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"Optically Active 3-Dimensional Semiconductor Quantum Dot Assemblies in Heterogeneous Nanoscale Hosts"

Principal Investigator: Professor Arto Nurmikko, Brown University

Division of Engineering, Providence, RI 02912

Arto_Nurmikko@brown.edu

Synopsis: In this project we focused on two-classes of semiconductor light emitting nanocrystals, II-VI compound quantum dots and inorganic-organic perovskite halides, both of which are synthesized by low-cost, solution based methods. The research has increased basic understanding of the relationship between electronic structure and its dimensionality in relation to optical properties, specifically optical gain for candidate laser devices. Using ultrafast, time-resolved spectroscopic methods and innovative thin film fabrication techniques has enabled us to identify excitonic components for the radiative gain, and formulate approached to reduce the non-radiative Auger recombination processes. By embedding the nanocrystal II-VI and perovskite optical gain media in several different types of optical microcavities, we have produces an assessment as to the promise of these materials for transformation to high performance semiconductor lasers across the blue-green-red spectral divide.

Summary:

Semiconductor light emitters are part of our everyday life. Compact lasers are the basic devices in fiber optic networks and drive optical storage devices such as DVDs, while white light LEDs are transforming the world of lighting. These technologies rely on sophisticated epitaxial single crystal growth techniques based on molecular beam epitaxy (MBE) and metal-organic chemical vapor deposition (MOCVD) that are routinely used in mass production. As an alternative to high-tech MBE and MOCVD, synthesis of wide range of nanocrystal materials has found remarkably simple creative routes via solution chemistry as well as by simplified CVD methods. This work has produced a plethora of high quantum yield emitters in the laboratory, including colloidal II-VI, III-V, and lead salt QDs, perovskite nano- and microcrystals and so on. Enticingly, their light emission (and absorption) capabilities span the wavelength range from blue into infrared. Hence an opportunity beckons: low-cost production of single nanocrystal material where wavelength of light emission can be tuned seamlessly across a huge range.

In this project we use basic material science research while translating solution-based nanocrystal synthesis techniques to optically pumped laser device prototypes. The prototype devices in clude different types of optical microcavities: distributed grating feedback resonators, vertical cavity surface emitting lasers, and photonic crystal lasers, respectively. The internal radiative efficiencies of both II-VI and perovskite nanocrystals have reached up to 90% across the range covering red-green-blue and near IR. Compared with incoherent emitters, a semiconductor laser requires three critical attributes: (a) atomic/electronic structure compatible with formation of optical gain; (b) a low-loss optical resonator for feedback; (c) means for energy efficient external excitation to achieve lowest possible threshold for lasing. Beyond the high temporal and spatial coherence (monochromaticity and collimated laser beams, respectively), the devices must sustain operation without degradation for thousands of hours. This project has produced an assessment whether these requirements can be met via state-of-the art nanocrystal science in the II-VI and perovskite materials, respectively.

<u>Publications and Products Based on DOE/BES Support</u>

"New Luminescent Material Incorporating Colloidal Semiconductor Quantum Dots within a Nanoporous Gallium Nitride Matrix", Cuong Dang, Yu Zhang , Jung Han, Arto Nurmikko, Craig Breen, Jonathan S. Steckel and Seth Coe-Sullivan, Phys. Stat. Sol (c) volume 8, Issue 7-8, July 2011, Pages: 2337–2339; *The research was supported by Department of Energy grant (ER-46387) and the National Science Foundation (ECCS-0725740)*

"Red, Green, and Blue Lasing enabled by Single-Exciton Gain in Colloidal, Densely Packed Quantum Dot Films", Cuong Dang, Joonhee Lee, Arto Nurmikko, Craig Breen, Jonathan S. Steckel and Seth Coe-Sullivan, Nature Nanotechnology 7, 335-339 (2012); *Research was funded by the Department of Energy (BES office), the Air Force Office for Scientific Research and the National Science Foundation.*

"A Wafer-Level Integrated White Light Emitting Diode Incorporating Colloidal Quantum Dot as Nanocomposite Luminescent Medium", Cuong Dang, Joonhee Lee, Yu Zhang, Jung Han, Craig Breen, Jonathan S. Steckel, Seth Coe-Sullivan and Arto Nurmikko, Advanced Materials, 24 AUG 2012, DOI: 10.1002/adma.201202354; *This research is supported by the Department of Energy–BES office (DE-FG02-07ER46387) and the National Science Foundation (ECCS-1128331).*

"Quasi-Continuous Distributed Feedback Lasers from Dense Colloidal Quantum Dot Films", Cuong Dang, Joonhee Lee, Kwangdong Roh, Hanbit Kim, Sungmo Ahn, Heonsu Jeon, Craig Breen, Jonathan S. Steckel, Seth Coe-Sullivan, and Arto Nurmikko (Applied Physics Letters, in press 2013); *Financial support for this research was provided by the National Science Foundation under grant: ECCS-1128331 and Department of Energy (BES office) under grant: DE-FG02-07ER46387.*

"Quasi-Continuous Distributed Feedback Lasers from Dense Colloidal Quantum Dot Films", Cuong Dang, Joonhee Lee, Kwangdong Roh, Hanbit Kim, Sungmo Ahn, Heonsu Jeon, Craig Breen, Jonathan S. Steckel, Seth Coe-Sullivan, and Arto Nurmikko, Applied Physics Letters, 103, 171104 (2013), *The authors acknowledge gratefully the support of grants from DOE/BES (DE-FG02-07ER46387)*

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“Reusable Inorganic Templates for Electrostatic Self-Assembly of Individual Quantum Dots”, Mingming Jiang, Jonathan A. Kurvits, Yao Lu, Arto V. Nurmikko, and Rashid Zia, Nanoletters 2015, 15, 5010−5016; DOI: 10.1021/acs.nanolett.5b01009. *The authors acknowledge gratefully the support of grants from DOE/BES (DE-FG02-07ER46387)*

“What Future for Nanocrystal-based Light Emitters?”, Arto Nurmikko, Nature Nanotechnology 10, 1001–1004 (2015), doi:10.1038/nnano.2015.288. *The authors acknowledge gratefully the support of grants from DOE/BES (DE-FG02-07ER46387)*

“A photonic crystal laser from solution based organo-lead iodide perovskite thin films”, S Chen, K Roh, J Lee, WK Chong, Y Lu, N Mathews, TC Sum, A Nurmikko, ACS nano 10 (4), 3959-3967 (2016). *The authors acknowledge gratefully the support of grants from DOE/BES (DE-FG02-07ER46387)*

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“High-Q, Low-Threshold Monolithic Perovskite Thin-Film Vertical-Cavity Lasers”, S Chen, C Zhang, J Lee, J Han, A Nurmikko, Advanced Materials 1604781(2017) DOI: 10.1002/adma.201604781. *The authors acknowledge gratefully the support of grants from DOE/BES (DE-FG02-07ER46387)*

“Towards Electrically Driven Perovskite Lasers – Prospects and Obstacles”, Arto Nurmikko and Songtao Chen, in *Halide Perovskites: Photovoltaics, Light Emitting Devices and Beyond*, Editors Tze-Chien Sum and Nripan Mathews**,** Wiley-VCH (in press). *The authors acknowledge gratefully the support of grants from DOE/BES (DE-FG02-07ER46387)*

“Coherent Light Emitters from Solution Chemistry: Inorganic II-VI Nanocrystals and Organometallic Perovskites” Songtao Chen and Arto Nurmikko, Journal of Selected Topics in Quantum Electronics (invited), under review; *The authors acknowledge gratefully the support of grants from DOE/BES (DE-FG02-07ER46387)*

“Stable Green Perovskite Vertical-Cavity Surface-Emitting Lasers on Rigid and Flexible Substrates", Songtao Chen and Arto Nurmikko (submitted to Nanoletters); *The authors acknowledge gratefully the support of grants from DOE/BES (DE-FG02-07ER46387)*

The work has been also presented in several conferences; approximately 20 presentations including many invited talks