwanyeol Park *et al.*, RSC Adv., 7 (2017) 55750

Strain effect on the electronic properties of Copper doped CdSe nanoplatelet

SeongJae Byeon¹, Yong-Hoon Kim^{1,*}

¹ *Department of Electrical Engineering, KAIST, 291 Daehak-ro, Yuseong-gu, Daejeon, Korea*

E-mail: y.h.kim@kasit.ac.kr

Cadmium Selenide (CdSe) nanoplatelets(NPL) are appealing due to their anisotropic growth controlled at the atomic scale and optical properties. However, to the best of our knowledge, not much work has studied the doping of the CdSe NPL. Herein, by means of density functional theory (DFT) calculation, we investigate the geometrical and electronic properties of the copper(Cu)-doped CdSe NPL at the atomic level. In particular, we study the strain effect on the electronic properties of the 3.5 monolayer Cu-doped zinc-blend(ZB) CdSe NPL capped with oleic acid organic ligand. The strain effect is indeed shown to change the Cu mid-gap states which are located near the fermi-level. To understand the origin of Cu states shift under strain, we analyze the local density of states for Cu-doped CdSe NPL and find that Jahn-Teller distortion plays an important role. On the basis of our results, we further show that the Cu states are shifted with ligand-induced strain upon exchange of oleic acid with thiol acid.

A novel superconducting Si allotrope and its high pressure chemical precursor

Ha-Jun Sung,¹ W.-H. Han,¹ In-Ho Lee,² and Kee Joo Chang^{1*}

¹*Department of Physics, Korea Advanced Institute of Science and Technology, Daejeon 34141, Korea*

²*Korea Research Institute of Standards and Science, Daejeon 34113, Korea*

The thermodynamic ground state of silicon is a cubic diamond structure at ambient pressure. Cubic diamond Si (*c*-Si) is a semiconductor with an indirect band gap that is the basis of modern semiconductor technology. Silicon forms various metallic under pressure and metastable semiconducting phases can be obtained after pressure release from metallic phases. Other approach to obtain novel Si allotropes is to use their chemical precursors synthesized at high temperature and high pressure conditions. Recently, *Cmcm*-Si₆ with a quasi-direct band gap of 1.3 eV was synthesized from a high-pressure NaSi₆ precursor followed by thermal degassing process. Although, numerous metastable Si allotropes can exist at ambient pressure, metallic Si phases have not been reported to date.

In this work, we report two novel metallic clathrates of silicon at ambient pressure [1], *P6/m*-Si₆ and *P6/m*-NaSi₆ allotropes. Using the evolutionary crystal structure search method at high pressure [2], we find that the *P6/m*-NaSi₆ clathrate can be synthesized at pressures above 12.4 GPa. A novel *P6/m*-Si₆ clathrate can be obtained from the *P6/m*-NaSi₆ precursor by the Na degassing process. We find that both *P6/m*-Si₆ and *P6/m*-NaSi₆ allotropes are stable and superconducting at ambient pressure, with the superconducting temperatures of 13.1 and 12.2 K, respectively. Our prediction of a novel Na-Si clathrate will stimulate experimentalists to synthesis various clathrate structures at high pressure.

References

- [1] H.-J. Sung, W. H. Han, I.-H. Lee, and K. J. Chang, *Phys. Rev. Lett.* 120, 157001 (2018).
- [2] I.-H. Lee, Y. J. Oh, S. Kim, J. Lee, and K. J. Chang, *Comp. Phys. Commun.* 203, 110 (2016).

Band Structure Modification of Cd_nSe_n Quantum Dots through External Electric Field: *Ab-initio* Studies

Dohyung Song¹, Jaichan Lee¹

¹*School of Advanced Materials Science and Engineering, Sungkyunkwan University, Suwon 16419, Republic of Korea*

Quantum dots (QDs) have been actively studied for their interaction with other materials due to their specific optical and electrical properties. In particular, we studied the change of the energy band structure of CdSe QDs under an external electric field in order to consider the situation where the electric characteristics change due to the effect of the electric field generated when the CdSe QDs is used in the device. We found that, the external electric field changes the energy band structure of CdSe QDs, due to the internal dipole moment.

In this study, we used density functional theory (DFT) to investigate the energy band structure and the process of change with an artificial external electric field. As a result, we verified that the CdSe QDs in wurtzite structure change of the CdSe QDs in wurtzite structure due to the external artificial electric field, in Cd_nSe_n QDs, n varied up to 33 with the corresponding diameters from 0.457 to 1.38 nm and the electric field from -0.1 to 0.1 V/Å range. When the electric field reaches at a critical value, the energy band gap of the QDs becomes significantly narrower than pristine case. We expect that band structure modification by the electrical field will provide the estimation of influence of physical properties when applying QDs to electrical devices.

Implementation of the DFT+DMFT: Natural atomic orbitals-based construction of the correlated subspace

Jae-Hoon Sim¹ and Myung Joon Han^{1,*}

¹Department of Physics, Korea Advanced Institute of Science and Technology (KAIST), Daejeon, Korea

E-mail address: jh_sim@kaist.ac.kr, mj.han@kaist.ac.kr

The strongly correlated electron system is of central importance in the condensed matter physics, exhibiting high-temperature superconductivity, correlation induced metal-insulator transition. With the fast progress in the experimental techniques for study novel materials, it is crucial to provide an appropriate theoretical formalism that can handle many-body effects in a material-specific context. While the standard density functional theory (DFT) is a successful method to describes real materials, correlated materials are not properly treated with this method. One of the successful approach to study these systems is the dynamical mean-field theory (DMFT) combined with the DFT [1, 2]. We implemented DFT+DMFT based on the non-orthogonal pseudo-atomic orbitals (PAOs) basis DFT code, OpenMX [3]. In our implementation, hybridization function is defined over a wide energy range, including ligand orbitals, e.g., oxygen *p* orbitals. The projector onto the strongly correlated subspace can be defined by given PAOs. While the different orthogonalization and projection methods are tested, the natural atomic orbitals, which form an orthogonal basis, have been shown to be stable to numerical parameter change [4]. Our implementations are applied to the typical transition metal oxide systems.

References

- [1] A. Georges and G. Kotliar, Phys. Rev. B **45**, 6479 (1992).
- [2] A. Georges et al., Rev. Mod. Phys. **68**, 13 (1996); G. Kotliar et al., Rev. Mod. Phys. **78**, 865 (2006)
- [3] <http://www.openmx.org>.
- [4] A. E. Reed, R. B. Weinstock, and F. Weinhold, J. Chem. Phys 83(2), 735 (1985).

Equilibrium crystal shape of GaAs and InAs by ab-initio thermodynamics

In Won Yeu^{1,2}, Gyuseung Han^{1,2}, Cheol Seong Hwang², and Jung-Hae Choi^{1*}

¹*Center for Electronic Materials, Korea Institute of Science and Technology, Seoul 02792, Korea*

²*Department of Materials Science and Engineering, and Inter-university Semiconductor Research Center, Seoul National University, Seoul 08826, Korea*

E-mail: choijh@kist.re.kr

The growth of the III-V compound semiconductors on Si is crucial to adapt their superior properties to the silicon-based CMOS technology. In order to achieve the high-quality hetero-epitaxy preventing defects such as dislocation, crack, and anti-phase boundary, the selective area growth (SAG) method has been developed. For this process, the atomic scale understanding is essential in terms of both the thermodynamics and kinetics. In this study, we establish a thorough methodology to calculate the surface energy as a function of temperature (T) and pressure (P) for the III-V compound materials in the combined framework of density functional theory (DFT) calculations and thermodynamics [1], which is competitive compared the previous DFT studies providing the surface energy as a function of chemical potential. Then, we construct the equilibrium crystal shape of GaAs and InAs as a function of T and P, including the effects of surface vibration. The calculated equilibrium shapes in various T and P conditions are compared with the experimentally grown shapes and in accordance with the experimental shapes. The correspondence between the calculated equilibrium shape and experimental shapes is first shown in the practical growth conditions, which confirms that the experimental growth was in near equilibrium. This work also shows the importance of anisotropic surface vibration on the equilibrium shape of the III-V compound semiconductors.

[1] In Won Yeu *et al.*, [Sci. Rep. 7, 10691 \(2017\)](#).

Development of a first-principles approach for the finite-bias simulation and its verifications using molecule junction systems.

Hyeonwoo Yeo[†], Juho Lee[†], Han Seul Kim[†], Yong-Hoon Kim[†]

[†]Graduate School of EEWS, KAIST, 291 Daehak-ro, Yuseong-gu, Daejeon 305-701, Korea

**E-mail address: y.h.kim@kaist.ac.kr*

ABSTRACT

Describing the non-equilibrium system induced finite bias within the first-principle calculations is one of the key challenges for the development of next-generation nano-electronic devices. However, non-equilibrium Green's function (NEGF) formalism, which is known as the most general and rigorous theoretical framework, has still a limitation of non-variational properties. In this presentation, we show new approach to describe the non-equilibrium system by introducing our recently developed method named multi-space density functional theory (MS-DFT). We describe the system in which the finite bias is applied to the molecular junctions, and verify their electrical properties as comparing to the NEGF formalism. This conclusion will give the justification for MS-DFT and critical insight in constructing transport calculation.

Time-reversal broken characteristics driven by circularly-polarized external perturbation: the comparison of a rotating electric field to magnetic field.

Mahmut Sait Okyay¹, Dongbin Shin¹, Bumseop Kim¹, Kim Jeongwoo¹, Noejung Park^{1,2}

¹Department of Physics, Ulsan National Institute of Science and Technology, Ulsan, 689-798 Korea

²Max Planck Institute for the Structure and Dynamics of Matter, Luruper Chaussee 149, 22761 Hamburg, Germany

It is widely known that external magnetic field breaks time-reversal symmetry and forms a new quantum state so called Landau level. By applying a uniform magnetic field along the surface-normal direction to two-dimensional electron systems such as GaAs and graphene, the quantum Hall effects has been successfully realized. In this presentation, we show that a transverse current can be induced by an in-plane circular electric field in the absence of magnetic field. As an example system, spinless Dirac graphene in which the electronic structure is well understood is chosen to be investigated. We investigate time-varying current values in response to various type of electric fields when subjected to a dc voltage drop. We show that an applied circular electric field induces a sizeable current along the perpendicular to the DC-bias, whereas a transverse current is absent for a linear electric field. We also discuss the origin of this effective magnetic field mimicked by circular electric fields. Our results expand our fundamental understanding of time-reversal symmetry and provide a key clue to finding a new quantum state without applying magnetic field.

Competing magnetic orderings in two-dimensional hexagonal organometallic lattices

Chongze Wang¹, Seho Yi¹, Hyun-Jung Kim², and Jun-Hyung Cho^{1,*}

¹ *Department of Physics and Research Institute for Natural Sciences, Hanyang University, 17 Haengdang-Dong, Seongdong-Ku, Seoul 133-791, Korea*

² *Korea Institute for Advanced Study, 85 Hoegiro, Dongdaemun-gu, Seoul 130-722, Korea*

Metal-organic frameworks have been received increasing attentions in the fields of nanotechnology and nano-electronics, because their advantages of low cost, easy fabrication, and mechanical flexibility. A previous density-functional theory (DFT) study predicted that two-dimensional (2D) hexagonal triphenyl-lead lattice $\text{Pb}(\text{C}_6\text{H}_4)_3$ supports a topologically nontrivial quantum spin Hall (QSH) state [1]. However, the subsequent DFT study reported that topologically trivial antiferromagnetic (AFM) insulating state is more stable than the QSH state [2]. Here, using a combined DFT and tight-binding approach, we systematically investigate the 2D hexagonal triphenyl-X lattices $\text{X}(\text{C}_6\text{H}_4)_3$ [$\text{X}=\text{Si}, \text{Ge}, \text{Sn}, \text{Pb}$]. It is found that the topological trivial AFM insulating state is the ground state for all these four lattices, while the magnetic moment mainly arises from X atoms. As the atomic number of X atoms increases, the magnetic moment m of X atoms increases, but the value of m in $\text{Pb}(\text{C}_6\text{H}_4)_3$ is smaller than $\text{Sb}(\text{C}_6\text{H}_4)_3$. Due to spin-orbit coupling, the valleys' degeneracy can be lifted in these four lattices, thereby giving rise to the band gap difference ΔE_g between the K and K' points. We find that ΔE_g increases monotonously with increasing the atomic number of X atoms. The present findings further enrich our understanding of 2D hexagonal organometallic lattices.

[1] Z.F Wang *et al.* Nat. Comm. 4, 1471 (2013)

[2] H. Kim *et al.* Phys. Rev. B **93**, 041404(R) (2016).

Atomic energies of high-dimensional neural network potential

Dongsun Yoo¹, Kyuhyun Lee¹, Wonseok Jeong¹, and Seungwu Han¹

¹Department of Materials Science and Engineering, Seoul National University, Seoul 08826, Korea

will1792@snu.ac.kr

Machine learning potentials are becoming popular method for large scale simulations with the accuracy of ab initio calculations. High-dimensional neural network potential (HDNN) of Behler and Parrinello is one of the most successful machine learning potentials that have been applied to many complex systems. In HDNN and some other machine-learning potentials, the total energy of a system is approximated as a sum of local atomic energies. However, since the atomic energy is not a physical quantity, HDNN can not learn atomic energies directly from reference atomic energies, but rather infers it from the total energy (and atomic forces) of ab initio calculations. During the training process, HDNN develops arbitrary energy decomposition scheme, or atomic energy mapping, of its own. We noticed that HDNN sometimes yields unphysical atomic energies, but is able to predict atomic energy difference of two distinct local configurations reasonably. In this presentation, we address the various aspects of atomic energy mapping of HDNN with neural network potentials trained on toy models, classical potentials, and density functional theory calculations.

SNU Materials Data Center (SNUMDC): The efficient and high performance material database platform

Youngchae Yoon¹, Yong Yoon¹, Miso Lee¹, Doyeon Kim¹ and Seungwu Han¹

¹Department of Materials Science and engineering, Seoul National University, Seoul 151-747, Korea

Recently, theoretical studies based on density functional theory (DFT) have been widely used to discovery high performance functional materials. However, because of high computational cost and high complexity of the DFT calculation, the experimental researchers have struggled in using theoretical studies to design new materials. To overcome the problems, various materials database are introduced such as AFLOW [1], OQMD [2] and Materials Project [3] but they are still difficult to utilize data.

In this study, we develop the novel material database platform based on web interface, SNU Materials Data Center (SNUMDC). We design the web application which can be easily used for the researchers and operated in high performance. Additionally, we exploit REpresentational State Transfer (REST) API module for search the properties of materials for various keyword.

Reference

- [1] Curtarolo, S. et al. Comput. Mater. Sci. 58, 227-235 (2012).
- [2] Saal, J. E., Kirklin, S., Aykol, M., Meredig, B. & Wolverton, C. JOM 65, 1501-1509 (2013).
- [3] Jain, A. et al. APL Mater. 1, 011002 (2013)

Structural stability and thermoelectric performance of two-dimensional ZnPS₃ and ZnPSe₃

Won Seok Yun and J. D. Lee *

Department of Emerging Materials Science, DGIST, Daegu, 42988 Republic of Korea

**E-mail: jdlee@dgist.ac.kr*

Recently, tremendous research attention has been focused on two-dimensional (2D) semiconductors due to their abundant physical properties and promising potential applications. In this regard, searching for the novel 2D semiconducting materials with excellent performance is an extremely important issue in the field of low-dimensional materials. In the present study, we perform the first-principles electronic structure calculations to scrutinize the dynamic and thermal stabilities, transport, and thermoelectric properties of single-layer (1L) ZnPS₃ and ZnPSe₃. Consequently, we find that 1L-ZnPSe₃ is thermodynamically more stable than 1L-ZnPS₃ through the *ab initio* molecular dynamics and phonon dispersion calculations. In addition, using the deformation potential method, an electron and hole mobilities of 1L-ZnPSe₃ are calculated to be ~ 440 and ~ 400 cm²V⁻¹s⁻¹, respectively, which is comparable to that of 1L-MoS₂. Furthermore, detailed discussion of thermoelectric properties of 1L-ZnPS₃ and 1L-ZnPSe₃ will be given.

Analytic continuation via “domain-knowledge free” machine learning

Hongkee Yoon¹, Jea-Hoon Sim¹, and Myung Joon Han¹

¹*Department of Physics, KAIST, 291 Daehak-ro, Yuseong-gu, Daejeon 34141, Republic of Korea*

We present a machine learning (ML) approach to the analytic continuation which is the notorious problem in quantum many-body physics. Especially analytic continuation is applied to obtain density of states (DOS) from imaginary time Green's function calculated from quantum Monte Carlo (QMC). However, the analytic continuation is an ill-conditioned problem therefore many numerical approaches exists; such as maximum entropy method [1,2], stochastic method [3], Pade', and approximation [4]. Here we show that using modern machine learning techniques, such as convolutional neural network and stochastic gradient descent based optimizers, ML-based Green's function to DOS kernel can be realized without detailed “domain-knowledge” about previous analytic continuation approaches. Furthermore, the ML-based kernel is faster than conventional analytic continuation algorithms and more robust to noise from Green's function. Our approach to tackling ill-posed problems by data-based ML shows the applicability of ML in other ill-posed problems.

[1] Jarrell, M. & Gubernatis, J. E. Bayesian inference and the analytic continuation of imaginary-time quantum Monte Carlo data. *Physics Reports* **269**, 133–195 (1996).

[2] Bergeron, D. & Tremblay, A.-M. S. Algorithms for optimized maximum entropy and diagnostic tool for analytic continuation. *Phys. Rev. E* **94**, 023303 (2016).

[3] Sandvik, A. W. Stochastic method for analytic continuation of quantum Monte Carlo data. *Phys. Rev. B* **57**, 10287 (1998).

[4] Vidberg, H. J. & Serene, J. W. Solving the Eliashberg equations by means of N-point Padé approximants. *J. Low Temp. Phys.* **29**, 179 (1977)

Optical-Harmonic-Generation from magnetic semiconductor EuX

Kyungmee Lee¹

*¹Department of Nano and Electronic Physics, College of Science and Technology, Kookmin University, 77
Jeongneung-ro, Seongbuk-gu, Seoul, 02707, Korea*

I investigate the electronic structure and magnetic properties of EuX and EuX oxide. Using Optical–second-harmonic-generation, the magnetic europium chalcogenide semiconductor EuX are investigated. Electric polarization $E(2\omega)$, magnetization(2ω) are calculated.

The electric level and electronic structure are demonstrated through Born-Haber Cycle , First-principle self-consistent calculation and tight-bind model. Theory and experiment are compared in SHG Intensity for different magnetic field strength and temperature.

Reference)

S. Di Matteo and M. R. Norman, Magnetic ground state of Sr₂IrO₄ and implications for second-harmonic generation, Phys. Rev. B 94, 075148 – Published 24 August 2016

B.Kaminski et al. Optical second harmonic generation in the centrosymmetric magnetic semiconductors EuTe and EuSe, Phys. Review B 81, 155201(2010)

Ab-initio calculations on Aluminum dopant in SrTiO₃

Kyeongpung Lee and Seungwu Han

Department of Materials Science and Engineering, Seoul National University, Seoul 151-744, Korea

Downsizing of MOSFET-based devices meets physical limitations, and one of them is leakage current tunneling through a dielectric layer under a critical thickness. It is well known that high-k dielectrics are less vulnerable to the leakage problem than conventional ones are. Without loss of device performance, they are advantageous for further miniaturizing. As a representative high-k material, SrTiO₃ (STO) has a large dielectric constant of about 300. However, because of its n-type nature as well as a bandgap of 3.25 eV, Schottky barrier between the metal-STO interface is small which leads to large leakage current. It is repeatedly reported that n-type nature of STO stems from thermodynamically stable oxygen vacancy which is electron donor [1]. To overcome this problem, it is one of the strategies that acceptor doping can reduce the carriers and enlarge the Schottky barrier. For instance, Paek et al. introduced Fe³⁺ or Cr³⁺ ions at Ti⁴⁺ sites which reduce the leakage current by one order [2]. In this manner, Al³⁺ dopants at Ti⁴⁺ site are also helpful. It is reported that leakage current in TiO₂ is reduced by doping Al [3], which has similar electronic structure to STO. Posadas et al. verified that Al doping over 10% enlarges bandgap of STO both experimentally and theoretically [4]. At this moment, it is in need to reveal the defect configuration which can lower the Fermi level against the stable donors, where the dopant concentration is dilute so that the dielectric properties are not degraded. In this study, Al dopant in STO is revisited, whose defect formation energies are calculated and compared to the intrinsic defects such as oxygen vacancies. Based on them, the concentration of each defect configuration is determined and the position of Fermi level is found. As a result, it is elucidated whether Al dopant reduces n-type nature of STO.

[1] A. Janotti et al., Phys. Rev. B 90, 085202 (2014)

[2] S. H. Paek et al., J. Mater. Sci. 33, 1239-1242 (1998)

[3] S. K. Kim et al., Adv. Funct. Mater. 20, 2989-3003 (2010)

[4] A. B. Posadas et al., Appl. Phys. Lett. 103, 142906 (2013)

First-principles study of metal-doped amorphous Ge₂Sb₂Te₅

Dongheon Lee¹, Gijae Kang¹, Kyeongpung Lee¹, Sungjoon Yoon², Jinwoo Kim² and Seungwu Han^{1,*}

¹*Department of Materials Science and Engineering and Research Institute of Advanced Materials, Seoul National University, Seoul 08826, Republic of Korea*

²*R&D Division, SK Hynix Inc., 2091, Gyeongchung-daero, Babul-eup, Icheon-si, Gyeonggi-do 17336, Republic of Korea*

**Corresponding Authors: hansw@snu.ac.kr*

Keywords: phase change materials, post-transition metal, doping, amorphous, GST, first-principles calculations, ab-initio molecular dynamics

The effects of post-transition metal dopants M (M = Al, Zn, and Ga) on structural and electronic properties of amorphous Ge₂Sb₂Te₅ (a-GST) were investigated through first-principles calculations. The doped a-GST were obtained by melt-quench simulations. We found that all dopants M were mainly surrounded by Te atoms, had tetrahedral geometry, and scarcely bonded to each other due to its cationic character. Local structures of dopants M in doped a-GST are similar with those in crystalline phase M_xTe_y. Bond lengths of Ge-Te and Sb-Te are little changed by doping. The number of wrong bonds like Ge-Ge, Ge-Sb, or Sb-Sb increases since metal dopants take up lots of Te atoms. It also leads to increasing the number of tail states localized at wrong bonds. Here, we came up with a method to extract mobility edges from DOS because any robust methods have not been developed. The method was validated by comparing mobility gap from DOS with the optical gap from Tauc model. The mobility gap and the optical gap increase by doping, which is in good agreement with experiments.

Screening Non-oxide High- k Materials by High-throughput *Ab Initio* Calculations

Miso Lee¹, Yong Youn¹, Kanghoon Yim², and Seungwu Han^{1,*}

¹ *Department of Materials Science and Engineering and Research Institute of Advanced Materials, Seoul National University, Seoul, 08826, Korea*

² *Korea Institute of Energy Research, Daejeon, 34129, Korea*

E-mail: opusg514@snu.ac.kr

High- k dielectrics, materials having a large band gap (E_g) and high dielectric constant (k) simultaneously, constitute critical components in microelectronic devices. Because of the inverse relationship between E_g and k , materials with large values in both properties are rare. Therefore, massive databases on E_g and k will be useful in identifying optimal high- k materials. While experimental and theoretical data on E_g and k of oxides are accumulating, corresponding information is scarce for non-oxide dielectrics with anions such as C, N, F, P, S, and Cl. To identify promising high- k dielectrics among these material groups, we screen 869 compounds of binary carbides, nitrides, sulfides, phosphides, chlorides, and fluorides, through automated *ab initio* calculations. Among these compounds, fluorides exhibit an E_g - k relation that is comparable to that of oxides. By further screening over ternary fluorides, we identify fluorides such as BiF₃, LaF₃, and BaBeF₄ that could serve as useful high- k dielectrics.

Evidence for Covalent Interaction in WS₂/Graphene van der Waals Heterostructure

Sungwoo Lee¹, Se-Yang Kim², Euijoon Yoon¹, Soon-Yong Kwon², Gun-Do Lee¹

¹*Department of Materials Science and Engineering, Seoul National University, Korea*

²*Department of Materials Science and Engineering, UNIST, Korea*

Vertically stacked two-dimensional (2D) heterostructures are promising for novel applications. Their properties and performances are profoundly affected by their stacking and interlayer geometries. Recently, WS₂ flakes were directly synthesized on polycrystalline CVD graphene template to present the significant effect of defects of the template layer to the subsequent layer growth and intrinsic properties of heterostructure. We can infer that defects of underlying template have a strong influence on the interlayer interaction between stacking layers through distinctively different deformability, thermal stability and beam damage behavior. We have tried to explain these phenomena using density functional theory calculations. Among several candidates, defective graphene with grain boundary was to be found to have covalent bonding with WS₂ flakes through W atomic bridge. Attributed to the larger orbital overlap compared to van der Waals contacts of general 2D heterostructures, this model allows to explain different intrinsic properties including high conductivity. This new defect-mediated heterostructure and its novel properties could extend the versatile possibilities for designing 2D heterostructures with desirable properties.

Novel two-dimensional van der Waals materials: layered IV-V compounds

Seungjun Lee and Young-Kyun Kwon

Department of Physics and Research Institute for Basic Sciences, Kyung Hee University

Since the successful exfoliation of graphene from graphite, a variety of two-dimensional (2D) van der Waals (vdW) materials have also emerged and extensively studied over the last decade. Nevertheless there have been continuing demands for new 2D vdW materials that exhibit interesting physical properties. Here, we propose layered IV-V compounds as novel 2D vdW materials based on first-principles calculations. Our predicted IV-V compounds are in the form of A_2B_2 , where A and B are elements in group IV (C, Si, Ge, Sn) and group V (N, P, As, Sb, Bi), forming the same crystal structure as those of layered III-VI compounds, such as GaSe or InSe. Our investigation reveals that these newly proposed IV-V compounds have two stable distinct phases, one with the mirror symmetry and the other with the inversion symmetry. To explore their physical properties, we evaluate their phonon dispersion relations; electronic band structure; transport properties; and estimate activation barrier of the phase transitions. Furthermore, we describe the local Rashba spin splitting and topological non-trivial properties found in some of our structures containing heavy elements.

Revisiting of molecular J_{eff} band structure in lacunar spinel compounds through DFT+ U with charge-dependent exchange-correlation functional

Hyunggeun Lee¹, Min Yong Jeong¹, Jae-Hoon Sim¹, Hongkee Yoon¹ and Myung Joon Han¹

¹*Department of Physics, KAIST, 291 Daehak-ro, Yuseong-gu, Daejeon 34141, Republic of Korea*

After the discovery of the $J_{\text{eff}} = 1/2$ Mott ground state in Sr_2IrO_4 [1], spin-orbit entangled J_{eff} state has attracted a great deal of attention in condensed matter physics because various quantum exotic phases such as unconventional superconductivity [2] and quantum spin liquid [3] can expect from J_{eff} state. The recent study predicted the J_{eff} Mott insulating ground state in the lacunar spinel compounds, GaM_4Se_8 ($M = \text{Nb, Mo, Ta and W}$) [4]. The molecular t_2 orbitals in the lacunar spinel compounds have same symmetry with the atomic t_{2g} orbitals. Therefore, the spin-orbit coupling can split three-fold degenerate t_2 orbitals to $J_{\text{eff}} = 1/2$ doublet and $J_{\text{eff}} = 3/2$ quartet. Furthermore, the $J_{\text{eff}} = 3/2$ ground state in GaTa_4Se_8 was directly observed through the resonant inelastic X-ray scattering (RIXS) experiment [5]. Lately, it is reported that DFT+ U calculation result can be affected by a choice of exchange-correlation functional dependent to spin density or charge density [6]. Therefore, we revisit the electronic structure of the lacunar spinel compounds with charge-dependent exchange-correlation functional instead of previously used spin-dependent exchange-correlation functional. As a result, we confirmed that the J_{eff} ground states in the lacunar spinel compounds with both exchange-correlation functionals and reasonable interaction parameters. Moreover, we analyzed the J_{eff} electronic structure with respect to Hubbard U and Hund J .

References

- [1] B. J. Kim, H. Jin, S. J. Moon, J.-Y. Kim, B.-G. Park, C. S. Leem, J. Yu, T. W. Noh, C. Kim, S.-J. Oh, J.-H. Park, V. Durairaj, G. Cao, and E. Rotenberg, *Physical Review Letters* **101**, 076402 (2008).
- [2] F. Wang and T. Senthil, *Physical Review Letters* **106**, 136402 (2011).
- [3] J. Chaloupka, G. Jackeli, and G. Khaliullin, *Physical Review Letters* **105**, 027204 (2010).
- [4] H.-S. Kim, J. Im, M. J. Han, and H. Jin, *Nature Communications* **5**, 3988 (2014).
- [5] M. Y. Jeong, S. H. Chang, B. H. Kim, J.-H. Sim, A. Said, D. Casa, T. Gog, E. Janod, L. Cario, S. Yunoki, M. J. Han, and J. Kim, *Nature Communications* **8**, 782 (2017).
- [6] S. Ryee and M. J. Han, arXiv: 1709.03214 (2017).

Experimental and Theoretical study for Spin State Transition in FeO₂

**Bo Gyu Jang¹, Jin Liu², Qingyang Hu^{2,3}, Kristjan Haule⁴, Ho-Kwang Mao³,
Wendy L. Mao^{2,5}, Duck Young Kim³, and Ji Hoon Shim^{1,3,4}**

¹ *Department of Chemistry, Pohang University of Science and Technology, Pohang 790-784, Korea,*

² *Department of Geological Sciences, Stanford University, Stanford, California 94305, USA,*

³ *Center for High Pressure Science and Technology Advanced Research (HPSTAR), Shanghai 201203, China,*

⁴ *Department of Physics and Astronomy, Rutgers University, Piscataway, New Jersey 08854, USA,*

⁵ *Stanford Institute for Materials and Energy Sciences, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA*

⁶ *Department of Physics and Division of Advanced Nuclear Engineering, Pohang University of Science and Technology, Pohang 790-784, Korea*

Iron oxide is a key compound to understand the state of the deep Earth. Since a pyrite-structured FeO₂ was discovered recently by both first-principles calculation and high-pressure experiments [1], there has been an increasing interest in this new iron oxide from a geophysical point of view. Nevertheless, the electronic structure and magnetic properties of FeO₂ under high pressure are still poorly understood. Also it is controversial about the oxidation state of Fe in FeO₂ [2, 3]

In this presentation, we provide evidence and origin of a spin state transition (SST) in FeO₂ under pressure by means of synchrotron experiments and first principles calculation. The phase diagram obtained from the relaxed structure with DFT+*U* calculation cannot describe the experimental results due to the unusual intermediate spin state. We found that this intermediate spin state comes from Fe^{(2+a)+} and O₂^{(2+a)-} state which is originated from long O₂ dimer bond length (short Fe-O distance). The DFT+*U* and DFT+DMFT calculations on the structure with short O₂ dimer give correct description of the SST observed in the experiment.

From this study, we can conclude that the oxidation state of Fe in FeO₂ is +2 rather than +3. The delocalization error of DFT may cause the controversial oxidation state between the previous theoretical studies and the discrepancy between the experiment and calculation results.

32-POS Board 32

- [1] Q. Hu, D. Y. Kim, W. Yang, L. Yang, Y. Meng, L. Zhang, and H.-K. Mao, Nature (London) **534**, 241 (2016).
- [2] B. G. Jang, D. Y. Kim, and J. H. Shim, Phy. Rev. B **95**, 075144 (2017).
- [3] S. S. Streltsov *et al.*, Sci. Rep. **7**, 1 (2017).

Charge density functional plus U theory of LaMnO₃: Phase diagram, electronic structure, and magnetic interaction

Seung Woo Jang¹, Siheon Ryee¹, Hongkee Yoon¹, and Myung Joon Han^{1,2,*}

¹ *Department of Physics, KAIST, 291 Daehak-ro, Yuseong-gu, Daejeon 34141, Republic of Korea*

² *KAIST Institute for the NanoCentury, KAIST, 291 Daehak-ro, Yuseong-gu, Daejeon 34141, Republic of Korea*

We perform charge density functional theory plus U calculation of LaMnO₃. While all the previous calculations were based on spin density functionals, our result and analysis show that the use of spin-unpolarized charge-only density is crucial to correctly describe the phase diagram, electronic structure and magnetic property. Using magnetic force linear response calculation, a long-standing issue is clarified regarding the second neighbor out-of-plane interaction strength. We also estimate the orbital-resolved magnetic couplings. Remarkably, the inter-orbital e_g - t_{2g} interaction is quite significant due to the Jahn-Teller distortion and orbital ordering.

Contact: mj.han@kaist.ac.kr

Molecular films on dielectrics probed by a graphene overlayer

Seunghun Jang¹, Hyunju Chang¹, Jeong-O Lee¹, and Bernard Doudin²

1. Korea Research Institute of Chemical Technology, Daejeon, Korea.

2. University of Strasbourg, Strasbourg, France.

We investigated graphene transistors with an ultra-thin quinonmonoamines zwitterions (QZW) molecule layer sandwiched between the graphene and SiO₂ layers. Graphene layers are transferred after surface functionalization, followed by a transistor fabrication that preserves the molecules integrity. Compact 1~5 nm thick molecular films of QZW over dielectric SiO₂ are validated by x-ray and mass spectroscopies. These dipolar layers impact the transfer curve of the transistors, with hysteresis developing when cooling down the sample below 20 K. To explain the experimental findings we calculated the energetics for the rotation of QZW molecule between graphene and SiO₂ under various applied electric field, using first-principles density functional theory (DFT) calculations. DFT calculations confirm that the reverse activation energy barrier (from 2nd perpendicular configuration to parallel configuration) increases as the applied electric field increases toward to negative direction, which can explain the hysteresis developing below 20 K in experiment. This study shows that molecules lying under a graphene layer can be designed and used to impact the graphene electrical properties.

Spin-orbit coupling effect in frustrated magnetism of $J_{\text{eff}}=3/2$ lacunar spinel GaTa_4Se_8

Min Yong Jeong¹, Jae-Hoon Sim¹ and Myung Joon Han^{1*}

¹*Department of Physics, Korea Advanced Institute of Science and Technology, Korea*

Large Spin-orbit coupling (SOC) have attracted increasing interest in condensed matter physics with novel J_{eff} states [1]. SOC introduces new exchange interaction such as Kitaev exchange interaction and multipolar interaction, which can produce intriguing magnetism which triggered people to study honeycomb lattice Iridates, d^1 double perovskite and other J_{eff} compounds.

Recently, theoretical studies proven that spin-orbit coupling can drive J_{eff} states even in the molecular orbitals with t_{2g} symmetry [2]. Moreover, in the lacunar spinel GaTa_4Se_8 (GTS) molecular $J_{\text{eff}}=3/2$ ground state is observed with advanced X-ray technique, resonant inelastic X-ray scattering combined with theory [3]. GTS is the first and only example of $J_{\text{eff}}=3/2$ ground state.

While the experimentally observed negative Curie-Weiss temperature indicates the antiferromagnetic interaction, no magnetic and structure transition are observed until 4K for GTS [4]. Concerning $[\text{Ta}_4\text{Se}_4]^{5+}$ cluster, the main molecular unit of GaTa_4Se_8 , has geometrically frustrated face-centered cubic structure, strongly indicating GTS has frustrated magnetism. On the other hand, isovalent GaV_4X_8 ($\text{X}=\text{S}, \text{Se}$) is quite different to GTS because it is ferromagnetic and has trigonal distortion. The SOC strength can be a clear difference between these sister compounds. Therefore, well organized study is needed to examine the effect of SOC in GTS.

In this presentation, we performed first-principles density functional theory calculations to understand effect of SOC in structural and magnetic properties of GTS. We find that the intra- and inter-cluster exchange interactions are dominated by antiferromagnetic interaction. We also show that the strength of exchange interactions can be weakened by a trigonal distortion while SOC dislikes the distortion.

[1] B. J. Kim *et al.* *Phys. Rev. Lett.* **101**, 076402 (2008)

[2] H.-S. Kim, J. Im, M. J. Han & H. Jin. *Nat. Commun.* **5**, 3988 (2014)

[3] M. Y. Jeong, M. J. Han *et al.* *Nat. Commun.* **8**, 782 (2017)

[4] S. Kawamoto *et al.* *J. Phys.: Conf. Ser.* **683**, 012025 (2016)

Effect of spin-orbit coupling on the zigzag nanoribbon structures of Bi(111) and Sb(111) bilayers

Sang-Min Jeong¹, Seho Yi¹, Hyun-Jung Kim^{2,1}, Gustav Bihlmayer³, and Jun-Hyung Cho^{1*}

¹ *Department of Physics, Research Institute for Natural Sciences, and HYU-HPSTAR-CIS High Pressure Research Center, Hanyang University, 222 Wangsimni-ro, Seongdong-gu, Seoul 04763, Korea*

² *Korea Institute for Advanced Study, 85 Hoegiro, Dongdaemun-gu, Seoul 02455, Korea*

³ *Peter Grünberg Institut and Institute for advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany*

One-dimensional (1D) edge states formed at the boundaries of 2D normal and topological insulators have shown intriguing quantum phases such as charge density wave and quantum spin Hall effect. Based on first-principles density-functional theory calculations including spin-orbit coupling (SOC), we show that the edge states of zigzag Sb(111) and Bi(111) nanoribbons drastically change the stability of their edge structures. For zigzag Sb(111) nanoribbon, the Peierls-distorted or reconstructed edge structure is stabilized by a band-gap opening. However, for zigzag Bi(111) nanoribbon, such two insulating structures are destabilized due to the presence of topologically protected gapless edge states, resulting in the stabilization of a metallic, shear-distorted edge structure. We find that the Bi(111) nanoribbon exhibits a larger Rashba-type spin splitting at the boundary of Brillouin zone, compared to the Sb(111) nanoribbon. Interestingly, the spin texture of the edge states in the Peierls-distorted edge structure of Sb(111) and the shear-distorted edge structure of Bi(111) has all three spin components perpendicular and parallel to the edges, due to their broken mirror symmetry. The present findings demonstrate that the topologically trivial and nontrivial edge states in normal and topological insulators play crucial roles in determining their edge structures.

Overcoming the sampling bias problem in neural network potentials using Gaussian density function

Wonseok Jeong,¹ Kyuhyun Lee¹, Dongsun Yoo and Seungwu Han¹

Department of Materials Science and Engineering, Seoul National University, Seoul, 151-744, Korea

email: hansw@snu.ac.kr

The machine-learning neural-network potential (NNP) is anticipated as promising next-generation interatomic potentials. While NNP has been demonstrated to be very accurate on describing the potential energy surface (PES) of various atomic systems, we found that NNP can suffer from inaccurate description of PES even with the structures included in the training set. The training failure is caused by a fundamental sampling bias which comes from highly non-uniform distribution of training data point in input feature vector space. Due to the problem, NNPs may cause a catastrophic simulation failure and inaccurate description of physical properties. Using the Gaussian density function (GDF) that quantifies the sparsity of training points, we propose a weighting scheme that can effectively rectify the sampling bias. Various examples confirm that the GDF weighting significantly improves reliability and transferability of NNP compared to the conventional training method, which is attributed to accurate mapping of atomic energies.

Crystal structure prediction of transition metal binary alloy: Orbital-spin occupancy rule

Taewon Jin,¹ Hyo Seok Ji,¹ Young Joo Lee², Joo Young Kim², S. K. Kwon³, Changhoon Lee^{1,4,*} and Ji Hoon Shim^{1,4,5,*}

¹*Department of Chemistry, Pohang University of Science and Technology, Pohang 37673, Korea*

²*Research Institute of Industrial Science and Technology, Pohang 37673, Korea*

³*Graduate Institute of Ferrous Technology, Pohang University of Science and Technology, Pohang 37673, Korea*

⁴*Division of Advanced Nuclear Engineering, Pohang University of Science and Technology, Pohang 37673, Korea*

⁵*Department of Physics, Pohang University of Science and Technology, Pohang 37673, Korea*

Prediction of structural phase in the composition of various transition metals is highly required because the material properties are deeply related to the crystal structures [1]. In the early pioneering work of Skriver, the d -orbital occupancy, n_d , has been suggested as a simple descriptor to predict the structural phase of transition metal composition in nonmagnetic ground state [2]. As an extension of previous works, we suggest an orbital-spin occupancy rule using two descriptors n_d and σ_d (spin moment) to predict crystal structure for pure transition metal [3]. We also apply the orbital-spin occupancy rule to predict the crystal structures of 200 kinds of transition metal binary alloys in random phase and ordered phase. The crystal structures of 95% binary alloys are correctly predicted using the orbital-spin occupancy rule. We also found that the Slater-Pauling curve can be used in the crystal structure prediction of the $3d$ - $3d$ binary alloys [4, 5], while the modified Slater-Pauling curve is needed in various $3d$ - $4d$ ($5d$) binary alloys.

[1] V. Lota, J. H. P. Klepeis, and C. S. Yoo Appl. Phys. Lett. **90**, 042505 (2007).

[2] H. L. Skriver, Phys. Rev. B **31**, 1909 (1985).

[3] T. Jin, *et al.*, AIP Advances (Submitted)

[4] J. C. Slater, Phys. Rev. **49**, 537 (1936).

[5] L. Pauling, Phys. Rev. **54**, 899 (1938).

Identification of intrinsic surface defects in a thin PtSe₂ film from first-principles

Presenter : Yichul Choi

Affiliation : Virginia Tech

Authors : Fazel Baniasadi, Husong Zheng, Yichul Choi, Chenggang Tao, and Kyungwha Park

Abstract : Among transition-metal dichalcogenide families, PtSe₂ is relatively new. In a bulk form, it was confirmed to be a Dirac semimetal, whereas as a monolayer, it was shown to be semiconducting with a small band gap. Recently, a PtSe₂ monolayer was grown on a Pt substrate and revealed interesting layer-selective Rashba splitting. Compared to MoS₂ family, PtSe₂ has a stronger bonding between neighboring monolayers, which allows to observe defects slightly deeper into the surface. Very recently, a scanning tunneling microscopy and spectroscopy (STM/S) study revealed interesting morphology changes depending on types of intrinsic surface defects on a thin PtSe₂ film grown on sapphire. Motivated by this, we investigate morphology, formation energies and electronic structures of five distinct surface defects in a thin PtSe₂ film, by using supercells within density-functional theory. Our results are compared with the STM/S experimental data.

Phase diagram and electronic structures of Ga(As,Sb) solid solution using cluster expansion method

Gyuseung Han^{1,2}, Mahesh Chandran³, Seung Cheol Lee³, Cheol Seong Hwang², and Jung-Hae Choi^{1*}

¹ *Center for Electronic Materials, Korea Institute of Science and Technology, Seoul 130-791, Korea*

² *Department of Materials Science and Engineering, Seoul National University, Seoul 151-744, Korea*

³ *Indo-Korea Science and Technology Center, Bengaluru 560065, India*

Ga(As,Sb) solid solution has been studied as one of the promising next-generation semiconductor channel materials. However, the studies on Ga(As,Sb) energetic property shows ambiguous results [1][2][3]. This is due to the characteristics of a solid solution. A solid solution is the chemical mixture of some components and forms a single phase in the composition of a certain range. In addition, many physical and electronic properties of the solid solution are related to atomic configuration [4][5]. Depending on experimental conditions, some atomic configuration can appear more often than others [6][7]. This leads the diverse results among previous experimental studies. Ab-initio calculations are able to calculate the properties of a limited number of atomic configurations due to the large computational cost. On the other hand, when we assume that materials properties come from the interaction between atoms, the properties of materials is the sum of the contribution of interactions of some atomic groups, which are called clusters. Cluster expansion method predicts the properties of any atomic configuration by calculating the contribution of each interaction on properties. Predicted properties of each configuration are combined into properties of materials by the canonical ensemble and grand canonical ensemble. In this study, the phase diagram and bandgap of Ga(As,Sb) are calculated by the combined methods using ab-initio calculation, cluster expansion, canonical ensemble and grand canonical ensemble and the calculated results are compared with the previous experimental results.

[1] J. Electron. Mater. 8, 1 (1979)

[2] J. Less-Common met., 142(1988) 135

[3] Jour. Appl. Phys. 59, 2728 (1986)

[4] Am. J. Opt. Photonics 4, 4, 32 (2016).

[5] Nano Lett. 14, 455 (2008).

[6] Appl. Phys. Lett. 85, 5589 (2004)

[7] Appl. Phys. Lett. 48, 1603 (1986)

Green phosphorus with tunable direct band gap and high electron mobility

Woo Hyun Han¹, Sunghyun Kim², In-Ho Lee³, and Kee Joo Chang¹

¹*Department of Physics, Korea Advanced Institute of Science and Technology, Daejeon 305-701, Korea*

²*Department of Materials, Imperial College London, London SW7 2AZ, UK*

³*Korea Research Institute of Standards and Science, Daejeon 34113, Korea*

Since graphene was discovered in 2004, a variety of 2D materials have been suggested and synthesized. For device applications, these materials require important factors, such as high carrier mobility and sizable band gap. Recently, a few layers of black phosphorus have attracted much attention due to its high carrier mobility and moderate band gap promising for applications to nanoelectronics and nanophotonics. Besides black phosphorus, various phosphorus allotropes have been predicted theoretically, with electronic properties different from those of black phosphorus. In recent experiment, blue phosphorus, one of the theoretically predicted P allotropes, has been successfully synthesized on a gold substrate. The existence of various P allotropes motivates us to search for new P allotropes that may have superior properties for device applications.

In this work, we report the prediction of a novel P allotrope, termed green phosphorus [1], which has been discovered using an *ab initio* evolutionary crystal structure search method [2]. Green phosphorus has a layered structure with interlayer interactions comparable to that of black phosphorus, indicating that it is possible to be exfoliated to a few layers. G_0W_0 -BSE calculations show that green phosphorus has tunable band gaps ranging from 0.7 to 2.4 eV. The nature of direct band gap is maintained even if the film thickness decreases from bulk to monolayer. We find that a monolayer of green phosphorus, called green phosphorene, exhibits anisotropic optical absorption spectra as well as high electron mobility, suggesting that green phosphorene is suitable for various device applications. Finally, we discuss a possible route to synthesizing green phosphorene on corrugated metal surfaces.

[1] W. H. Han, S. Kim, I.-H. Lee, and K. J. Chang, *J. Phys. Chem. Lett.* 8, 4627 (2017).

[2] I.-H. Lee, Y. J. Oh, S. Kim, J. Lee, and K. J. Chang, *Comp. Phys. Commun.* 203, 110 (2016).

Theoretical Study on the Origin of Giant Rashba-type Spin Splitting in Bi/Ag(111) Alloy System

Jisook Hong¹, Jun-Won Rhim², Inkyung Song³, Changyoung Kim^{3,4}, Seung Ryong Park^{5,*}, Ji Hoon Shim^{1,6,*}

¹Department of Chemistry, Pohang University of Science and Technology, Pohang 37673, Korea

²Max-Planck-Institut für Physik komplexer Systeme, 01187 Dresden, Germany

³Center for Correlated Electron Systems, Institute for Basic Science, Seoul 08826, Korea

⁴Department of Physics and Astronomy, Seoul National University, Seoul 08826, Korea

⁵Department of Physics, Incheon National University, Incheon 22012, Korea

⁶Department of Physics and Division of Advanced Nuclear Engineering, Pohang University of Science and Technology, Pohang 37673, Korea

Rashba-type spin splitting (RSS) has recently drawn added attention due to its central role in the field of spintronics. In that regards, designing materials with giant RSS is highly desirable for practical spintronic applications, and thus disclosing the origin of the giant RSS could pave the way for it. Here, we theoretically demonstrate that the giant RSS emerges from the spin-dependent interatomic-hopping, not from a uniform electric field. Our density functional theory calculation and tight-binding analysis show that the interatomic-hopping strength between the surface and subsurface is spin-dependent as a result of the spin-dependent asymmetric charge distribution around the surface. This new interpretation on the RSS successfully explains the giant RSS in Bi/Ag(111) surface states and has implication for the RSS mechanism in general.

Electrochemical CO₂ reduction reaction at sulfur vacancy of MoS₂: Role of hyper-charged states

Sungwoo Kang¹, Youngho Kang^{2*} and Seungwu Han^{1*}

¹*Department of Materials Science and Engineering and Research Institute of Advanced Materials, Seoul National University, Seoul 08826, Korea*

²*Materials Modeling and Characterization Department, Korea Institute of Materials Science, Changwon 51508, Korea.*

MoS₂ is receiving much attention as a promising electrochemical CO₂ reduction reaction (CO₂RR) catalyst. However, the performance for CO₂RR of MoS₂ is limited without ionic liquid or co-catalyst [1-3] because hydrogen evolution reaction (HER) is superior to CO₂RR at edge site [4], which is considered as a main active site. Through first-principles calculation, we investigate the catalytic performance for CO₂ reduction reaction at sulfur vacancy (V_S) of MoS₂, which was recently discovered as a significant active site for HER [5]. We search possible reaction paths by modifying computational hydrogen electrode (CHE) model as considering hyper-charged states [6] of intermediates. H₂CO, CH₄ and CH₃OH are identified as possible products of which theoretical overpotentials (η_{theory}) are 0.56 V_{RHE}, 0.56 V_{RHE} and 0.64 V_{RHE}, respectively, which is comparable to Cu (211) plane (0.68 V_{RHE} in the same computational setting). η_{theory} of those products are much higher when hyper-charged states are not considered (0.82 V_{RHE} and 1.10 V_{RHE}, respectively), which demonstrates that hyper-charged states are possible to reduce thermodynamic step of the electrochemical reaction. The kinetic barriers of CO₂RR and HER at the first protonation step are negligible at applied potential of -0.64 V_{RHE}, respectively, which indicates that CO₂RR is expected to take place comparably to HER even without ionic liquid. Finally, we suggest new C2 mechanism for V_S-MoS₂ at η_{theory} of 0.56 V_{RHE} which shows comparable selectivity with C1 products.

[1] M. Asadi, B. Kumar, A. Behranginia B. A. Rosen, A. Baskin, N. Repnin, D. Pisasale, P. Phillips, W. Zhu, R. Haasch, R. F. Kile, *et al.*, *Nat. Commun.*, 2014, **5**, 4470.

[2] M. Asadi, K. Kim, C. Liu, A. V. Addepalli, P. Abbasi, P. Yasaei, P. Phillips, A. Behranginia, J. M. Cerrato, R. Haasch, *et al.*, *Science*, 2016, **242**, 467-470.

[3] W. Tu, Y. Li, L. Kuai, Y. Zhou, Q. Xu, H. Li, X. Wang, M. Xiao and Z. Zou, *Nanoscale*, 2017, **9**, 9065-9070.

[4] K. Chan, C. Tsai, H. A. Hansen and J. K. Nørskov, *ChemCatChem*, 2014, **6**, 1899-1905.

[5] H. Li, C. Tsai, A. L. Koh, L. Cai, A. W. Contryman, A. H. Fragapane, J. Zhao, H. S. Han, H. C. Manoharan, F. Abild-Pedersen, J. K. Nørskov and X. Zheng, *Nat. Mater.*, 2016, **15**, 48-53.

[6] K. Y. Kim, J. Lee, S. Kang, Y. -W. Son, H. W. Jang, Y. Kang and S. Han, *ACS Catal.*, 2018, **8**, 4508-4515.

Memo

Memo

Memo

Memo

Memo

Memo

