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Heterogeneous integration of CdS filters with GaN LEDs for fluorescence detection microsystems

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Abstract

Microassembly of a hybrid fluorescence detection microsystem by heterogeneous integration of a CdS thin-film filter, an (In,Ga)N thin-film blue LED, and a disposable PDMS microfluidic device onto a Si PIN photodetector substrate is described. The CdS thin film filter was deposited directly onto a photodetector by pulsed-laser deposition. A thin-film (In,Ga)N LED was then transferred by a novel “pixel-to-point” laser lift-off process from the sapphire growth substrate to the silicon photodetector substrate. The final integration step was achieved by positioning a disposable polymer microfluidic device onto the excitation/detection subsystem. Pixel-to-point transfer is potentially an enabling micromanufacture process for the fabrication of multicolor fluorescence-based biosensors and chemical detection microsystems.

Keywords: Heterogeneous integration; Fluorescence detection; Optical thin-film filter; Microsystem; Pixel-to-point transfer

1. Introduction

Laser-induced fluorescence (LIF) detection is a popular detection technique for bioanalytical detection applications because of its selectivity and high sensitivity [1]. Miniaturization of fluorescence detection components into microsystems would provide the advantages of portability and low manufacturing cost. In addition, miniaturization could enable new functionality. For detection components on microsystems, most researchers employ either avalanche photodiodes (APDs) [2] or PIN photodiodes [3,4], the advantage of the latter being that substantially lower voltages can be employed (<1 V, compared to 40–100 V). Since photodiodes do not allow wavelength-discrimination, separating the fluorescence emission photons selectively from the more intense excitation light becomes of paramount importance.

Thin-film interference filters such as distributed Bragg reflectors (DBRs) have been used by various groups to filter the excitation signal [4–6]. One inherent difficulty with this approach is that the fabrication process of the DBR is expensive, time-consuming, and complicated since it requires the precise control of deposition conditions to maintain film thickness specifications for many layers. Furthermore, such filters display undesirable angular dependence due to the Bragg design constraint [4].

Cadmium sulfide, with a room temperature absorption edge of 513 nm, is a viable candidate for an optical filter in bioanalytical detection applications in which the dye’s excitation is in the blue (470–490 nm) and emission in the green (510–530 nm). Being a direct bandgap semiconductor, it possesses a steep absorption edge, resulting in a sharp transition between absorbed and transmitted wavelengths. This is important because some dyes (e.g., YOYO) have Stokes’ shifts as small as 18 nm. Table 1 provides a list of Stokes’ shifts for several commercially relevant dyes.
In this paper, the heterogeneous integration of a CdS thin-film filter, an (In, Ga)N blue LED, a Si PIN photodetector, and a disposable PDMS microfluidic device is described for the development of a sensitive, portable fluorescence detection system.

2. Design and fabrication

A schematic of the heterogeneous integration of a CdS filter, an (In, Ga)N blue LED, and a disposable PDMS microfluidic device is shown in Fig. 1. Wiring to both the LED and photodetector are shown. Rather than employing microlenses to couple excitation beams into microchemical components [5], the fluorescence detection microsystem described here is considerably simpler, using direct, normal-incidence excitation from an integrated light source in close proximity (~2 mm) to the sample.

The excitation signal in the emission range of the dye is a significant source of noise limiting the sensitivity of on-chip LIF analysis systems. Placing the detectors on the same side as the excitation source minimizes the effect of the LED’s green component. An aluminum mirror beneath the LED eliminates backside emission and enhances the frontside excitation signal significantly. The LED’s light passes through the microfluidics after exciting the sample and the LED’s green component does not reach the detector. The emitted fluorescent signal from the sample is incident on the PIN detector from a direction opposite to the primary emission cone from the LED excitation source. Whereas if the detector were placed behind the microchannel (in the direct path of the light from the LED), the LED’s green component would be directly incident on the detector, significantly reducing the signal-to-background ratio. This approach also enables the replacement of disposable microfluidic channels since the LED, filter, and detector are on one monolithic substrate.

The process flow begins with PECVD deposition of 1500 Å of SiO2 onto a silicon photodiode substrate [7]. An indium tin oxide (ITO) film of 2000 Å thickness was sputtered at 300 W power in an RF sputtering system using gas flow rates of 95 sccm Ar and 5 sccm Ar:O2 (20% concentration of O2 in Ar). With these conditions, the deposition rate was approximately ~200 Å/min. A 1–3 μm CdS film was then deposited onto the ITO/SiO2 stack using pulsed laser deposition (PLD). The SiO2 layer also provides electrical insulation and a diffusion barrier, as the ITO is conductive and would otherwise short the p- and n-contacts of the LED. The (In, Ga)N LED, grown on sapphire due to epitaxial growth constraints, is transferred

Table 1

<table>
<thead>
<tr>
<th>Dye name</th>
<th>Excitation peak (nm)</th>
<th>Emission peak (nm)</th>
<th>Stokes shift (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fluorescein FITC</td>
<td>494</td>
<td>518</td>
<td>24</td>
</tr>
<tr>
<td>YOYO-1, YO-PRO-1</td>
<td>491</td>
<td>509</td>
<td>18</td>
</tr>
<tr>
<td>Lucifer yellow CH</td>
<td>428</td>
<td>536</td>
<td>108</td>
</tr>
<tr>
<td>Rhodamine 110</td>
<td>496</td>
<td>520</td>
<td>24</td>
</tr>
</tbody>
</table>

Note: Stokes shift can vary by an order of magnitude.

Fig. 1. (a) Schematic of the heterogeneous integration of a CdS filter with an (In, Ga)N LED, a Si PIN photodetector, and a microfluidic device. With electrical wiring as shown, biases of about 15 V for the photodetector and about 4.5 V for the LED were applied. (b) Similar schematic, highlighting light paths for the excitation and Stokes-shifted emission signals.
onto the pre-fabricated silicon photodiode substrate by a pixel-to-point double-transfer technique using excimer laser lift-off and Pd-In transient-liquid-phase bonding (Fig. 2). The pixel-to-point process involves three steps: (1) temporarily bonding the LED pixel to a specially designed pick-up rod with sapphire substrates facing up using Super Glue®, (2) removing the sapphire substrates using laser lift-off, and (3) permanently bonding the LED pixel to a designated area. Previous work has demonstrated that the laser lift-off and transfer process of (In, Ga)N LEDs does not degrade performance [8]. Fig. 3 shows the electroluminescence spectra of LEDs before and after lift-off and transfer.

Deposition of CdS by PLD has been previously demonstrated [9]. The technique employs a solid source (1.3 cm in diameter, and 0.65 cm thick) as in evaporation or sputtering. A laser pulse of 38 ns duration and a fluence of 3 J/cm² were applied to the target at a pulse frequency of 10 Hz. A base pressure of $5 \times 10^{-5}$ Pascals and a substrate temperature of 350 °C were used. A target-to-substrate distance of

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**Fig. 2. Schematic representation of pixel-to-point (In, Ga)N LED transfer process.**

**Fig. 3.** The emission spectra GaN LEDs before (a) and after (b) laser lift-off and transfer.

**Fig. 4.** Schematic of the KrF excimer laser ($\lambda = 248$ nm) PLD system (Courtesy: Eric J. Carleton).

**Fig. 5.** (a) Fabrication process of disposable microfluidics and (b) SEM image of a fabricated channel.
6.2 cm was used in order to obtain a smooth film of reasonable thickness uniformity. A 2.4 μm film exhibited about 500 Å rms surface roughness. The deposition rate was about 0.5 Å/pulse under these process conditions. Faster deposition rates could be obtained by moving the substrate closer to the target, but at the expense of greater thickness non-uniformity. A schematic representation of the PLD system is shown in Fig. 4. Wire bonding or metallization to form contacts for the LED and the photodiode complete the process of the non-disposable substrate.

Disposable microchannels were fabricated on a PDMS mold, as is commonly performed by various groups [2,3]. The molds were prepared using deep-reactive-ionetching (DRIE) as shown in Fig. 5. Various mask designs have been attempted in order to maximize the area from which the dye’s signal can be collected. Microchannel depths vary from 20 to 150 μm, and channel widths of 50 μm, 100 μm, 1 mm, and 2 mm were evaluated.

3. Results and discussion

One difficulty associated with DBR filters is angular dependence. Fig. 6 displays simulations that characterize the angular dependence of the CdS and a ZnS:NaAlF DBR filter (Omega Optical) [10]. While the cutoff wavelength of the CdS film is almost independent of incident angle and thickness, the ZnS:NaAlF shows both an angular and thickness dependence. Angular dependence is not desirable, particularly if one is collecting signal from a wide range of incident angles, as is usually the case. Another drawback to DBR filters is the requirement for severe process control. In Fig. 6d, the effect of slightly varying the film thickness of just one of the compounds by a few percent is shown. Note that the cut-off wavelength of the CdS filter is independent of thickness variation.

Fig. 7 compares the performance of the CdS thin-film filter and a commercial ZnS/NaAlF DBR filter. Both the 2.4 μm CdS film and the DBR transmit only 1 × 10−6 of the blue incident excitation light. However, the DBR transmits about 80% of the green emission light whereas the 2.4 μm CdS film transmits only about 40%. Nevertheless, considering the simplicity and greater tolerance to process variations associated with CdS film deposition, it appears that a single thin CdS film is a viable alternative to a DBR.

Using the fabrication process described above, a 1.2 μm CdS filter was incorporated into a prototype device. Fluorescence detection data obtained with this prototype is shown in Fig. 8. Carboxylate-modified microspheres from Molecular Probes, 0.04 μm in size, were excited from nominally 2 mm wide, 100 μm deep, PDMS microfluidic channels by a blue LED at maximum intensity. Factors which limit the detectable concentration to 1 × 10−6 M include measurement imprecision associated with background noise, channel positioning inconsistency (relative to the LED and detector), uncertainty in the concentration, channel dimensional deviations, dye–dye quenching, collisional quenching, and disparity in the actual amount of dye under excitation due to sample preparation inequalities. Three measurements were recorded at each concentration. Each error bar represents two standard deviations (2–20 μV) both above and below the average. Due to photobleaching, the last of the three fluorescence signal measurements generally was lower. Fig. 9 shows images of the prototype microsystem with the LED power on (a), and with the microchannel being excited (b).

Optical filtering with II–VI compounds such as CdS can be extended to other wavelengths by adding a third element (e.g., Se) to vary the bandgap. CdS and CdSe form a continuous range of solid solutions via substitution, with little bandgap bowing [11]. This means that CdS1−xSex thin-film filters with cutoff wavelengths varying between 513 and 714 nm (the absorption edges of CdS and CdSe, respectively) can be fabricated using PLD by adjusting the target composition, which also takes advantage of the fact that PLD is known for providing excellent stoichiometric control [12]. These filters would be appropriate for numerous dyes. Analyses of PLD-deposited films of CdS1−xSex have revealed relatively steep absorption edges across the composition range [13].

Laser lift-off and transfer introduce the possibility of multicolor LEDs on the same substrate. As demonstrated in this work, pixel-to-point transfer allows individual LEDs to be lift-off a source wafer and placed at specific locations on a photodetector substrate. It is an obvious extension of this work that the LEDs need not be from the same growth substrate, thereby enabling integration of In, GaN LEDs with electroluminescence peaks ranging from the UV through the green portion of the visible spectrum. Likewise, filters deposited onto individual photodiodes need not be of the same composition, and hence could filter different wavelengths. This type of flexibility could also be explored at the wafer level, as large-area lift-off and transfer has been demonstrated [14], although adaptation of “pixel-to-point” transfer to a manufacturing scale would require considerable process development.

Although the prototype device illustrates the potential of this microsystem concept, there remains much room for performance improvement even at the single channel level. For example, it was observed that the dark current of the commercial photodetector increased from ~150 pA to ~15 nA during the course of the fabrication. Pinholes in the initial PECVD SiO2 layer are a potential contributor. If this undesirable increase in dark current could be avoided, an improvement in detection sensitivity could be expected.

As noted above, the design of the current prototype is such that light from the blue excitation source is not directly incident on the photodetector. This suggests the possibility that the filter itself may be superfluous. Clearly, having no filter at all would result in an increase in sensitivity (since currently, a 2.4 μm CdS filter only transmits 40% of the emission signal). Unfortunately, initial observations indicate that an appreciable amount of blue light is reflected from the
Fig. 6. Transmission as a function of both wavelength and incident angle for (a) a 2.4 μm CdS filter and (b) a 90-layer ZnS/NaAlF DBR stack. (c) The band shift as a function of incident angle is shown for three different DBR stacks. (d) The cut-off wavelength (1% transmission) is shown as a function of percent thickness variation.
backside of the microfluidic device, or otherwise scattered, in the direction of the photodetector. While the filter may be necessary, another way to improve detection sensitivity would be to add an anti-reflection coating to the CdS film to suppress the reflection of the green emission signal, which is estimated to be ~15% [15].

One weakness with the present configuration is that fluorescence signal exiting the backside of the transparent PDMS is not collected by the photodetector. Similarly, the solid angle captured by the detector is limited by the fact that the active area is 4 mm in diameter whereas the fluidic channel is approximately 2 mm away from the detector. A larger detector active area, in addition to a reflective surface at the backside of the PDMS, would significantly increase the amount of emission signal collected. The chief difficulty with the latter is that the green component of the LED’s electroluminescence spectrum would also reach the detector via reflection. This approach should therefore only be utilized for an LED with a minimal green component. In this paper, approximately 2.5% of the LED’s electroluminescence spectra was above the CdS filter’s cut-off wavelength. Improvements to the microsystem design can be quantitatively evaluated, to some degree, by their contribution toward lowering the detection limit.

4. Conclusions

Heterogeneous integration of a CdS filter, an (In, Ga)N blue LED, a Si PIN photodetector, and disposable PDMS microfluidic channels has been described and demonstrated for the development of fluorescence detection microsystems. The prototype microsystem features normal incidence excitation, but reduces the green excitation signal by placing the photodetectors in the same plane as the excitation source. A single 2.4 μm film of CdS deposited by pulsed-laser deposition transmitted only $1 \times 10^{-6}$ of the blue incident excitation, which was similar to the performance of a commercial multilayer DBR stack. However, the DBR transmits about 80% of the green emission light whereas the CdS transmits only about 40%. Nevertheless, considering the simplicity of the CdS film deposition, and its relative insensitivity to thickness and incident angle variations, these results were promising. An integrated microsystem employing a 1.2 μm CdS film was able to detect fluorescein concentrations as low as $1.2 \times 10^{-7}$ M. It is anticipated that the transmission of the green emission signal would be higher for the CdS film if an appropriate anti-reflective coating is added to suppress reflection, and thus an even lower detection limit should be realizable.

The CdS$_{x}$Se$_{1-x}$ materials system has the potential of enabling thin-film filters over a broad wavelength range. Heterogeneous integration of CdS$_{x}$Se$_{1-x}$ filters with (In, Ga)N
LEDs and disposable microfluidics has the prospect of enabling the assembly of multiwavelength chemical detection systems-on-a-chip.

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References

[10] Simulations were made with IMD software from Columbia University (http://idus.phys.columbia.edu/~wind/idm/).