SAND2013-8312P

PEEM studies of surface defects and compositional inhomogeneities in InGaN-based heterostructures

1. Describe the main scientific or technology-related question you are trying to address (5000 character max):

InGaN LEDs are the enabling technology of a global effort to replace conventional lighting with energy-efficient Solid-State Lighting (SSL). However, the remarkable properties of InGaN alloys remain poorly understood, with major fundamental questions still unanswered. Foremost among these is: Why do blue-emitting InGaN alloys have high radiative efficiency despite having threading-dislocation densities that quench light emission in traditional semiconductors? This is generally attributed to localization of the carriers that drives them away from crystalline defects. However, the nature of carrier localization and its microscopic origin are highly controversial. Also, the potential variation (landscape) around dislocations, which determines how non-radiative recombination of the charge carriers happens, is yet to be understood. These nano-scale material properties (carrier localizing structures/dislocations) may also contribute to long-standing (~ 15 year) roadblocks to high-efficiency SSL, including the "green-yellow gap" in LED efficiency (where the efficiency of InGaN degrades at longer wavelengths) and the "efficiency droop" observed for InGaN LEDs operated at high current. Understanding the nano-scale material properties in InGaN LED structures is therefore of major importance.

2. Explain how this project relates to recent work in the field and its expected significance and impact (5000 character max):

Multiple hypotheses have been proposed to explain the high quantum efficiency of InGaN-based LED containing high threading-dislocation densities, many of which invoke the carrier-localizing structures that drive carriers away from non-radiative recombination centers [Humphreys, Phil. Mag. 87, 1971 (2007)]. The proposed carrier-localizing structures include, but are not limited to, alloy composition fluctuation [Watson-Parris, et al., PRB 83, 115321 (2011)], indium-rich dots [Gerthsen, et al., Phys. Status Solidi A 177, 145 (2000)], and quantum-well (QW) thickness variation. Nonetheless, the scientific community has not reached consensus on which of these proposed structures may actually contribute to the carrier localization. As for the dislocations that contribute to the non-radiative recombination, v-defect (which nucleates from a threading-dislocation) is suggested as a main source [Hangleiter et al., PRL 95, 127402 (2005)], though more recent work proposes that only the clusters of v-defect (trench defects) are responsible for non-radiative recombination [Massabuau, et al., J. Appl. Phys., 113, 073505 (2013)].

One of the difficulties in solving this problem is the order of magnitude difference in the spatial length scale of the proposed structures and defects spanning from atomic-scale alloy composition variation to the sub-micron scale v-defect and trench defect (a few 100 nano-meters). Thus, this problem calls for an experimental tool to reveal the microscopic surface structure, the chemical composition variation, and the surface electronic properties concurrently on InGaN heterostructures.

The impact of addressing this scientific question could be immense. As mentioned in the previous section, solving the issue of "efficiency droop" and "green-yellow gap" could once again revolutionize the lighting technology leading to a few percent reduction in our electricity consumption.

3. Outline the technical approach you are taking to answer the questions stated in question 1 (5000 character max):

Our approach is to employ low-energy electron microscopy (LEEM) and photo-emission electron microscopy (PEEM) to reveal the carrier localization mechanisms and the non-radiative recombination impacting efficiencies of InGaN LED structures. The samples are prepared using the state-of-the-art growth capability for nitride semiconductor at Sandia Labs. Using LEEM-PEEM with micro- to nano-meter spatial resolution, we will spatially probe surface potential, density-of-states (DOS), and alloy composition based on the spectroscopic operation of the instrument.

4. List the aspects of the project will be done at your home institution or elsewhere (5000 character max):

At Sandia Labs, we conduct LEEM-PEEM study of InGaN single and multi-quantum well (QW) structures. We have been working on optimizing the sample preparation methods that are suitable for LEEM-PEEM experiment. So far, we have verified that we can conduct LEEM and PEEM using helium-lamp experiments to examine the v-defects and trench defects, and to probe the surface potential variation on freshly prepared samples with brief air exposure. We note that reasonably good LEED patterns are obtained using this experimental procedure.

In addition to LEEM-PEEM studies, the InGaN samples are characterized using high-resolution x-ray diffraction, scanning electron microscopy, atomic force microscopy, and x-ray photoelectron spectroscopy using magnesium/aluminum dual source at Sandia labs.

5. Describe very clearly and specifically the research tasks you expect to perform at the CFN and the expected outcomes (5000 character max):

At CFN, we plan to conduct photoemission spectroscopy using PEEM. The first focus will be to study the alloy composition variation in the imaging mode using In-4d and the Ga-3d states lying at the low binding energies of ~17 eV and ~19 eV, respectively. Absolute compositional variations will then be estimated from the relative variations of these two core-levels by referencing to the known average composition and thickness of the QW structure, which is separately determined at Sandia Labs. A particular emphasis will be given to the vicinity of v-defects and trench defects (a few 100 nano-meters in lateral size) with the spatial resolution aimed at 50 nanometer or less. This spatial resolution is attainable using a field of view (FOV) of 10 micron or less. We plan to conduct the core-level spectroscopy work using the variable photon energy of 50-100 eV.

The second focus will be the study of the valence band (VB) DOS and the dispersion. These measurements will be conducted using photon of the energy of 20-100 eV. Again, we will use the imaging mode with the 10 micron FOV or less for the VB-DOS mapping. Particular attention should be given to the band alignment at the vicinity of v-defects and trench defects. For VB dispersion measurements, we will use an area-selection aperture and operate PEEM in the diffraction mode. It is known that for an $In_xGa_{1-x}N$ with the high indium content ($x > \sim 0.3$), a significant downward band bending occurs forming a two-dimensional surface electron gas [Veal, et al., Phys. Stat. Sol. (a) 203, 85 (2006)] similar to the case of InN [Colakerol, et al., PRL, 97, 237601 (2006)]. An analogous effect can be expected on InGaN structures if indium composition is raised locally.

6. Thinking short term, describe what you expect to accomplish during the next 4-month cycle (5000 character max)

We request 4 days of the PEEM beamtime using synchrotron radiation during the first 4 month (Jan-April 2014). During this period, we will first focus on accessing the photon flux with the photon spectral width needed for core-level and VB PEEM spectroscopic studies. An appropriate energy resolution of the electron filter, FOV, and acquisition time will be determined for our InGaN QW structures. We will also vary the photon energy to look for the condition for a high photoemission cross section as well as accessing the possibility of a depth profiling by varying the photon energy. We expect to obtain enough data to evaluate a proof-of-principle for our proposed work during this period.

7. Provide a short description of how you will utilize each facility and instrument you have selected (5000 character max)

We will conduct photoemission spectroscopy to study shallow core-levels (In-4d and the Ga-3d states at binding energies of ~17 eV and ~19 eV, respectively) and VB using PEEM. LEEM mode operation will be used to verify that the sample condition measured at synchrotron PEEM is qualitatively the same as LEEM-PEEM measurement at Sandia labs.

8. Describe your team's experience and expertise relevant to this proposal. Include up to three publications that will assist the Proposal Review Panel in evaluating your work (5000 character max):

Our team proposing this work has expertise in LEEM-PEEM as well as photoemission spectroscopy. Ohta (PI) is known for his angle-resolved photoemission spectroscopy (ARPES) work on two-dimensional crystal (graphene, molybdenum disulfide, etc.) [Ohta, et al., Science, 313, 951 (2006)] and lately using the combination of LEEM-PEEM [Ohta, et al., PRB 85, 075415 (2012)] and ARPES [Ohta, et al., PRL 109, 186807 (2012)]. He is also an experienced user at various synchrotron facilities (Advanced Light Source, BESSY II). Kellogg is a world renowned expert in LEEM-PEEM research with >16 years of experience. He is well known for his work on self-assembly and pattern formation of metal and semiconductor films [Plass, et al., Nature 412, 875 (2001)]. He also serves as a member of international advisory board of biannual LEEM/PEEM Workshop. Diaconescu has substantial experience in LEEM-PEEM research [Robinson, et al., ACS Nano 7, 637 (2013)]. InGaN samples are prepared and characterized by our collaborators, Mary Crawford, Stephen Lee, and Daniel Koleske, at Sandia National Laboratories. They are all well-known experts the research field of nitride-based solid state lighting.