

Monday Afternoon, October 31, 2011

Energy Frontiers Focus Topic

Room: 103 - Session EN+EM+NS-MoA

Nanostructured Materials for Third Generation Solar Cells

Moderator: W.A. Tisdale, Massachusetts Institute of Technology

2:00pm **EN+EM+NS-MoA1 Fabrication of Two-dimensional Array of Sub-10nm GaAs Nanodisk using Bio-template Neutral Beam Etching Process, S.-H. Lin, X.-Y. Wang, C.-H. Huang, Y. Ohno, M. Igarashi, Tohoku University, Japan, A. Murayama, Hokkaido University, Japan, S. Samukawa, Tohoku University, Japan**

Recently, 3-dimensional (3D) or 2-dimensional (2D) quantum dot superlattice are widely investigated to develop the new generation devices, such as quantum dot solar cell. Quantum dot superlattice consists of quantum dots with lower band gap energy in the matrix with higher band gap energy. For the device application, the structure requires high QD density, periodic QD array and uniform dot size and inter-dot spacing. Molecular beam epitaxy (MBE) and Metal-organic chemical vapor deposition (MOCVD) are two attractive methods among various bottom-up fabrication methods to fabricate quantum dots. However, there is limitation of bottom-up process to control the size, spacing and density of quantum dots. To breakthrough these problems, we have proposed the ultimate top-down process by combination of bio-template and damage-free neutral beam etching (NBE) for fabricating defect-free 2D array of quantum dots.

In this study, we developed a series of novel process for fabricating uniform size and high density 2D array of GaAs nanodisk with uniform inter-dot-spacing. Firstly, the hydrogen-radical treatment was used to remove the native oxide on GaAs surface. To utilize two-dimensional array of ferritin (protein including 7-nm-diameter iron core) as an etching mask, the formation of a hydrophilic 1-nm thick GaAs neutral beam oxidation film is key point. It is found that protein shell can be removed with oxygen-radical treatment at a low temperature of 280°C without thermal damage to GaAs. After protein shell removal, the iron oxide cores inside the ferritins remained on the surface. Then, using the iron core as an etching mask, neutral beam could etch the defect-free nanodisk structure of GaAs. Finally, we developed that iron oxide core was removed by wet etching with diluted hydrogen chloride and completed a fabrication process without inflicting any damage to GaAs. The result shows the quantum dot superlattice structure with a two-dimensional array of GaAs quantum dots with a diameter of ~7-nm, a height of ~10-nm, and a quantum dot density of more than $7 \times 10^{11} \text{cm}^{-2}$ was successfully demonstrated without any damage to GaAs.

This work is supported by the Core Research of Evolutional Science and Technology (CREST) of Japan Science and Technology Agency (JST).

2:40pm **EN+EM+NS-MoA3 Solution Processed Quantum Dots for Low Cost Photovoltaics, E.J.D. Klem, J. Lewis, C. Gregory, G. Cunningham, D. Temple, RTI International** **INVITED**

For solar energy to be a significant component of our energy supply new technologies are needed that enable the fabrication of low cost, high efficiency solar cells. Research into solar energy devices which incorporate carbon fullerenes and semiconducting polymers represent one such technology. One factor limiting their further advance is their lack of absorption in the infrared (IR). As half the sun's energy lies beyond 700 nm and one third beyond 1000 nm, low-cost device technologies are needed which capture this lost infrared potential. An additional factor limiting the further advance of these devices is the relatively poor electrical transport properties of most semiconducting polymers.

The use of solution processed quantum dots provides a potential route towards overcoming both of these limitations. Solution processed quantum dots can be tuned to absorb light well into the infrared, and quantum dot composite thin films have been shown to have charge carrier mobilities approaching that of amorphous silicon.

In this presentation we will present a brief overview of colloidal quantum dots and the field of quantum dot photovoltaics. We will discuss a range of device architectures and material systems that have been explored experimentally. This includes quantum dot-metal Schottky junctions, quantum dot heterojunctions, and quantum dot-metal oxide junctions.

We will also present a device architecture which is based on the heterojunction formed between infrared-sensitive PbS quantum dots and C_{60} fullerenes. In this device pre- and post-deposition treatments are used

passivate carrier traps and increase the conductivity of the quantum dot films. A device stack is presented that is designed to steer photo-excited charge carriers to the charge-separating interface, reducing recombination pathways and improving carrier extraction efficiency. Under simulated solar illumination the devices exhibit short circuit current densities greater than 20 mA/cm², power conversion efficiencies greater than 5%, and spectral sensitivity out to 1500 nm. This represents a significant step towards demonstrating the commercial viability of solution processed quantum dot technology

3:40pm **EN+EM+NS-MoA6 Role of Quantized and Mid-Gap States in "Dark" Charge Transport and Photoconductivity in Semiconductor Nanocrystal Films, P. Nagpal, Los Alamos National Laboratory**

Colloidal semiconductor nanocrystals (NCs) have attracted significant interest for applications in solution-processable devices such as light-emitting diodes and solar cells. However, poor understanding of charge transport in NC assemblies, specifically the relation between electrical conductance in dark and under light illumination, hinders their technological applicability. Here, we simultaneously address the issues of "dark" transport and photoconductivity in films of PbS NCs by incorporating them into optical field-effect transistors (OFETs), in which the channel conductance is controlled by both gate voltage and incident radiation. Spectrally resolved photoresponses of OFETs reveal a weakly conductive mid-gap band (MGB) which is responsible for charge transport in dark. The mechanism for conductance, however, changes under illumination when it becomes dominated by band-edge quantized states. In this case, the MGB still plays an important role as its occupancy (tuned by the gate voltage) controls the dynamics of band-edge charges. Our study has broad implications for NC-based electronics and optoelectronics, and specifically, suggests that design guidelines for NC devices might be different depending on whether they are intended for operation in dark (diodes and transistors) or under illumination (photodetectors and solar cells).

4:00pm **EN+EM+NS-MoA7 Using Surface Chemistry to Modulate the Bandgap of Ge Nanowires, S. Sivaram, M.A. Filler, Georgia Institute of Technology**

Small-diameter semiconductor nanowires are highly attractive building blocks for next generation photovoltaic devices because they exhibit highly tunable optoelectronic properties as a result of quantum confinement. Bottom up approaches, such as the vapor-liquid-solid (VLS) growth mechanism, are controllable down to ~20 nanometers but significant challenges exist at smaller length scales where property tuning is maximized. Furthermore, it is expected that the optoelectronic properties of quantum-confined nanowires will be heavily dependent on surface chemistry, yet there are no experimental studies that fundamentally probe this relationship. Germanium is an ideal system to study because of its large Bohr exciton radius, low rate of oxidation, and chemical similarity to silicon. In this work we controllably synthesize germanium nanowires with diameters from 5 to 20 nanometers using gold catalyst particles with a narrow size distribution. To obtain epitaxial Ge nanowires with uniform diameters and lengths, a two-step growth process is employed that includes a brief, high-temperature nucleation (390°C) above the Au-Ge eutectic point, followed by elongation at various process conditions, generally below the eutectic point (280 – 340°C, 1×10^{-7} – 1×10^{-4} Torr). To limit catalyst diffusion and formation of the $\sqrt{3} \times \sqrt{3}$ Au/Si reconstruction on the Si(111) surface, a key problem for controllable sub-20 nm growth, the substrate is passivated with -CH₃ groups prior to nanowire synthesis by introducing small quantities of methylgermane. Nanowire surface chemistry is probed with *in-situ* transmission infrared (IR) spectroscopy both during and after growth. We introduced different adsorbates (-H and -CH₃) and monitored their influence on the band gap as a function of surface coverage. We show that the band gap of Ge nanowires blue shifts with H passivation and red shifts with CH₃ termination.

4:20pm **EN+EM+NS-MoA8 Production of Multi-milligram Yields of Ternary II-VI Semiconductor Nanocrystals Under Non-coordinating Amine Activated Synthesis, M. Plaisant, P.H. Holloway, University of Florida**

Thermolysis of Zn and Cd precursors under a non-coordinating one-pot wet-chemical synthesis has successfully produced a heterogeneous ternary Cd_xZn_{1-x}Se alloy with an extensive spectral red shift of 200nm in the visible range. Large yields of twenty milligram of the Cd_xZn_{1-x}Se alloy are minimally required for inclusion of the material in the active layer of an inorganic/organic photovoltaic device. The facile synthesis of such large yields of ternary semiconductor nanocrystal alloys has not commonly been reported in the literature. Herein we discuss first the thermolytic synthesis

of the material beginning from the ZnSe core through the inclusion of the Cd-precursor. We then discuss the production of the ternary II-VI semiconductor $Cd_xZn_{1-x}Se$ material through a process of non-coordinating synthesis with amine-activation to produce the high multi-milligram yields required for device inclusion.

4:40pm **EN+EM+NS-MoA9 Type-II ZnTe/ZnSe Quantum Dots for Intermediate Band Solar Energy Conversion**, *C. Chen, B. Juang, J. Hwang, S. Kim, X. Pan, J. Phillips*, University of Michigan

Intermediate band solar cells and impurity photovoltaics have been proposed to achieve high efficiency solar energy conversion by introducing electronic states within the bandgap of the host material. The intermediate electronic states provide enhanced photocurrent through the addition of sub-bandgap optical transitions while maintaining a voltage that follows the host material. Approaches to realize these solar cells include the incorporation of dopants/impurities, dilute alloys, and nanostructures such as quantum dots. Self-assembled quantum dots such as InAs/GaAs possess excellent optical properties and have been applied to numerous optoelectronic devices including demonstration of the intermediate band solar cell concept. The highly radiative transitions in these type-I materials result in short radiative carrier lifetimes – a feature that is desirable for light emitters, but can be problematic for photodetectors and solar cells. Semiconductor nanostructures with type-II band alignment result in spatially separated electron and hole wavefunctions, resulting in reduced oscillator strength and corresponding reduction in optical absorption, spontaneous radiative recombination rate, and increased radiative carrier lifetime. The tradeoff between carrier lifetime and optical absorption may be effectively used to provide a closer match to the generation-recombination rates desired for intermediate band solar energy conversion. In this work, ZnTe/ZnSe type-II quantum dots are proposed for intermediate band solar energy conversion. The theory of intermediate band solar energy conversion in this material system will be presented along with initial experimental results on the epitaxial growth, structural properties, and optoelectronic response of the materials. The ZnTe/ZnSe quantum dots were grown by molecular beam epitaxy, where three-dimensional island formation is observed via strained layer growth in the Stranski-Krastanow growth mode. Low temperature photoluminescence spectra reveal optical transitions from ZnSe (2.8eV), Te_{Se} isoelectronic centers (2.6eV), and broad emission in the range of 2.0-2.4eV attributed to type-II quantum dots.

Authors Index

Bold page numbers indicate the presenter

— C —

Chen, C.: EN+EM+NS-MoA9, **2**
Cunningham, G.: EN+EM+NS-MoA3, **1**

— F —

Filler, M.A.: EN+EM+NS-MoA7, **1**

— G —

Gregory, C.: EN+EM+NS-MoA3, **1**

— H —

Holloway, P.H.: EN+EM+NS-MoA8, **1**
Huang, C.-H.: EN+EM+NS-MoA1, **1**
Hwang, J.: EN+EM+NS-MoA9, **2**

— I —

Iagarashi, M.: EN+EM+NS-MoA1, **1**

— J —

Juang, B.: EN+EM+NS-MoA9, **2**

— K —

Kim, S.: EN+EM+NS-MoA9, **2**
Klem, E.J.D.: EN+EM+NS-MoA3, **1**

— L —

Lewis, J.: EN+EM+NS-MoA3, **1**
Lin, S.-H.: EN+EM+NS-MoA1, **1**

— M —

Murayama, A.: EN+EM+NS-MoA1, **1**

— N —

Nagpal, P.: EN+EM+NS-MoA6, **1**

— O —

Ohno, Y.: EN+EM+NS-MoA1, **1**

— P —

Pan, X.: EN+EM+NS-MoA9, **2**
Phillips, J.: EN+EM+NS-MoA9, **2**
Plaisant, M.: EN+EM+NS-MoA8, **1**

— S —

Samukawa, S.: EN+EM+NS-MoA1, **1**
Sivaram, S.: EN+EM+NS-MoA7, **1**

— T —

Temple, D.: EN+EM+NS-MoA3, **1**

— W —

Wang, X.-Y.: EN+EM+NS-MoA1, **1**