

Nanomaterials

Room Hau - Session NM-TuM

Nanofabrication and Nanodevices I

Moderator: Adam Schwartzberg, Lawrence Berkeley Lab, USA

8:00am **NM-TuM1 Fabrication of Nanoplasmonic Pore on the Pyramid for Single Molecule Analysis**, *SeongSoo Choi, M.J. Park, C.H. Han, S.J. Oh, T. Yamaguchi*, Research Center for Nanobio Science, SunMoon University, Republic of Korea; *Y.S. Kim*, Sungkyunkwan University, Korea, Republic of Korea; *N.K. Park*, School of Electrical Engineering, Seoul National University, Republic of Korea

There have been tremendous interests about the portable single molecule sensor device by using nanopore detection techniques. Recently the portable genome sequencing device based on the electrical detection technique was fabricated, however, the very high error rate during detection process was reported. In this report, we will report the nanoplasmonic Au nanopore fabrication by using diffusion of atoms, or drilling a freestanding thin film under high electron beam irradiation. For diffusion techniques, the Au apertures with ~ 100 nm diameter were fabricated by using focused ion beam technique followed by electron beam irradiations. The Au nanoclusters were formed on the diffused membrane several months later under the room environment after electron beam irradiations. In addition, drilling a freestanding 40 nm thick Au membrane was successfully carried out. Due to unstable electron beam irradiation on the nanoscale spot, we observed a crescent shape aperture with 0.75 nm width on the freestanding Au film. The fabricated nanopore surrounded with periodically patterned grooves can enhance the optical transmission and enhance the optical intensity due to plasmonic hot spot at the pore area. The controlled nanopore can be utilized as single molecule translocation and analysis by using optical detection and optically driven technique.

8:20am **NM-TuM2 Reversible Diels-Alder attachment of Organic Molecules to Single Atoms on Hydrogenated Semiconductors**, *Szymon Godlewski, M. Kolmer*, Jagiellonian University, Poland; *M. Englund*, Donostia International Physics Center, Spain; *H. Kawai*, Institute of Materials Research and Engineering; *R. Zuzak*, Jagiellonian University, Poland; *A. Garcia-Lekue*, Donostia International Physics Center, Spain; *A.M. Echavaren*, Institute of Chemical Research of Catalonia; *D. Pena, D. Perez, E. Guitian*, Universidad de Santiago de Compostela; *C. Joachim*, CEMES-CNRS, France; *D. Sanchez-Portal*, Donostia International Physics Center, Spain; *M. Saeys*, Ghent University, Belgium; *M. Szymonski*, Jagiellonian University, Poland

In recent years the on-surface, local chemistry attracts growing attention, inspired by the interest in fundamental processes occurring on surfaces, as well as by potential technological applications. In particular, the controlled and reversible bonding between single molecules and atoms could be advantageous in construction of prototypical molecular switches, rotors and electronic circuits. To control the electronic properties of organic species the isolation from the underlying substrate is required. This could be achieved by application of passivated surfaces that enable retention of originally designed properties [1-3]. Hydrogen passivated silicon and germanium surfaces are of particular interest enabling creation of atomic scale defects – hydrogen vacancies that could interact with organic molecules and form artificial wiring [4-5].

In the presentation we will demonstrate reversible Diels-Alder attachment of prototypical three-branch conjugated molecules to paired surface dangling bonds. We will show that molecules initially undergo the [4+2] cycloaddition forming chemical bonds with the unsaturated surface dangling bonds [6]. Further, it will be discussed that the bonds could be broken and restored using the low temperature scanning tunnelling microscope. We will demonstrate that at low temperature the molecules located on surface dangling bonds could be switched between the chemisorbed configuration and the physisorbed one held by van der Waals interactions only. Interestingly, the molecules adsorbed in the latter configuration could be controllably switched rotationally with the STM tip, which constitutes the first molecular switch on a passivated surface. Further it will be shown that by tuning the structure of the molecules the attachment geometry could be controlled with high selectivity. Finally we will introduce prospects for utilization of the control over connecting individual molecules with surface single atoms.

[1] Szymon Godlewski et al. ACS Nano 7 (2013) 10105–10111

[2] Amandine Bellec et al. Nano Lett. 9 (2009) 144–147.

[3] Szymon Godlewski et al. Phys. Chem. Chem. Phys. 18 (2016) 3854

[4] Marek Kolmer et al. Phys. Rev. B 86, 125307 (2012)

[5] Marek Kolmer et al. Nanoscale 7 (2015) 12325

[6] Szymon Godlewski et al. Phys. Chem. Chem. Phys. 2016, 18, 16757-16765

This research was supported by the National Science Centre, Poland (contract no. UMO-2014/15/D/ST3/02975) and the 7th Framework Programme of the European Union Collaborative Project PAMS (contract no.610446).

8:40am **NM-TuM3 Full Color Light Emitting Diodes using Perovskite Quantum Dots as Emissive Materials**, *Y.-H. Suh*, Ulsan National Institute of Science and Technology (UNIST), Republic of Korea; *J.-W. Chio*, Gwangju Institute of Science and Technology (GIST), Republic of Korea; *T. Kim*, Ulsan National Institute of Science and Technology (UNIST), Republic of Korea; *S.-H. Han*, Mokpo National Maritime University, Republic of Korea; *J. Park*, Ulsan National Institute of Science and Technology (UNIST), Republic of Korea; *Chang-Lyoul Lee*, Gwangju Institute of Science and Technology (GIST), Republic of Korea

The cesium lead halide (CsPbX₃, X = Cl, Br, and I) based perovskite quantum dots (QDs) were synthesized using precipitation and hot-injection methods. And also, the device performance of QD-LEDs using inorganic perovskite (CsPbX₃) as emissive layer was characterized. The CsPbX₃ showed excellent properties, such as high quantum yield (>80%), easily tunable band gap, and high color purity as well as high stability. The PL spectra of CsPbX₃ were varied by changing the composition of halides. The inverted structure QD-LEDs consisted of ITO/ZnO/poly-electrolyte/PeQDs/MoO₃/Ag showed the high color purity EL (Blue : ~ 480 nm, Green : ~ 510 nm, Red : ~ 680 nm) and high brightness (Blue : ~ 200 cd/m², Green: $\sim 1,200$ cd/m², Red : ~ 300 cd/m²).

9:00am **NM-TuM4 Nanoarchitecture Design for Independent Control of Carrier and Phonon Transports**, *Yoshiaki Nakamura*, Osaka University, Japan

INVITED

Thermoelectric (TE) conversion has been expected to be one of the ideal energy sources, where waste heat is converted to be usable electrical energy. Therefore, TE performance enhancement had drawn much attention. TE performance is related to dimensionless figure of merit, $ZT = S^2 \sigma T / \kappa$, where S is Seebeck coefficient, σ is electrical conductivity, κ is thermal conductivity, and T is absolute temperature. This means that the materials with high S , σ , and low κ are required. However, high σ and low κ are difficult to achieve simultaneously because they are correlated. Therefore, the identical control of σ and κ has been a vital goal in thermoelectric study for long.

We proposed the nanoarchitecture where phonon was scattered effectively at the nanostructure interfaces, but carriers could go through them using ultrasmall epitaxial nanodots (ND) with well-controlled interfaces. Herein, carriers feel that nanomaterials are single crystal because crystals of NDs are oriented. On the other hand, the interfaces of ultrasmall NDs scatter phonon. In this study, we use our original technique for forming epitaxial ND structures. We achieve the identical control of phonon and carrier transports using the above nanoarchitecture design.

Clean Si surfaces were oxidized at 500°C at the O₂ pressure of 2×10^{-4} Pa to form ultrathin (~ 0.3 nm) SiO₂ films. Si or Ge was then deposited to form epitaxial Si or Ge NDs grown on Si substrates with ultrahigh density of $\sim 10^{12}$ cm⁻² as the following. At first stage of Si or Ge deposition, nanowindows were created in the ultrathin SiO₂ films through the reaction of Si+SiO₂→2SiO↑, or Ge+ SiO₂→GeO↑+SiO↑. By subsequent deposition, Si or Ge NDs formed on the ultrahigh density nanowindows. In the case of Ge NDs, Si layers were formed on Ge NDs at 400°C. The above formation of Si NDs or Si layer/Ge NDs and the oxidization process were repeated to fabricate the ND stacked structures.

The formation of the epitaxial nanoarchitectures including NDs was succeeded. The κ values were measured by 2ω method. The smallest κ value of stacked Si NDs with 3 nm diameter was close to the amorphous value. In the case of Ge NDs, the κ values were also drastically reduced compared with bulk Si. The σ of Ge ND structures was enhanced to be a similar value to the bulk Si cases by the interface designing, indicating the independent control of κ and σ . This demonstrates the possibility of our nanostructure as a Si-based thermoelectric material with high ZT .

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Tuesday Morning, December 13, 2016

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9:40am **NM-TuM6 DNA self-assembled Nanostructures using a Scaffold with a Minimum Size**, *Enrique Samano*, Universidad Nacional Autónoma de México, México; *T.H. LaBean*, North Carolina Central University, USA

Artificial DNA nanostructures such as DNA origami have garnered significant interest as templates for sub-20 nm assembly of materials because their design allows for the incorporation of binding sites to assemble nanocomponents with 6nm resolution. The DNA origami strategy for assembling designed supramolecular complexes requires ssDNA as a scaffold strand. A system is described that was designed using approximately one third of the M13 bacteriophage genome as a scaffold. Folding of the short 2404-base ssDNA scaffold into a variety of two-dimensional origami shapes (rectangle, triangle, and circle) with high assembly yields is demonstrated. DNA origami templates, modified to have DNA binding sites with a uniquely coded sequence, can be used to produce complex metallic nanostructures of programmable design. Gold nanoparticles functionalized with a complementary DNA sequence were attached to specific binding sites on the scaffold in a highly controllable manner to form a pre-designed metallic array on two nanostructures.

10:20am **NM-TuM8 Feedback-Controlled Electromigration (FCE) Method with Automatically Optimized Parameters**, *Noriaki Numakura*, *Y. Iwata*, *J. Shirakashi*, Tokyo University of Agriculture & Technology, Japan

Feedback-controlled electromigration (FCE) has been used as a useful technique for fabricating quantum point contacts (QPCs) and nanogaps [1]. The conventional FCE method is performed on a general-purpose operating system (GPOS). In contrast, real-time operating system (RTOS) is specifically designed to run applications with precise timing and reliability. Hence, we have previously reported the FCE method using RTOS, in order to run FCE algorithms time-deterministically and to control quantized conductance of Au nanowires, at room temperature in ambient air [2]. In this report, we propose a new FCE system with automatically optimized parameters for the improvement of control performance of FCE using RTOS.

In this system, we designed the FCE algorithms with randomly varying parameters. First, FCE experiments for the Au nanowires were repeatedly performed to obtain various FCE experimental data. Then, the FCE parameters were evaluated using cost functions, and stored in a database. Finally, we performed the FCE experiments with a suitable set of evaluated FCE parameters, which was generated using the stored data. As a result, the conductance of Au nanowire was successfully quantized and decreased without catastrophic break. Therefore, it is considered that the FCE system based on automatically optimized parameters can show the conductance quantization of Au nanowires even at room temperature in ambient air.

References

- [1] D. R. Strachan, D. E. Smith, D. E. Johnston, T.-H. Park, M. J. Therien, D. A. Bonnell, and A. T. Johnson, *Appl. Phys. Lett.* 86, 043109 (2005).
- [2] S. Sato, Y. Kanamaru, Y. Katogi and J. Shirakashi, 43rd International Conference on the Physics and Chemistry of Surfaces and Interfaces (PCSI-43), Mo0950, January 17-21, 2016, Palm Springs, CA, USA.

10:40am **NM-TuM9 Controlled Switching and Charge State Alteration of Surface Dangling Bond Nanostructures, Realization of a Quantum Hamiltonian Boolean Logic Gate**, *S. Godlewski*, *Rafal Zuzak*, Jagiellonian University, Poland; *M. Kolmer*, Jagiellonian University, Poland; *M. Engelund*, Donostia International Physics Center, Spain; *H. Kawai*, Institute of Materials Research and Engineering, Singapore; *G. Dridi*, CEMES-CNRS, France; *A. Garcia-Lekue*, Donostia International Physics Center, Spain; *C. Joachim*, CEMES-CNRS, France; *D. Sanchez-Portal*, Donostia International Physics Center, Spain; *M. Szymanski*, Jagiellonian University, Poland

In recent years we observe an increasing interest in the precise atomic-scale fabrication of surface nanostructures. This is because the miniaturization of various electronic devices has pushed many fundamental research efforts toward creating atomic-scale circuits [1]. Among different substrates, the hydrogen passivated silicon and germanium surfaces are regarded as the promising ones due to the fact that they allow precise fabrication of complex dangling bond (DB) nanostructures, which introduce new electronic states within the intrinsic band gap of the substrate [2]. These structures could be further utilized in fabrication of atomic scale devices [3] or may be directly applied in quantum circuitry.

In the presentation we will describe fabrication of various surface DB nanostructures. Their geometrical and electronic properties will be analysed on the basis of scanning tunnelling microscopy/spectroscopy (STM/STS) measurements corroborated by density functional theory (DFT)

Tuesday Morning, December 13, 2016

calculations. We will show that upon electron/hole injection dangling bonds exhibit dynamical behaviour, which due to the high frequency of oscillations leads to smooth STM appearance. A new imaging model, based on a dynamical two-state rate equation, which allows detailed reproduction of observed STM pattern will be introduced [4]. Moreover, the influence of single dangling bond charging on the STS spectra will be analysed [5]. We will further envision that our new model might be applied to simulate other bi-stable systems where fluctuations arise from transiently charged electronic states. Finally, we will present successful design and construction of the first prototypical QHC (Quantum Hamiltonian Computing) atomic scale Boolean logic gate operated by the STM tip [6].

- [1] M. Fuechsle et al. *Nat. Nanotechnol.*, 7, 2012, 242–246
- [2] M. Kolmer et al. *Phys. Rev. B* 86, 2012, 125307
- [3] B. Weber et al. *Ohm's Law Survives to the Atomic Scale*. *Science* 335, 2012, 64–67
- [4] M. Engelund et al. *Phys. Chem. Chem. Phys.*, 2016, 18, 19309
- [5] M. Engelund et al. *Scientific Reports* 2015, 5, 14496
- [6] M. Kolmer et al. *Nanoscale* 2015, 7 12325

This research was supported by the National Science Centre, Poland (contract no. UMO-2014/15/D/ST3/02975) and the 7th Framework Programme of the European Union Collaborative Project PAMS (contract no.610446).

11:00am **NM-TuM10 Synthesis of Transition Metal Dichalcogenide Heterostructures and Nanodevices by Atomic Layer Deposition**, *Christoph Kastl*, *C.T. Chen*, Lawrence Berkeley National Laboratory, USA; *B. Shevitski*, University of California, Berkeley, USA; *T.R. Kuykendall*, *S. Aloni*, *A.M. Schwartzberg*, Lawrence Berkeley National Laboratory, USA

Interest in layered transition metal dichalcogenides (TMDs) has been renewed since the discovery of emergent properties when reduced to single two-dimensional layers. The current state-of-the-art fabrication of heterostructures involves exfoliation from bulk crystals and subsequent manual stacking of the atomic layers. The lack of reproducible and large scale synthetic methods for high quality, consistent TMD samples has become a major bottleneck to research on and application of these materials.

The following two-step process involving atomic layer deposition (ALD) and chalcogenization can be used as a scalable and highly versatile method for TMD synthesis. First, high quality transition metal oxide films are deposited by ALD. Second, the transition metal oxide films are converted into layered TMDs by chemical chalcogenization, i.e. annealing under H₂S atmosphere in a high temperature furnace. [1] We leverage the particular advantages of ALD to further develop this approach into a fabrication process for encased nanodevices and heterostructures. Planar films of MoO₃ and/or WO₃ are deposited on the growth substrate, and SiO₂ is used as an inert capping layer. Then, device structures are defined by photolithography and reactive ion etching. In the subsequent conversion step, the nucleation starts at the exposed cross sections of the heterostructure, and the TMD growth proceeds laterally from the nucleation site. Cross-sectional TEM reveals that this lateral growth mode forms highly crystalline MoS₂ (WS₂) layers which are aligned parallel to the substrate. We characterize the optical and electronic properties of the encased heterostructures by photoluminescence spectroscopy, ultrafast transient absorption spectroscopy, and field effect measurements. Furthermore, controlling the extent of the lateral growth via annealing time and temperature, we show that encased TMD nanoribbons can be fabricated with lateral dimension down to ~10 nm.

- [1] C. Kastl et al., The Important Role of Water in Growth of Monolayer Transition Metal Dichalcogenides, *under review* (2016).

11:20am **NM-TuM11 Growth and In situ Electronic Transport and Scanning Probe Studies of Topological Materials Bi₂Se₃ and Na₃Bi**, *Michael Fuhrer*, Monash University, Australia

INVITED

Topological materials are fascinating novel electronic phases with important device implications. Topological insulators are insulating in their interiors but conduct on their boundaries; two-dimensional topological insulators can realize dissipationless conduction along their one-dimensional edges. Topological Dirac semimetals lie at the boundary between conventional and topological insulators, and can be pushed toward one or the other by electric or magnetic fields, realizing new types of switches.

Tuesday Morning, December 13, 2016

I will discuss the development of a novel experimental capability combining the growth of topological materials by molecular beam epitaxy (MBE) with *in situ* magnetotransport and low-temperature scanning tunneling microscopy at 5 K. This capability has been used to study the transport properties of topological insulator Bi_2Se_3 *in situ* during MBE growth, monitoring the carrier concentration and mobility as a function of film thickness[1]. More recently, topological Dirac semimetal Na_3Bi thin films have been prepared on $\alpha\text{-Al}_2\text{O}_3(0001)$ substrates with low temperature charge carrier mobilities exceeding $6000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ with n-type carrier densities below $1 \times 10^{18} \text{ cm}^{-3}$, comparable to the best single crystal values[2]. Perpendicular magnetoresistance at low field shows the perfect weak-antilocalization behaviour expected for Dirac fermions in the absence of intervalley scattering. Our ongoing efforts to tune the carrier density using physical and chemical schemes to realize topological devices will also be discussed [3].

References

1. Jack Hellerstedt et al., Appl. Phys. Lett. **105**, 173506 (2014).
2. J. Hellerstedt et al., Nano Lett. **16**, 3210 (2016).
3. M.T. Edmonds et al., ACS Appl. Mater. Interfaces **8**, 16412 (2016).

Author Index

Bold page numbers indicate presenter

— A —

Aloni, S.: NM-TuM10, 2

— C —

Chen, C.T.: NM-TuM10, 2

Chio, J.-W.: NM-TuM3, 1

Choi, S.: NM-TuM1, 1

— D —

Dridi, G.: NM-TuM9, 2

— E —

Echavaren, A.M.: NM-TuM2, 1

Engelund, M.: NM-TuM2, 1; NM-TuM9, 2

— F —

Fuhrer, M.: NM-TuM11, 2

— G —

Garcia-Lekue, A.: NM-TuM2, 1; NM-TuM9, 2

Godlewski, S.: NM-TuM2, 1; NM-TuM9, 2

Guitian, E.: NM-TuM2, 1

— H —

Han, C.H.: NM-TuM1, 1

Han, S.-H.: NM-TuM3, 1

— I —

Iwata, Y.: NM-TuM8, 2

— J —

Joachim, C.: NM-TuM2, 1; NM-TuM9, 2

— K —

Kastl, C.: NM-TuM10, 2

Kawai, H.: NM-TuM2, 1; NM-TuM9, 2

Kim, T.: NM-TuM3, 1

Kim, Y.S.: NM-TuM1, 1

Kolmer, M.: NM-TuM2, 1; NM-TuM9, 2

Kuykendall, T.R.: NM-TuM10, 2

— L —

LaBean, T.H.: NM-TuM6, 2

Lee, C.-L.: NM-TuM3, 1

— N —

Nakamura, Y.: NM-TuM4, 1

Numakura, N.: NM-TuM8, 2

— O —

Oh, S.J.: NM-TuM1, 1

— P —

Park, J.: NM-TuM3, 1

Park, M.J.: NM-TuM1, 1

Park, N.K.: NM-TuM1, 1

Pena, D.: NM-TuM2, 1

Perez, D.: NM-TuM2, 1

— S —

Saeys, M.: NM-TuM2, 1

Samano, E.C.: NM-TuM6, 2

Sanchez-Portal, D.: NM-TuM2, 1; NM-TuM9, 2

Schwartzberg, A.M.: NM-TuM10, 2

Shevitski, B.: NM-TuM10, 2

Shirakashi, J.: NM-TuM8, 2

Suh, Y.-H.: NM-TuM3, 1

Szymonski, M.: NM-TuM2, 1; NM-TuM9, 2

— Y —

Yamaguchi, T.: NM-TuM1, 1

— Z —

Zuzak, R.: NM-TuM2, 1; NM-TuM9, 2