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13. ABSTRACT *(Maximum 200 words)*

Semiconductor interfaces such heterojunctions and metal/semiconductor contacts are crucial components of modern solid state electronic devices ranging from MOSFETs and MODFETs to LEDs and lasers. The possibility of tuning the interface potential barrier to a given device application and optimizing carrier injection or confinement would offer an important new degree of freedom in device design. The focus of our work was the characterization and control of the band alignment in a number of interfaces of current interest for the development of a viable blue optoelectronic technology. These included wide gap II-VI and III-V semiconductor materials fabricated by molecular beam epitaxy (MBE) on GaAs wafers. We have exploited the MBE growth parameter to vary the interface composition and morphology, and examined the resulting variations in the structural and electronic properties of the junctions. Among the major successes of our program was the fabrication of local interface dipoles in the interface region of II-VI/III-V heterojunctions to independently tune band offsets and structural properties, the implementation of metal/ZnSe contacts with unpredentedly low resistivity, and the development of new substrates lattice-matched to the active layer for the fabrication of II-VI based blue-green lasers.

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## 1. STATEMENT OF THE PROBLEM

Semiconductor interfaces such heterojunctions and metal/semiconductor contacts are crucial components of modern solid state electronic devices ranging from MOSFETs and MODFETs to LEDs and lasers. The possibility of tuning the interface potential barrier to a given device application and optimizing carrier injection or confinement would offer an important new degree of freedom in device design. The focus of our work was the characterization and control of the band alignment in a number of interfaces of current interest for the development of a viable blue optoelectronic technology. These included wide gap II-VI and III-V semiconductor materials fabricated by molecular beam epitaxy (MBE) on GaAs wafers, II-VI/II-VI heterostructure and multiple quantum wells, as well as metal/semiconductor contacts. On the one hand, little was known about the basic properties of such interfaces, and the possibility that the local interface environment might affect the band alignment. On the other hand, the development of a viable blue optoelectronic technology is currently hindered by the insufficient performance of such interfaces, which leads to high dissipation and low device lifetime. We have exploited the MBE growth parameter to vary the interface composition and morphology, and examined the resulting variations in the structural and electronic properties of the junctions.

## 2. SUMMARY OF THE MOST IMPORTANT RESULTS

Much of our work has been focussed on heterovalent semiconductor heterojunctions, i.e. heterostructures and superlattices comprised of materials with different chemical valence (e.g. II-VI/III-V or III-V/IV). Heterovalent semiconductor heterojunctions are becoming increasingly important for the development of future optoelectronic devices, and offer promise to be the most easily tunable class of semiconductor heterojunctions. This is because ideally abrupt heterovalent heterojunctions with polar orientation would be thermodynamically unstable, so that different interface reconstruction may develop to lead to neutral interfaces. The resulting different interface configurations may lead to widely different heterojunction parameters.

Our approach has been to use the growth parameters to kinetically favor one or the other of the different interface configurations, and modify the band discontinuities. The direction in which we have attempted to drive the interface reconstruction has been generally suggested by the results of first principle calculations of the band discontinuities as a function of the interface atomic configuration. The growth parameters at our disposal were those controllable during molecular beam epitaxy (MBE), namely the atomic or molecular fluxes of the elemental constituents, temperature, and substrate orientation. All heterostructures examined were therefore synthesized by MBE, and characterized in-situ by reflection high energy electron diffraction and photoemission spectroscopy, and ex-situ by x-ray diffraction, and transmission electron microscopy (TEM). In general, we determined by photoemission spectroscopy the composition profile achieved at the interface, the band discontinuities, and the charged versus neutral character of the interfaces.

We targeted a family of II-VI/III-V which include strained ZnSe/GaAs heterostructures as well as lattice-matched $Zn_{1-x}Cd_xSe/In_yGa_{1-y}As$ heterostructures, which are crucial components of new optoelectronic devices operating in the blue region of the visible spectrum. For such heterojunctions, the large value reported for the valence band offset hinders hole injection in the active region for LEDs and lasers. However, first principle calculations suggest that a 1-eV wide range of band offsets might be achieved by varying the local atomic configuration at the interface. Experimentally we found that the Zn/Se beam pressure ratio (BPR) employed in the early stage of MBE fabrication of the heterostructure determines the local composition of the interface. High BPRs produce a Zn-rich interface composition, while low BPRs yield a Se-rich interface composition. The excess Zn or Se at the interface is of the order of one monolayer. Using synchrotron radiation photoemission spectroscopy for enhanced sensitivity we found that high Zn/Se flux ratios during the early stages of growth give rise to relatively abrupt interfaces, with little evidence of atomic intermixing, and an apparently Zn-rich Zn/Se relative composition.

Correspondingly, valence band offsets as high as 1.32-1.34eV were observed. Conversely, low Zn/Se flux ratios lead to enhanced atomic intermixing, with evidence of Ga-Se and As-Se bond formation, Ga outdiffusion in the overlayer and As surface segregation. The apparently Se-rich Zn/Se relative composition was accompanied by valence band offsets as low as 0.48-0.55eV. Such a wide range of tunability of the interface parameters is totally unprecedented, and could be exploited to substantially increase hole collection efficiency in blue LEDs and lasers.

Tuning the band alignment to optimize hole injection could have a substantial impact on device performance, but unfortunately comes at a price. The BPR employed controls also the optical properties of the ZnSe layer, and minimum incorporation of extended and point defects, as well as unintentional dopants is found in samples fabricated with near-stoichiometric growth conditions (BPR~1). Lowest densities of stacking faults are observed for Zn-rich growth conditions (BPR>1). Fabricating Se-rich interfaces requires BPR's much lower (~0.1) than those necessary to optimize such properties. We developed, however, a promising solution to this problem. We exploited the local character of the parameters that control the band offset and confine non-stoichiometric growth to a thin composition-control interface layer (CIL). We showed 2nm-thick CILs are sufficient to set the desired value of the band offsets, and that the band alignment does not change during the following growth stages in near-stoichiometric conditions. We used optical transmission measurements, complemented by photoluminescence studies, to demonstrate that the optical properties of CIL-incorporating heterostructures were dramatically improved as compared to those of structures fabricated in non-stoichiometric conditions.

We also conducted a series of studies on the stability of such engineered heterojunctions, in collaboration with colleagues at Xerox Corporation. ZnSe-GaAs heterojunctions were probed for deep level emission as a function of temperature by means of photoluminescence and cathodoluminesce spectroscopy. A series of previously unreported deep levels at 0.8, 0.98, 1.14, and 1.3 eV were identified which are characteristic of the ZnSe overlayer, and relatively independent in energy of overlayer thickness. These levels lie far below those of the near-band-edge features commonly used to characterize the ZnSe crystal quality. The relative intensity and spatial distribution of the deep level emission was found to be strongly affected by the BPR employed during ZnSe growth. In heterostructures fabricated in Se-rich growth conditions, that minimize the valence band offset and the concentration of Se vacancies, the dominant deep level emission is at 1.3 eV. For heterostructures fabricated in Zn-rich growth conditions, emission by multiple levels at 0.88, 0.98, and 1.14 eV dominates. Additional deep levels at 1.9-2.0eV appear related to thermal degradation of the interface and interdiffusion. The spectral energies and intensities of these deep level transitions might provide a new and improved characteristic indicator of ZnSe epilayer stoichiometry and near-interface defect densities.

The above in-situ studies were complemented by transport studies in which interfaces with different compositions were incorporated in functional n-ZnSe/p-GaAs heterojunction diodes. The goal of this second type of study was twofold. First, to gauge directly the effect of the local interface composition on the conduction-band offset. Second, to verify if engineered band offsets are observable in a complete device, having survived the following stages of device fabrication and processing. We designed heterostructures in which carriers could be photogenerated only in a 10nm-thick GaAs layer near the interface to avoid field induced transfer into GaAs satellite valleys before tunneling. From the systematics of the low-temperature current-voltage (I-V) spectra, and of the corresponding tunneling current, we deduced conduction band-offset values in remarkable agreement with those measured by electron spectroscopy in the thin-overlayer samples. This suggests that the metastable interface configurations responsible for band offset tuning in the thin overlayer samples are sufficiently stable to sustain the following stages of device fabrication and processing. Engineered band offsets can therefore be observed and exploited in practical devices. This was, to our knowledge, the first verification of a locally engineered heterojunction band offset in a fully functional device by transport methods. The only previous instance we are aware of dates back to the seminal work by Capasso et al. who used, however, doping-related dipoles as opposed to local modifications of the atomic configuration of the interface.

Our understanding of wide-gap II-VI semiconductor heterostructures lead us to develop a new method to obtain low-resistance metal/n-ZnSe contacts through fabrication of a Cl-doped graded n-$Zn_{1-x}Cd_xSe$ interface layer. Our interest in the effect of $Zn_{1-x}Cd_xSe$ interfacial layers on the properties of Al/$n^+$-ZnSe junctions was stimulated by three main considerations: 1)The ternary alloy $Zn_{1-x}Cd_xSe$ can be grown epitaxially on ZnSe by MBE and exhibits a bandgap which decreases monotonically with increasing Cd concentration x; 2)Recent studies indicate that the conduction band discontinuity accounts for most (70-80%) of the $Zn_{1-x}Cd_xSe$/ZnSe bandgap difference; and 3)Among the technologically relevant metals, aluminum is known to exhibit one of the lowest Schottky barriers to n-type ZnSe. We first performed SXPS studies of Al/$Zn_{1-x}Cd_xSe$ Schottky barriers fabricated in ultra-high-vacuum conditions. We found that the p-type Schottky barrier varies only slightly with x, and that most of the bandgap variation in the alloy series is reflected by the parallel variation in the n-type barrier. We then performed I-V measurements on a number of Al/n-$Zn_{1-x}Cd_xSe$ contacts fabricated with standard photolithographic techniques and found that the specific contact resistivity decreased monotonically with increasing Cd concentration and doping level. The lowest contact resistance observed in the presence of the ternary interface layer ($1.5\pm0.5\times10^{-5}$ $\Omega\cdot cm^2$) was, to our knowledge, the lowest ever reported for a n-type ZnSe-based, wide gap semiconductor.

We pursued the elimination of the single remaining region of high strain in current II-VI based blue-green lasers, i.e., the lasing medium. Most lasers include $Zn_{1-x}Cd_xSe$ quantum wells with x=0.25 (blue-green emitters) or x=0.15 (blue emitters) as optically active materials. The deeper quantum wells (x=0.25) exhibit a better temperature-dependence of the current threshold, but shorter low-temperature device lifetime. This has been related to the substantial compressive strain existing in the quantum wells (about 1.3% for y=0.15 and 1.9% for y=0.25 in pseudomorphic wells) which would assist defect formation and propagation. Strain in the quantum well is mandated by the lattice mismatch of the ternary active layers with both ZnSe and GaAs substrates. It could be eliminated, provided that suitable substrates lattice-matched to the ternary alloy wells are developed, since the remaining components of the device - namely the cladding and waveguiding alloy layers - can be easily grown lattice-matched to the active layers rather than to GaAs. For appropriate value of the In concentration y, $In_yGa_{1-y}As$ alloys can be designed to match the lattice parameter of ZnSe or that of the $Zn_{1-x}Cd_xSe$ active layers in blue or blue-green lasers. However, high quality $In_yGa_{1-y}As$ wafers with $x\geq0.04$ are not available, because of homogeneity problems in high-x bulk single crystals, and growth of $In_yGa_{1-y}As$ buffer layers with optimized concentration and strain relaxation profile on GaAs seems the most promising option.

We focused on $Zn_{1-x}Cd_xSe$/$In_yGa_{1-y}As$/GaAs(001) heterostructures which included $In_yGa_{1-y}As$ epilayers with homogeneous composition y=0, 0.18 and 0.27 as well as graded-composition layers in which x was gradually varied from x=0 to $x_s$=0.05, 0.23 and 0.33. The ternary buffer layers were designed to obtain in-plane surface lattice constant matching that of $Zn_{1-x}Cd_xSe$ with x=0, 0.15 and 0.25. On such substrates, dislocation-free growth of thick $Zn_{1-x}Cd_xSe$ films was readily achieved both on homogenous-composition as well as on graded-composition buffers in which the indium concentration was varied a superlinear, parabolic profile. The more complex, graded-composition profile was found to retard the nucleation of new dislocations due to the grading of the strain and reduces dislocation interaction, since the dislocations are distributed throughout the graded volume, and this leads to lower threading dislocation densities in both the III-V buffer and the II-VI active layer. Photoluminescence studies indicated an improvement of over two orders of magnitude in the ratio of the near-band-edge versus defect-related emission as a result of the presence of the ternary buffer.

Progress in eliminating dislocations and strain from the laser structure may reduce but not eliminate the problem of early laser degradation, so that attention has also been focused on other types of native defects which are preferentially located at the II-VI/III-V interface and may contribute to device degradation. In particular, degradation during cw operation of pseudomorphic laser structures at room temperature has been associated with stacking faults at the II-VI/III-V interface. Such defects would propagate through the structure during growth and generate dislocation sources within the strained $Zn_{1-x}Cd_xSe$ active layers. The existence of relatively high densities of native stacking faults at II-VI/III-V interfaces has been known for some time and tentatively explained by mechanisms as different as island coalescence during II-VI epilayer growth, doping, or substrate roughness. Both intrinsic and extrinsic stacking faults, which are formed by removing or inserting, respectively, a portion of a closed-packed {111} plane have been reported, in pairs or as isolated defects, starting in all cases at or near the II-VI/III-V interface and propagating throughout the II-VI epilayer, bounded by Frank or Shockley partial dislocations.

We found evidence that the Zn/Se flux ratio employed during the early stages of ZnSe growth by MBE on GaAs as well as $In_yGa_{1-y}As$ surfaces has a profound effect on the resulting density of native stacking faults. We found that the use of low Zn/Se BPRs, i.e., Se-rich growth conditions, during interface fabrication yields stacking fault densities dramatically lower than those obtained with high Zn/Se BPRs. Since we had shown earlier that the BPR controls the interface composition and the band alignment, the present results indicate that the local atomic configuration of the interface contributes to determine the probability of stacking fault nucleation.

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## 3. REPORTABLE INVENTIONS

None.

## 4. PARTICIPATING SCIENTIFIC PERSONNEL SUPPORTED

2.1 G. Bratina, Research Associate
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2.3 D. Coldwell, Research Assistant
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