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13. ABSTRACT (*Maximum 200 words*).

The overall goal of this program has to develop novel light emitting optoelectronics and photonic devices. We have investigated MBE-grown GaN thin films doped with rare earth (RE) species for emission of light over a wide wavelength range from the near-ultraviolet to the near-infrared.

We have shown that Er-doped GaN is an excellent emitter both in the infrared at 1.5 µm and in the visible (green light at 537 and 558 nm). This makes Er-doped GaN a very promising material candidate for light sources to be utilized in optical communication and interconnections because the IR photoluminescence at room temperature is much stronger than that in other semiconductors. Furthermore, the presence of strong and spectrally pure visible emission indicates that applications such as bright, robust display devices can be developed within the same materials technology base. We have shown the other primary visible colors can be emitted with high intensity at room temperature in RE-doped GaN: red from Pr and Eu, and blue from Tm. We conclude that bright and robust full-color displays using RE-doped GaN are technically feasible and commercially attractive.

14. SUBJECT TERMS

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I. Introduction

The overall goal of this program has been the development of novel light emitting optoelectronics and photonic devices. We have investigated MBE-grown GaN thin films doped with rare earth (RE) species for emission of light over a wide wavelength range from the near-ultraviolet to the near-infrared. In this final report we review our research with the incorporation of trivalent rare earth (RE^{3+}) elements into GaN films and resulting light emitting devices. GaN is a wide bandgap semiconductor that is intensely investigated[1] for optical and electronic applications. The lanthanide elements, commonly known as the "rare earths" (RE), have been an increasingly important ingredient in a variety of photonic applications[2], ranging from solid-state lasers to color displays to optical fiber telecommunications. RE elements have a partially filled inner ($4f^{n}$) shell shielded from its surroundings by complete filled outer ($5s^2$ and $5p^6$) outer orbitals. This results in optical emission of very sharp lines at wavelengths from the UV to the IR which are relatively independent of the host material and are determined by the energy of the transition between $4f$ states of the RE. While the host material has a weak influence on the emission wavelength, it does have a very strong effect on the radiative transition probability. In general, doping of conventional semiconductors (Si, GaAs, etc.) with REs has suffered from limited solubility and severe temperature quenching, which has made the room temperature operation impractical.

II. Optical Emission Characteristics

At Cincinnati, we have obtained for the first time photoemission from higher excited RE states in GaN covering the entire visible spectrum: light emission in the green[3,4,5] (from Er at 537/558 nm), red[6,7] (Pr at 650 nm, Eu at 621nm), and blue[8] (Tm at 477 nm). Emission in the near-infrared (IR) was also obtained at 801 nm from Tm[8], at 1000 and 1540 nm from Er[5], and at 956, 1303 and 1914 nm from Pr[6]. The rare earths were introduced during growth of the GaN layer by MBE on either sapphire or Si substrates. **Fig. 1** shows emission spectra from electroluminescent devices (ELD) for GaN doped with Tm, Er, and Eu and a photoluminescent (PL) spectrum from Pr-doped GaN. Also shown in **Fig. 1** is the intrinsic GaN characteristic emission in the ultraviolet part of the spectrum at ~365 nm. The primary visible colors emitted by GaN ELDs doped with these individual REs are very "pure" and match very well the CIE coordinates adopted by the National Television System Committee (NTSC). In addition to the pure colors, mixed colors have been obtained by co-doping GaN films with a combination of REs. As discussed in more detail below, GaN:RE ELDs have been developed which emit in a variety of pure and mixed colors. In general, emission from GaN:RE is surprisingly strong, being observable with the naked eye at room temperature. Thermal quenching of the emitted light is frequently not observed until well above room temperature. Thus, rare-earth-doping of GaN represents an interesting alternative to semiconductor alloying (GaN/InN/AlN) for visible light emission applications and has the additional attractive aspect of strong IR emission for telecommunications and other applications. In addition, Er^{3+} doping of GaN has been shown to produce strong near-IR 1.5 µm emission suitable for fiber optic telecommunications from the lowest excited state.

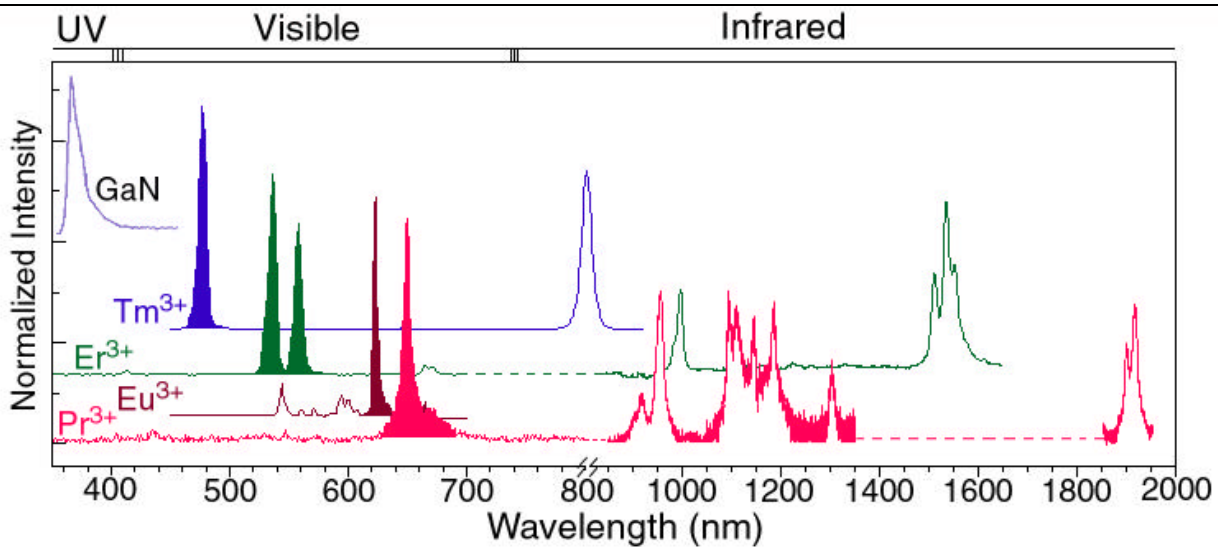

Fig. 1 The emission spectrum of several different RE-doped GaN films from the visible out to the IR wavelengths. All spectra are normalized to their own highest value and are not readily comparible to each other in intensity. The color of the GaN:Tm emission is blue, GaN:Er is in the green, and both GaN:Eu and GaN:Pr emit in the red. For the visible region, all spectra were taken from working ELDs. In the IR region, the GaN:Tm and the GaN:Er spectra are from working EL devices while the GaN:Pr is from PL data.

III. Rare Earth Incorporation

Theoretically, RE intra-$4f^n$ atomic transitions are parity forbidden by the Laporte selection rule. RE ions incorporated into a partially ionic solid frequently substitutionally occupy the cation site. In the cation site an uneven ligand crystal field relaxes the selection rule and increases the probability of intra-$4f^n$ transitions[9]. These *4f*-*4f* transitions, however, are still not fully allowed, resulting in excited state lifetimes of ~10^{-3} s for RE-doped systems. In wurtzitic GaN, which has a significant component of ionic bonding, the RE^{3+} ions have strong optical activity levels, since they are generally substitutionally located on the Ga sub-lattice where the lack of inversion symmetry produces strong ligand fields thereby increasing the *4f*-*4f* transition probability. These substitutional RE dopants are therefore likely to be the optically active RE centers observed in GaN:RE. A preliminary model of the GaN:RE crystal structure has been developed as part of a collaboration with scientists at Bell Laboratories and in Europe. A simple view of the structure is shown in **Fig. 2** A strongly bonded GaN lattice, in conjunction with substitutional incorporation allows[10] unusually high RE doping concentrations (up to ~3-5 at.%), while preserving the optical activation of RE dopants. By comparison, the use of RE^{3+}-doped II-VI semiconductors as emitters and phosphors suffers from a more weakly bonded lattice, and substitutional location of the RE^{3+} ions on the 2^+ cation sites, which generates additional electrically-active defects due to lack of charge neutrality. RBS channeling analysis[11] has confirmed that a great majority (~90%) of the Er ions occupy substitutional sites on the Ga sublattice even at relatively high concentrations of >0.1 at.%. The Er-N bond has been measured by EXAFS analysis[12] to be 2.17 Å, versus a Ga-N bond length of 1.95 Å. This unusually short Er-nearest neighbor bond length in GaN is thought to be due to two major factors: (a) the low 4-fold coordination, compared for example to a 12-fold coordination in $ErSi_2$; (b) a more polar bond for the Er-N than for Ga-N (due to electronegativity differences), which helps to energetically compensate for the Ga - Er size mismatch.

Light emission from GaN:RE has been demonstrated via photoluminescence (PL), cathodoluminescence (CL), and electroluminescence (EL). The dominant mechanisms for excitation and subsequent relaxation of RE dopants in GaN are depicted in **Fig. 3**. In PL electron-hole pairs are generated by above band-gap photon absorption, carrier generation is provided by a high energy electron beam in CL, and in EL carrier injection

occurs by the application of bias voltage to electrical contacts on the GaN layer.

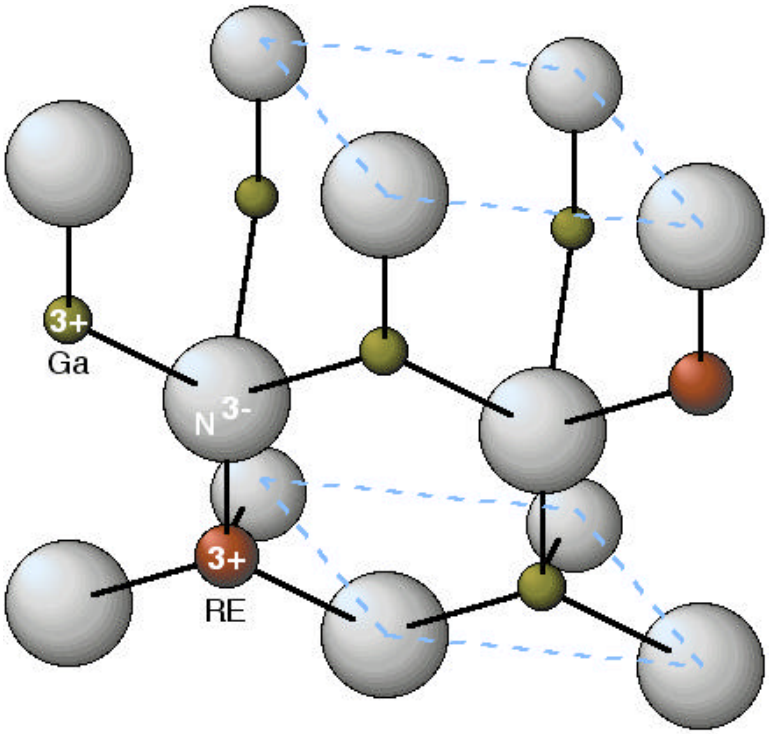

Fig. 2 Preliminary model of the GaN:RE crystal structure. The Er-N bond has been measured to be 2.17 Å, versus a Ga-N bond length of 1.95 Å.

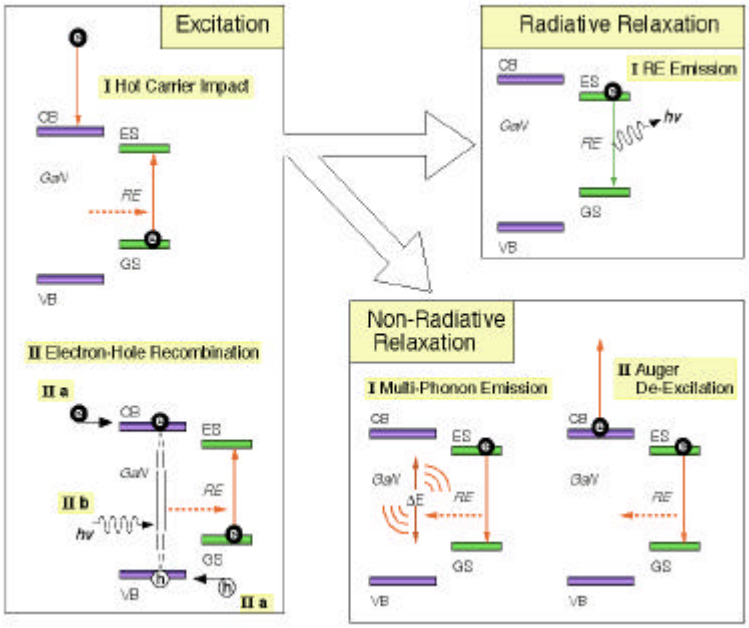

Fig. 3 The dominant mechanisms for excitation and subsequent relaxation of RE dopants in GaN. Shown schematically in the diagrams are the conduction band (CB) and valence band (VB) edges of GaN and the ground state (GS) and excited state (ES) for RE transitions.

IV. Electroluminescent Devices

The form of EL that the GaN:RE ELDs utilize involves impact excitation of RE dopants. In order for hot carrier excitation to occur, a high electric field is applied across the GaN layer. The minimum field strength required for GaN:RE ELDs is an order of magnitude lower than the 1-2 MV/cm required for II-VI:RE based ELDs. This lower field requirement results in light emission at much lower voltages (as low as ~5V) compared to ELDs based on other semiconductor materials. In **Fig. 4**, a low voltage GaN:Er ELD is shown emitting green light at a bias of 7.5 V, along with a corresponding graph of current and light intensity vs. applied voltage. The device consists of an Er-doped GaN layer grown by MBE on a n+-Si substrate and a top-side circular ("ring contact") transparent bias electrode of In-Sn oxide (ITO). The combination of this very simple device structure with the use of Si *substrate* indicates the potential of this technology for low-cost displays and for integration with Si technology for drive and control circuitry.

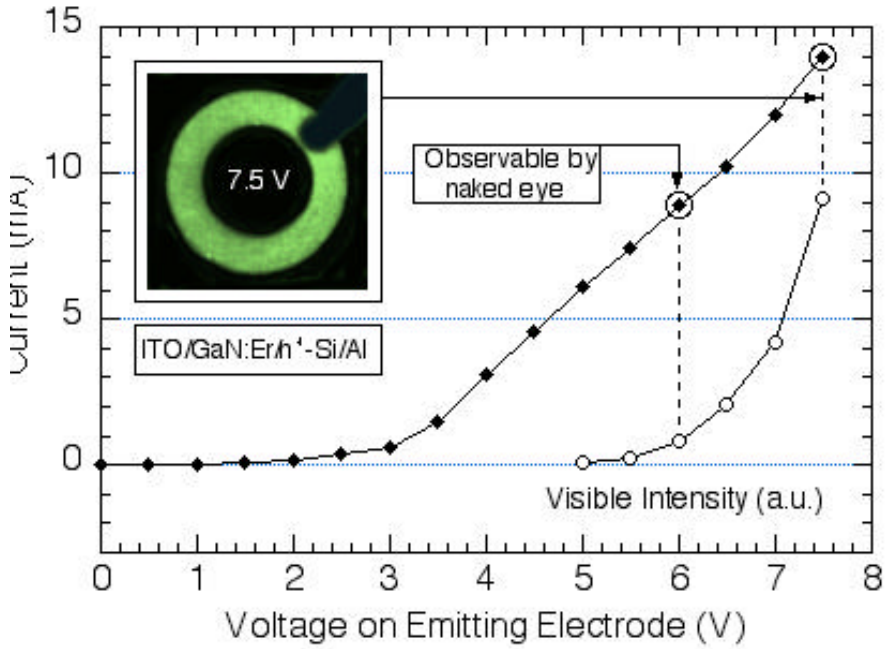

Fig. 4 GaN:Er ELD current and EL intensity as a function of voltage on emitting electrode. A thin (~300 nm) GaN layer and n^+ Si substrate are used to obtain low-voltage optical turn-on at 6V. The inset is a photograph of green emission under the ITO bias electrode at 7.5 V.

V. 1.5 µm Operation

A second major application of RE-doped GaN is in the area of fiber optic telecommunications, which has seen explosive growth in terms of both new network installation and network bandwidth. Fiber optic signals carry near-IR wavelengths, primarily 1.5 µm and secondarily 1.3 µm, which are the wavelengths of minimum loss and minimum dispersion in silica fibers. Fiber optic sources and amplifiers have been developed using primarily Er^{3+} and Pr^{3+} doped in a variety of glasses. Semiconductor-based equivalents that could take advantage of semiconductor technology for miniaturization and operation, have suffered from low solubility and thermal quenching. Extensive research has been carried out on Er-doped Si [13,14]. The IR radiation is nearly completely quenched at temperatures around 200 K. The addition of a significant oxygen concentration has been shown to improve the Er temperature quenching in silicon[15,16] but at the expense of a more complicated process and degraded electrical properties. An increasing semiconductor bandgap has been shown to reduce temperature quenching[17]. GaP, which has a larger bandgap than Si, still exhibits[18] a reduction in EL intensity at 300K compared to low temperature values. In Er-doped GaN, the thermal

quenching is for all practical purposes, no longer a problem. **Fig. 5** shows the temperature dependence of the 1.5 µm electroluminescence of a GaN:Er ELD. Notice that the EL intensity actually peaks at around 375 K. Furthermore, allowing for only a 10% degradation of the peak emission value results in an operating temperature range from 275K to 410K. This is important especially when considering the temperature quenching of the IR light emission in other semiconductor hosts. We, therefore, conclude that GaN has a significant advantage as a host for erbium compared to GaAs, GaP and Si, which have been extensively studied for Er-based light emitting applications. In the case of GaN:Er, it appears that devices could be developed for semiconductor light sources and amplifiers of 1.5 µm signals.

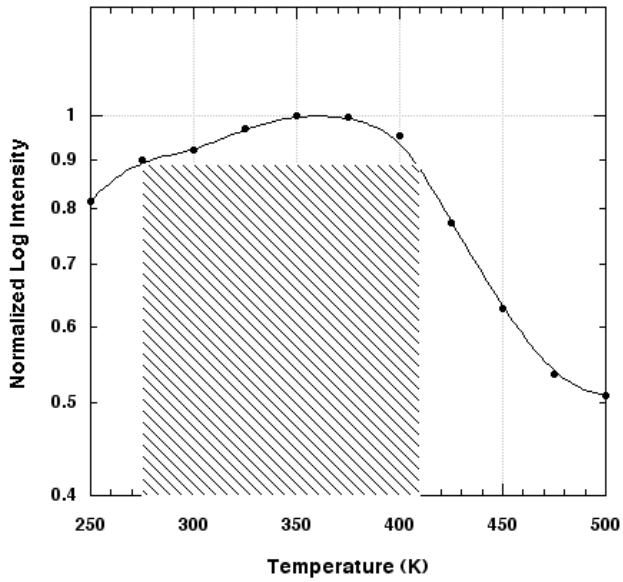

Fig. 5 Temperature dependence of the 1.5 µm electroluminescence of a GaN:Er ELD. The shaded region corresponds to 10% degradation of the peak emission (275K to 410K).

VI. Pure and Mixed Colors

The versatility of the GaN:RE technology is illustrated using the CIE chromaticity diagram shown in **Fig. 6**. The triangle in the diagram connects the three primary visible colors emitted by the GaN doped with Tm (blue), Er (green), and Eu and Pr (red). The coordinates of this triangle match very well the standard primary colors of the NTSC. In addition to primary colors, mixed colors or hues can be obtained by incorporating multiple REs into the GaN layer, whose combined emission is perceived by the eye as a range of colors depending on the relative intensities of the constituents. Examples of mixed colors shown in **Fig. 6** are a combination of Er and Tm yielding the cyan color and Er and Eu producing orange and yellow colors. The ability of RE-doped GaN technology to cover the visible color spectrum with both primary and mixed colors makes it a strong candidate for a variety of display and lighting applications.

In conclusion, on this contract we have developed a very versatile and robust rare-earth-doped GaN technology for both optical communications and visible displays. We plan to further develop RE-doped GaN devices as part of a follow-on contract. Ultimately, we foresee the insertion of this technology into significant applications of interest to the Department of Defense and to the commercial market.

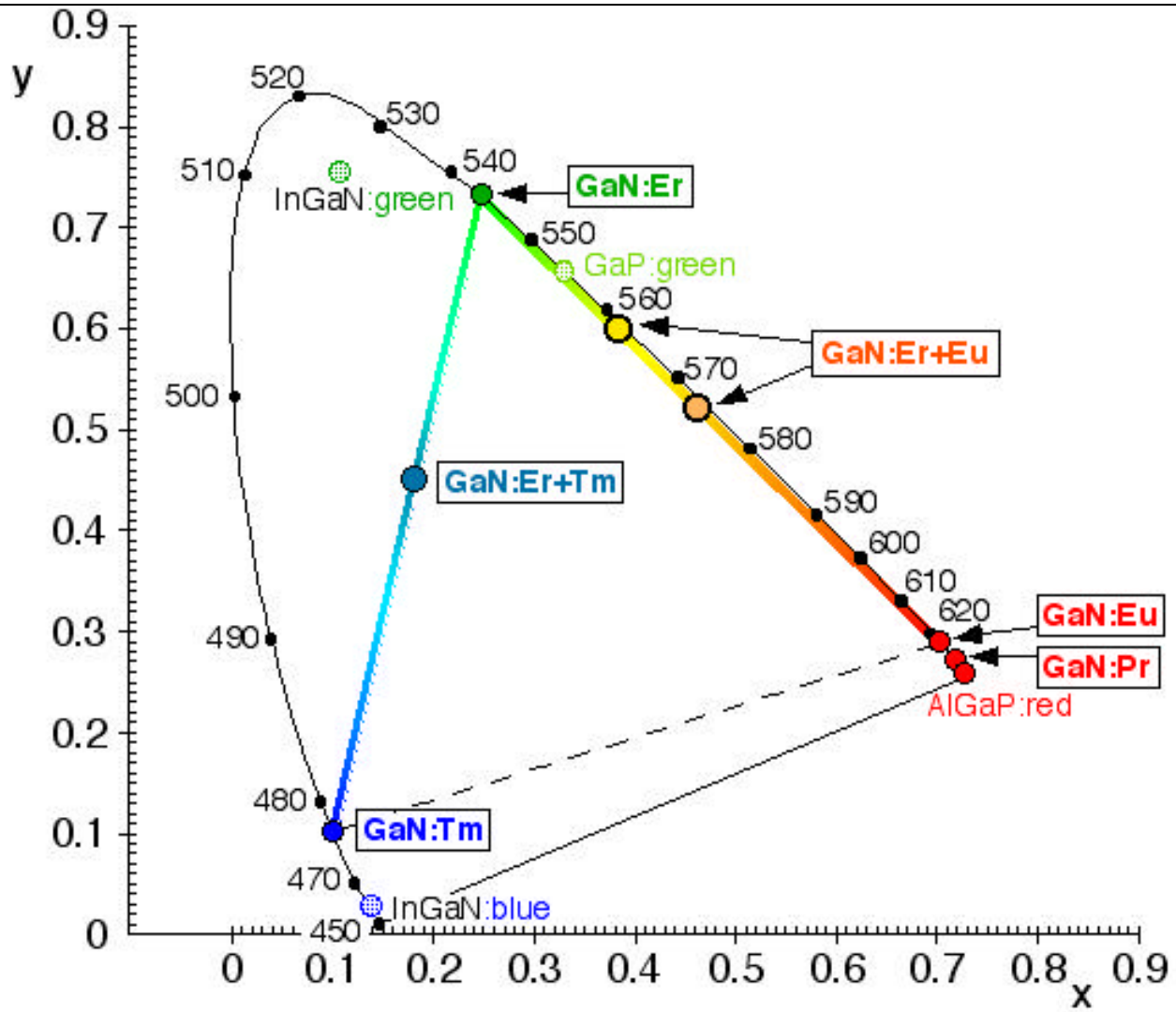

Fig. 6 CIE x-y chromaticity diagram showing the location of pure color emission from GaN ELDs doped with Tm (blue), Er (green), Eu (red) and Pr (red) and of mixed color emission using GaN:Er+Tm (cyan) and yellow and orange emission from the GaN:Er+Eu ELD. Also shown are the coordinates of other commercial LEDs.

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VII. Listing of Publications Under This Contract

(a) Papers published in peer-reviewed journals:

1. A.J. Steckl and R.H. Birkhahn, "Visible Emission from Er-doped GaN Grown by Solid Source Molecular Beam Epitaxy", Appl. Phys. Lett., Vol. 73, pp. 1700-1702, Sept. 1998.

2. A.J. Steckl and I. Chyr, "Focused Ion Beam Micromiling of GaN", J. Vac. Sci. and Technol. B, Vol. 17, pp. 362-365, March/April 1999.

3. A.J. Steckl, M.J. Garter, R.H. Birkhahn and J. Scofield, "Green Electroluminescence from Er-doped GaN Schottky Barrier Diodes", Appl. Phys. Lett., Vol. 73, pp. 2450-2452, Oct. 1998.

4. R.H. Birkhahn and A.J. Steckl, "Green Emission from Er-doped GaN Grown by Molecular Beam Epitaxy on Si Substrates", Appl. Phys. Lett., Vol. 73, pp. 2143-2145, Oct. 1998.

5. M.J. Garter, J.D. Scofield, R.H. Birkhahn and A.J. Steckl, "Visible and Infrared Emission from ITO/GaN:Er/Si Schottky Diodes", Appl. Phys. Lett. Vol. 74, pp. 182-184, Jan. 1999.

6. F.J. Pacheco, A.M. Sanchez, S.I. Molina, D. Araujo, J. Devrajan, A.J. Steckl and R. Garcia, "Electron Microscopy Study of SiC Obtained by Carbonization of Si (111)", Thin Solid Films, Vol. 343-344, pp. 305-308, 1999.

7. R.H. Birkhahn, M.J. Garter and A.J. Steckl, "Red Light Emission by Photoluminescence and Electroluminescence from Pr-doped GaN on Si Substrates", Appl. Phys. Lett. Vol. 74, pp. 2161-2163, Apr. 12, 1999.

8. L.C. Chao and A.J. Steckl, "Development of an Er-Ni Liquid Metal Ion Source", J. Vac. Sci. Technol. B Vol. 17, pp. 1051-1053, May 1999.

9. R.H. Birkhahn, R.A. Hudgins, D.S. Lee, A.J. Steckl, R.J. Molnar and J. M. Zavada, "Growth and Morphology of Er-doped GaN on Sapphire and HVPE Substrates", J. Vac. Sci. Technol.B, Vol. 17, pp. 1195-1199, May/Jun 1999.

10. R.H. Birkhahn, R. Hudgins, D.S. Lee, A.J. Steckl, A. Saleh, R.G. Wilson and J.M. Zavada, "Optical and Structural Properties of Er^{3+} - Doped GaN Growth by MBE", MRS Internet J. Nitride Semicond. Res. **4S1**, G3.80, 1999.

11. I. Chyr and A.J. Steckl, "Focused Ion Beam Micromachining of GaN Photonic Devices", MRS Internet J. Nitride Semicond. Res. **4S1**, G10.7, 1999.

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13. L.C. Chao and A.J. Steckl, "Room Temperature Visible and Infrared Photoluminescence from Pr-Implanted GaN films by Focused Ion Beam Direct Write", Appl. Phys. Lett. Vol. 74, pp. 2364-2366, Apr. 19, 1999.

14. J. Chen, A. J. Steckl, and M. J. Loboda, "*In-Situ* N_2 Doping of SiC films Grown on Si(111) by Chemical Vapor Deposition from Organosilanes", J. Electrochem. Soc., Vol. 147 (6), pp. 2324-2327, June 2000.

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(b) Papers published in conference proceedings:

1. F. J. Pacheco, A. M. Sanchez, S. I. Molina, D. Araujo, R. Garcia, and A. J. Steckl, “Effect of Temperature Ramp Rate During Carbonization of Si (111) on the obtained SiC Crystalline Quality”, Proc. Microscopy of Semicond. Materials XI Congress (Oxford, England), March 1999.

(c) Papers presented at meetings, but not published in conference proceedings:

1. A. J. Steckl, “Focused Ion Beam Fabrication of Optoelectronic Components", ARO Workshop on Low Power Optoelectronics, Lake Arrowhead, CA, Jan. 1996.

2. R. Gass, P. Chen, A. J. Steckl, H. E. Jackson, "Model for Selective Compositional Mixing in Al GaAs/GaAs Superlattices Induced by FIB Implantation", American Physical Society Meeting, St. Louis, MO, March 1996.

3. A. J. Steckl, J. Devrajan, C. Tran, Z.C. Feng, and R. Karlicek, “SiC SOI Substrates for GaN Growth”, 3rd Nitride Workshop, St. Louis, Mo., March, 1996.

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7. A.J. Steckl, J.Devrajan, C. Tran, R.A. Stall, “SiC Based Substrates for Growth of GaN Thin Films”, 4th Wide Bandgap Nitride Workshop, St. Louis, Mo., March 1997.
8. A.J. Steckl, “A Review of Focused Ion Beam Implantation for the Fabrication of Optoelectronic Components”, Invited Paper, Scanning Microscopy International Conference, Symposium on Nanometer Ion Beams, Chicago, May 1997.

9. J. Devrajan, A.J. Steckl, C. Tran, and R.A. Stall, “Optical Properties of GaN Films Grown on SiC/Si”, Int’l Conf. SiC and III-Nitrides, Stockholm, Sept. 1997.
10. J. Chen and A.J. Steckl, “Heteroepitaxial Growth of SiC on Si by Gas Source MBE with Silacyclobutane”, Int’l Conf. SiC and III-Nitrides, Stockholm, Sept. 1997.

11. J. Chen and A.J. Steckl, “MBE Growth of SiC on Si (111) with SCB”, North American Conf. on Molecular Beam Epitaxy, Ann Arbor, MI, Oct. 1997.

12. J. Chen, R. Birkhahn and A.J. Steckl, “Growth of SiC and GaN Thin Films by Gas Sources MBE”, Ohio MatNet Symposium, Bowling Green, OH, Nov. 1997,

13. R.H. Birkhahn, R.A. Hudgins, D. Lee, A.J. Steckl, R.J. Molnar and J.M. Zavada, “Growth and Morphology of Er-doped GaN on Sapphire and HVPE Substrates”, North American MBE Conference, State College, PA, Oct. 1998.

14. A. J. Steckl, R. Birkhahn, M. Garter, “Rare Earth Luminescence in GaN and Related Device Performance”, Materials Research Society Meeting, San Francisco, CA, April 1999.

15. A. J. Steckl, M. Garter, and R. Birkhahn, “Visible and Infrared Electroluminescence from Er-doped GaN Schottky Diodes”, Device Research Conference, Santa Barbara, CA, June 1999.

16. A. J. Steckl, R. Birkhahn, M. Garter, L. C. Chao, D. L. Lee and J. Heikenfeld, “Optical Properties of Rare-Earth-Doped GaN and Related Light Emitting Devices”, 41st Electronic Materials Conference, Santa Barbara, CA, July 1999.

(d) Manuscripts submitted but not published during reporting period:

1. A. J. Steckl, M. Garter, D. S. Lee, J. Heikenfeld, and R. Birkhahn, “Blue Electroluminescence from Tm-doped GaN Light Emitting Devices”.

2. A. J. Steckl and J. M. Zavada, “Optoelectronic Properties and Applications of Rare-Earth-Doped GaN”

3. J. Heikenfeld, M. Garter, D. S. Lee, , R. Birkhahn, and A. J. Steckl, “Red Light Emission by Photoluminescence and Electroluminescence from Eu-doped GaN”.

4. L.C. Chao, B. K. Lee, C. J. Chi, J. Cheng, I. Chyr, and A. J. Steckl, “Upconversion Luminescence of Er-Implanted GaN Films by FIB-Direct Write”, Appl. Phys. Lett.

5. A. J. Steckl and J. M. Zavada, “Photonic Applications of Rare-Earth-Doped Materials”.

VIII. Participating Personnel

Ronald Birkhahn (completed Ph.D. in Feb.1999)
Dr. David Chao
Dr. John Chen
Michael Garter
Robert Hudgins
Jason Heikenfeld
Don Lee

IX. Report of Inventions

“Visible light emitting diode formed from wide bandgap semiconductor doped with a rare earth element”.