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13. ABSTRACT (Maximum 200 words)

A high resolution study of the optical emission from nitrogen-plasmas produced by an Oxford Applied Research radio frequency plasma source is reported.
The use of electron cyclotron resonance (ECR) plasma sources has led to recent advances in the growth of GaN and other III-V nitride semiconductors by molecular beam epitaxy (MBE).
Methods have been developed bor the MBE-growth of integrated heterostructure devices containing both narrow-band-gap and wide-band-gap II-VI materials. Device processing procedures were also developed.

14. SUBJECT TERMS

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Final Technical Report

Blue Semiconductor Lasers Based on Wide-Band-Gap II-VI Materials

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I. SUMMARY OF RESEARCH RESULTS

NITROGEN PLASMA DOPING. A high resolution study of the optical emission from nitrogen-plasmas produced by an Oxford Applied Research radio frequency plasma source is reported. Strong atomic emission lines in the near-infrared spectral region provide unequivocal evidence that the nitrogen-plasma source generates an appreciable flux of nitrogen atoms. Nitrogen atoms, rather than N_2 molecules in the plasma, are the most-likely nitrogen-plasma species responsible for the successful p-type doping of ZnSe:N and related alloys. Emission spectra from the rf nitrogen-plasma source are compared with similar emission spectra obtained from an electron cyclotron resonance microwave nitrogen-plasma source.

The use of electron cyclotron resonance (ECR) plasma sources has led to recent advances in the growth of GaN and other III-V nitride semiconductors by molecular beam epitaxy (MBE). These materials show promise for high-power and high-temperature electronic applications, and short wavelength optoelectronic applications because of their large direct band gaps and structural stability. In an effort to better understand the physical properties of nitrogen-plasmas produced by an MBE-compatible ECR source, the optical emission from an ASTeX compact ECR microwave plasma source was investigated. High resolution optical emission spectra clearly show that the ASTeX ECR plasma source produces an appreciable flux of nitrogen atoms in addition to various species of molecular nitrogen. Atomic nitrogen production as a function of source input power, ECR magnet current, and N_2 gas flow rate is reported.

Variable-temperature photoluminescence (PL) experiments have been performed for a series of nitrogen-plasma-doped ZnSe and ZnSSe thin films. Values for the acceptor ionization energy E_a and compensating donor ionization energy E_d, as calculated from the PL data, were found to be temperature dependent. The PL data also indicate that luminescence from the electron-acceptor (e,A) and donor-hole (D,h) transitions is linearly polarized along the [011] direction. These results provide evidence that nitrogen doping of ZnSe and ZnSSe may not produce a simple hydrogenic acceptor at high doping levels. Rather, the nitrogen may be incorporated in asymmetric, non-tetrahedral configurations. Our results suggest that at high doping levels interstitial nitrogen may play an important role in both the p-type doping and compensation processes in these materials, as has been proposed by Chadi and Troullier.

OHMIC CONTACTS. The presence of large barriers is manifested by the need to apply very large voltages (10-20 V or more) to activate both n-on-p and p-on-n ZnSe-based light emitting diodes (LEDs) and laser diodes (LDs). In the case of n-on-p structures, the forward-biased diode characteristics are dominated by tunneling at the reverse-biased ZnSe/GaAs interface, where a barrier ~1.4 eV occurs as a consequence of the large valence band offset at the hetero-interface between these two materials, as is illustrated schematically in fig. 2(a). Similar tunneling, with the requisite large applied voltages, also occurs for p-on-n ZnSe-based diodes grown on n-type GaAs substrates. In this case the forward-bias characteristics of the diode are dominated by tunneling phenomena at the reversed-biased metal semiconductor contact, as shown in fig. 2(b).

At North Carolina State University (NCSU), we have addressed the above fundamental issue in a novel way by employing an epitaxial layer of the semimetal HgSe to decrease the interfacial energy barrier (valence band offset) to about 0.6 eV. This estimate is based on a "modified" common-anion rule for the mercury calcogenides which assumes a valence band offset of $\Delta E_v \sim 0.2\ E_g$, where E_g is the band gap of the corresponding wide band gap common-anion II-VI material (cadmium or zinc chalcoginide). Correspondingly, the conduction band offset is given by $\Delta E_c \sim 0.8\ E_g$. This empirical "80%-20%" rule holds in the case of CdTe (E_g = 1.5 eV) and HgTe for which a valence band offset ΔE_v ~0.3 eV has been measured [4]. It also holds approximately for ZnTe (E_g = 2.35 eV) and HgTe for which ΔE_v = 0.3 - 0.4 eV has been reported .

The use of epitaxial HgSe has resulted in improved ohmic contacts for p-type ZnSe films and related diode structures;'. Multilayered structures with intermediate or graded layers of $Hg_{1-x}Zn_xSe$ or $ZnTe_{1-y}Se_y$ have also been fabricated to further improve the electrical properties of II-VI light emitters. We refer to such structures as *integrated heterostructure devices (IHDs)* [5,6]. The term integrated heterostructure or integrated heterostructure device is here defined as a multilayered structure in which particular layers, or combination of layers, perform distinctly different functions. An example of an IHD is a semiconductor surface-emitting laser which contains (a) multilayers for optical mirrors, (b) an active light generation region which might consist of one or more additional layers or quantum wells, (c) p-type and n-type layers which supply the active light generation region with electron and holes under forward bias, and (d) additional top layers for optically and electrically coupling the laser output to the outside world. These various functions are integrated into a single epitaxial multilayered structure using sophisticated growth techniques such as molecular beam epitaxy (MBE) or metal-organic chemical vapor deposition (MOCVD). The p-on-n IHD structures being studied at NCSU consist of: (a) multilayers of II-Vi compounds that form an optical-emission heterostructure, (b) abrupt or graded II-VI layers that serve as an electrical bridge to the p-type contact layers, and (c) HgSe/metal contacts.

This grant provided the funds for methods that have been developed at NCSU for the MBE-growth of integrated heterostructure devices containing both narrow-band-gap and wide-band-gap II-VI materials. Device processing procedures were also developed. Properties of light-emitting devices were described within the context of IHD design principles, including those of recently-fabricated high-brightness, high-efficiency blue and green LEDs [7]. The feasibility of using other II-VI semimetals such as HgTe, HgS, HgSSe, or HgSeTe as electrical contact layers in particular light-emitting and/or light-detecting IHD structures was also addressed.

HALL EFFECT STUDIES. Van der Pauw Hall effect measurements from 77-350 K WERE reported for a series of p-type nitrogen-doped ZnSe thin films. Epitaxial HgSe electrodes were used as ohmic contacts in these experiments. Hole mobilities of 7.5 - 17.3 cm^2/V-s were obtained at 300 K. The hole mobility was found to increase with decreasing temperature. Hole concentrations as large as 8×10^{17} cm^{-3} at 300 K were obtained. Nonlinear least-squares fitting procedures were employed to fit the hole concentration versus temperature data in order to obtain values for the acceptor concentration N_a, the donor concentration N_d, and the acceptor ionization energy E_a for a series of p-type N-doped samples. Our results provide the first direct evidence that N-doping of ZnSe, using a remote-plasma source, does not produce a simple hydrogenic acceptor. Instead, plasma-doping may produce atomic complexes which give rise to a relatively shallow acceptor (E_a = 60-80 meV), but which also create atomic environments which presently limit the maximum p-type doping of ZnSe to $p_0 \sim 1 \times 10^{18}$ cm^{-3} because of compensation effects that accompany the doping process.

UV MULTILAYERS. Multilayers of $ZnS/ZnS_{1-x}Se_x$ and $ZnS/Zn_{1-x}Cd_xS$ were successfully grown by molecular beam epitaxy. Photoluminescence (PL) experiments were performed at 4.2 K to evaluate the optical quality and carrier-confinement properties of these multiple-quantum-well (MQW) structures. The PL emission from a $ZnS/Zn_{0.93}Cd_{0.07}S$ MQW structure consisted of a single sharp peak (FWHM=12 meV) at 3.736 eV. In general, emission peaks from the $ZnS/Zn_{1-x}Cd_xS$ films were much sharper and more intense than those from $ZnS/ZnS_{1-x}Se_x$ films. These PL characteristics indicate improved electron confinement by the Cd based wells due to an appreciable conduction band offset for $Zn_{1-x}Cd_xS$ with respect to ZnS. This research suggests that $ZnS/Zn_{1-x}Cd_xS$ quantum wells may be useful in the development of light emitting devices in the ultraviolet and blue/violet spectral region.

BLUE/GREEN LASER DIODES. The first high-resolution study of emission spectra from ZnSe-based blue-green laser diodes WAS reported. Analysis of the longitudinal (Fabry-Perot) mode spacing in output spectra from cleaved-cavity, stripe-geometry lasers having cavity lengths ranging from 185 -1100 μm yields values for the equivalent index of refraction n_e = 3.91 - 4.41. Under certain conditions, single mode output was observed. Several samples exhibited emission spectra with satellite peaks located adjacent to each longitudinal (Fabry-Perot) resonance which are associated with lateral resonances. Analysis of the lateral mode spacings yields $y_\varepsilon \sim 350$ μm, where y_ε is a characteristic length associated with the spatial fall-off of the refractive index in the junction plane.

Blue and green laser and light emitting diodes based on II-VI heterostructures have been grown on pre-deposited GaAs epilayers. Properties of blue laser diodes are discussed. Blue and green light emitting diodes which employ quaternary alloys of ZnCdSSe and ZnTeSSe as the active region of double heterostructure devices have also been prepared and tested.

Laser diodes have been fabricated by MBE which emit blue light at wavelengths as short as 467.5 nm at 77 K. This is the shortest wavelength ever obtained from a semiconductor diode laser. The lasers employ ZnCdSe quantum wells, ZnSe confinement layers, and ZnSSe cladding layers. Chlorine was used to dope the n-type layers of the structures; nitrogen from a plasma source was used as a p-type dopant. Lasing has been observed at temperatures up to ~200K, and cw emission has been achieved at 77K. Under appropriate excitation, some of the laser structures exhibit single-mode output. A high-resolution study of emission spectra from a series of laser diodes has been completed. Analysis of the longitudinal (Fabry-Perot) mode spacing in output spectra from cleaved-cavity, stripe-geometry lasers having cavity lengths ranging from 185 -1100 μm yields values for the equivalent index of refraction n_e = 3.91 - 4.41. Several samples exhibited emission spectra with satellite peaks located adjacent to each longitudinal (Fabry-Perot) resonance which are associated with lateral resonances. Analysis of the lateral mode spacings yields y_ε ~ 350 μm, where y_ε is a characteristic length associated with the spatial fall-off of the refractive index in the junction plane.

BLUE/GREEN LIGHT-EMITTING DIODES. Blue and green light emitting diodes (LED)s based on ZnSSe double-heterostructures were grown on (100) GaAs substrates by molecular beam epitaxy. The quaternary alloys ZnCdSSe and ZnSSeTe were used as active layer regions in the blue light emitting and green light emitting structures, respectively. The green LEDs produce 238 μW of light at room temperature when operating at 10 mA (3.8 V) and exhibit an external efficiency of 0.62 %. At 20 mA, the devices produce 428 μW of green light. The blue LEDs produce 213 μW of light when operating at 10 mA and exhibit an external efficiency of 0.57 % at room temperature. At 20 mA, the devices produce 370 μW of blue light.

High-brightness blue and green light-emitting diodes (LEDs) operating at peak wavelengths in the range 489 -514 nm have been successfully synthesized, processed, and tested. The high-brightness LEDs are II-VI heterostructures grown by molecular beam epitaxy (MBE) at NCSU using (100) ZnSe substrates produced at Eagle-Picher Laboratory by the Seeded Physical Vapor Transport (SPVT TM) process. Substrate wafers were cut and polished from oriented 50 mm diameter by ~25 mm thick single-crystalline ZnSe ingots. The ZnSe wafers are twin-free and contain no small-angle grain boundaries. Etch pit densities range from 2 - 5 x 10^4 cm^2. Double crystal x-ray diffraction rocking curve studies yield FWHM (400) = 11-16 arc sec, indicating ZnSe crystal quality comparable to that of GaAs substrates. The double-heterostructure (DH) LED devices consist of a ~1.8 μm thick layer of n-type ZnSe:Cl, a ~0.1 μm thick active region consisting either of a ZnCdSe MQW (blue) or a ZnTeSe layer (green), and a ~1.8 μm thick p-type ZnSe:N layer deposited using a nitrogen plasma source. Thin (~100 Å) epitaxial surface layers of HgSe/ZnTeSe were deposited by MBE to obtain excellent ohmic contact to the top p-type ZnSe layer, as shown in Fig. 1. Standard photolithographic techniques were used to fabricate 250 μm x 250 μm mesa diode structures. Gold was used as a metal contact to the top HgSe layer of each device; indium was used to contact the n-type ZnSe layer. Direct contact to the n-type ZnSe epilayer was necessitated because of the insulating nature of current ZnSe substrates. The devices were packaged in a standard T-13/4 clear-epoxy lamp configuration for testing. The blue LEDs produce 327 μW @ 10 mA with external quantum efficiency of 1.3%. We have compared the ZnCdSe and Nichia Chemical InGaN blue LED characteristics. It is seen that the Nichia device is about three times brighter but displays a much broader spectral output. In contrast, the ZnCdSe blue LED output is sharply peaked at 489 nm with a spectral purity of 96%. In terms of photometric units, the luminous performance of the ZnCdSe LED is 1.6 lumens/watt at 10 mA. The brightest ZnTeSe green LEDs tested to date produce 1.3

mW at 10 mA peaked at 512 nm with an external quantum efficiency of 5.3% . The luminous performance of the green LED is 17 lumens/watt at 10 mA. This exceeds the luminous performance of super-bright red LEDs (650 nm) based on AlGaAs DHs and greenish-yellow (570 nm) AlGaInP DH devices; it also exceeds the luminous performance of the super-bright Nichia blue LEDs based on GaN/InGaN DHs (3.6 lumens/watt). The green LEDs which we are reporting are more than fifty times brighter than commercial GaP LEDs which produce outputs peaked in the yellow-green at 555 nm. At NCSU, our best green LEDs presently have useful lifetimes (40% degraded) of more than 750 hr when operated at 10 mA -- although several have exceeded 2000 hrs. Improvements in nucleation and growth are expected to increase the device lifetime substantially in the months ahead. For a given defect density, the degradation process appears to depend on the total charge/unit area flowing through the device. Thus, it may be expected that initial commercial-grade devices will operate with high efficiency and low currents (~0.5 mA; 65 μW), for which there is a substantial market, in order to insure long lifetimes.

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