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Nanowire LEDs and Applications: Device Structure, Fabrication and Characteristics

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Abstract: *Recent years have witnessed significant progress in the fabrication technologies and the performance of the flexible nanowire light-emitting diodes (LEDs). Organic material has traditionally been the best candidate in flexible LED fabrication, although there is increasing interest in flexible inorganic LEDs, new methods based on micro-structuring and transferring stan- dard thin films to bendable substrates are now underway. One of the developments that have made these developments promising is the emergence of flexible nanowire-based LEDs which have been found to have high efficiency and have mechanical durability. Two large fabrication approaches, in which nanowires are grown directly on flexible surfaces and in which nanowire membranes made of rigid wafers are formed and transferred, have been used. The transfer-based approach, specifically, does not degrade qual- ity of crystals and makes high-performance equipment possible. Based on these methods, scientists have already demonstrated blue, green, white and bi-color flexible LEDs with transferred nanowire membranes demonstrating particularly high optical performance, flexibility, and reliability.*

I. INTRODUCTION

Flexible optoelectronic components provide new features and will be a long-term drive of next-generation technology, with flexible light-emitting diodes (LEDs) having received specific focus because of their potential applications in rol- lable displays, wearable electronics, deformable lighting, and biomedical systems (soft lighting platforms). Most of the current flexible LEDs are based on organic materials deposited on lightweight plastic platforms, and organic LEDs (OLEDs) occupy this market due to their high level of flexibility, ability to be made on a variety of substrates, low processing cost, and the ability to be produced on a larger scale. Nevertheless, along with such benefits, organic devices are characterized by serious stability problems, such as oxidation, recrystallization, and degradation due to temperature, which adversely affect electrical conductivity and interfacial properties of the active layers. Such limitations are especially pronounced in the blue spectral range, where blue OLEDs have low luminance (-10 2 -10 4 cd/m 2), low external quantum efficiency (23 -100%), and short operation lifetime (usually about 3700 h at T 0 at 1000 cd/m 2), causing color imbalance and differential aging around the display. Conversely, blue emission has the highest performance in luminance and external quantum efficiency using nitride-based inorganic semiconductors like InGaN/GaN, which may provide higher luminance levels than 10 6 cd/m 2, EQE of over 80 percent and lifetimes of over 100,000 hours.

Such high-quality properties have prompted vigorous research activities to develop flexible inorganic LEDs, that seek to integrate the strength and efficiency of inorganic constituents with the mechanical versatility needed of flexible electronic devices of the next generation.

II. FLEXIBLE LEDS BASED ON TWO-DIMENSIONAL FILMS

Traditional inorganic LEDs generally are constructed in the form of a pn junction, comprising of a quantum well between the layers of the knee with high brightness and high operation life. Nevertheless, these devices are typically grown on a hard substrate and thus their application in deformable electronics and biomedical devices suffers mechanically and geometric limitations. To overcome this difficulty, flexible LEDs prepared with the application of inorganic materials (thin-film and micro-pyramid) of the thin film have been developed through micro-transfer printing, which is a universal method that allows incorporation of structured materials at microscales (down to nanoscale) and centimeter scales onto nearly any substrate. This is performed by loading patterned layers of devices onto the source wafer to be transferred onto an elastic rubber stamp and transferred to a flexible host substrate, but laser lift-off or epitaxial lift-off techniques are used to release the active layers. In other systems, a layer of quasi-2D in between the interface (like graphene or hexagonal boron nitride) is added so that it becomes possible to mechanically delaminate the atomic bond by using weak van der Waals forces. Kim et al. were the first to demon- strate the flexible IIIV LEDs, printing AlInGaP quantum- well microstructures on ultrathin plastic and rubber, where a process relying on epitaxial growth, lateral patterning, lift-off, transfer, electrical interconnect fabrication, and final device integration was used. These systems have been integrated into waterproof light emitting sutures, implantable optoelectronic sheets, plasmonic crystal illuminators and optical proximity sensors, among others, which are of biomedical interest. In addition to red and infrared

emitters of the AlInGaP material, blue InGaN-based micro-LEDs have further increased the capability of flexible platforms of solid-state lighting and implantable medical systems, with an upcoming generation of injectable, cellular-scale optoelectronics providing the ability to perform fully wireless, programmable behavior control in free-moving animals. Such stacked multifunctional systems incorporate micro-LEDs, photo detectors, thermal sensors and electrophysiological probes and the performance of such micro-LEDs is capable of state-of-art application of GaN LEDs and can provide optical power required to perform optogenetic stimulation, without such bulky lasers or fiber- couples. Regardless of these developments, development of fabrication of flexible devices based on standard thin films has been difficult due to the numerous processes of micro- structuring combined with the tendency to undergo a strain due to bending as it changes the emission wavelength. To minimize these and to gain higher mechanical flexibility, the size of the active elements is better shrunk and bottom-up nanostructures shown advocated. Flexible LEDs realized by transferred micro-array pyramids of nanoparticles based on nano-void creation of the heterointerface have been realized via this strategy, and the future development of even smaller components remains practical using bottom-up nanowires in future applications of flexible optoelectronic device development.

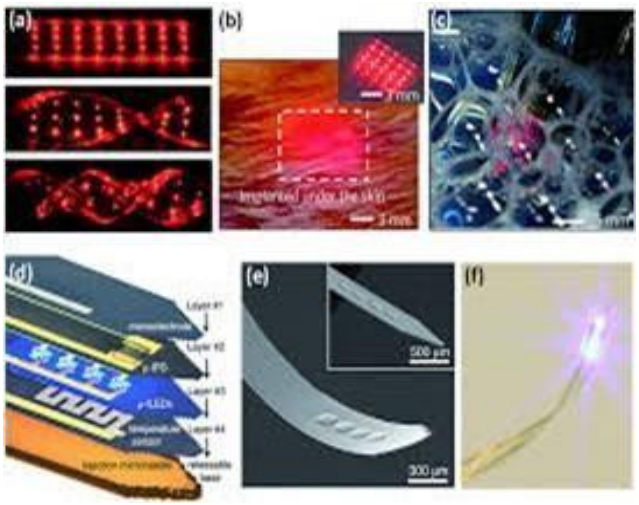

Fig. 1. shows different flexible and bio-integrated micro-LED systems. (a) Micro-LED arrays on a PDMS substrate twisted at various angles. (b) A light- emitting suture on a 700 µm thread in an animal model. (c) A stretchable LED–photodetector proximity sensor on a glove fingertip after immersion in soapy water. (d) An ultrathin injectable optoelectronic device with multiple components. (e) SEM image of the 8.5 µm-thick injectable LED array. (f) The integrated system emitting blue light. Panels (a–c) are adapted from Ref. 28 and (d–f) from Ref. 34.

III. FLEXIBLE LEDS BASED ON INORGANIC BOTTOM-UP NANOWIRES

The anisotropic geometry and high surface-to-volume ratio and extraordinary crystalline quality of semiconductor nanowires render them superior performance mechanically and optoelectrically. Their quasi-one dimensional structure makes it possible to resist very large elastic strains - often in the order of few per cent - without plastic deformation occurring. This makes nanowires excellent candidates for next-generation flexible, wearable and bendable opto-electronic devices, where it is easy to fracture conventional planar structures.

In addition, nanowires have free sidewalls which allow them to relax lattice mismatch strain effectively during epitaxial growth. This removes the stringent lattice matching require- ments that suffer the traditional thin film LEDs, and alleviates formation of threading dislocations. As a consequence, the structure of nanowires are in a defect-free or low-defect heterostructure and so the internal quantum efficiency of the devices becomes greater and the device lifetime is longer.

From the optical point of view, nanowires are natural optical waveguides. Their high refractive-index-contrast thus helps confine and guide photons along the nanowire axis which greatly improves the light extraction efficiency. Lessening the total internal reflection reduces or avoids the issues of light trapping that are common to planar LEDs, and produces much brighter, up to 30 times brighter, and more directional emission.

The bottom-up growth techniques of nanowires used for example MOCVD, MBE or VLS enable to precisely control diameter, length, doping concentration, and material composi- tion. This allows to fabricate sophisticated axial and radial heterostructures. In radial (core-shell) nanowire LED, the active region is wrapped around the core, and this results in a larger emission area with lowered current density, and reduces droop of efficiency at high injection levels.

Moreover, the nanowires offer a wide bandgap tunability, which enables the development of LEDs spanning exactly the entire visible range all the way into the ultraviolet portion of the electromagnetic spectrum. This tunability is derived from the nano-precise mankind-manipulation of the alloy makeup and parameters of the quantum-wells. Combined with their high thermal stability and high radiative recombination rates, nanowires are on the way to high-brightness, high-efficiency and long-lived inorganic LEDs for use in mechanically flexible (on polymer or metal-foil substrates) integrated circuits.

Overall, nanowire-based LEDs are a promising platform for future optoelectronics as a unique combination of mechanical robustness, good optical performance, compositional control, and flexible and large-area fabrication technologies.

IV. DIRECT NANOWIRE GROWTH ON FLEXIBLE SUBSTRATES

A simple method to produce flexible nanowire LEDs is simply to grow nanowires directly on flexible substrates and then redeposition them into a polymer matrix to make them mechanically stable. This technique is especially effective with substances

which are easily synthesized at relatively low temperatures. As an example, ZnO nanowires that can be fabricated with lower temperatures (less than 200 0 C) can be used on plastic substrates, including PET or PEN. Under this method, scientists have already made visible-emission LEDs, flexible pressure-mapping sensors, and bendable white-light sources all of which have been facilitated by the mechanical compliance and high optical gain of the ZnO nanowires.

Nevertheless, this method is restricted by the fact that most polymer and plastic substrates are unable to sustain the elevated temperatures necessary to use material such as GaN, InGaN or AlGaN, which tend to require growth temperature in excess of 700-900 o C. This leaves solution-based nanowire growth, which is based on low-temperature chemical path- ways, only capable of applying to a narrow set of materials. This limits the optical performance due to which high-quality III-V nanowires would typically need to be prepared by high- temperature epitoxy to achieve the best possible crystalline quality and emission efficiency.

In order to counter these limitations, scientists have con- sidered the application of flexible metal foils like titanium, molybdenum and stainless steel as other substrates. High temperatures and good thermal conductivity can be offered by these materials. High quality GaN and AlGaN nanowires have also been grown directly onto metal foils using molecular beam epitropy (MBE); this has resulted in fully flexible inorganic nanowire LEDs with reproducible emission even after repeated bending cycles. This is a significant advance in the high-performance flexible X-ray optoelectronics of III- nitride materials.

There is also the emerging technology of transfer printing and nanowire harvesting and subsequent re-assembly, which is becoming of interest. In these processes, nanowires are initially grown on a hard and high temperature compatible substrate and then are released either mechanically or chemically and then transferred onto flexible materials. This circumvents temperature constraints and maintains high crystalline quality, to provide a scaleable solution to the production of high- efficiency flexible nanowire based LED arrays.

In general, low-temperature growth on plastics, high- temperature growth on metal foils and advanced methods of transfers are converging to form a flexible toolkit to accom- plish mechanically flexible nanowire-based LED technologies.

In-plane transferred nanowire LEDs

There is one additional technique that can be used ef- fectively to produce flexible nanowire LEDs, other than di- rect epitaxial growth, and this is the transfer of pre-grown nanowires onto flexible substrates. In this approach, high- quality nanowires can be grown on hard and high-temperature substrates and later incorporated into the flexible device archi- tectures. In this technique, scientists have developed in-plane and vertical nanowire LED designs.

Nanowires in in-plane structures are laid down in the horizontal direction on the substrate surface. One such popular method is roller-pressing or contact printing, in which n-type ZnO nanowires developed in large mass proceeds to transfer the nanoparticles onto elastic surfaces patinaed with p-type organic semiconductors. This creates easy hybrid LED which is efficient, cheap and very flexible to large-scale roll-to- roll. The good mechanical compatibility also provides such hybrid devices with stretchable and wearable lighting elements because organic materials and flexible polymers have.

The latest developments in the process of alignment of nanowires have additionally enlarged the range of possibilities concerning the bottom-up construction of devices. Electric and magnetic field-assisted assembly, fluid flow driven by shear, dielectrophoresis and capillary-force assisted alignment are now controlled methods of alignment. Those techniques serve to assemble nanowires into regular pattern to enhance carrier transport, uniformity and manufacturing of large-scale nano- wire-based LEDs.

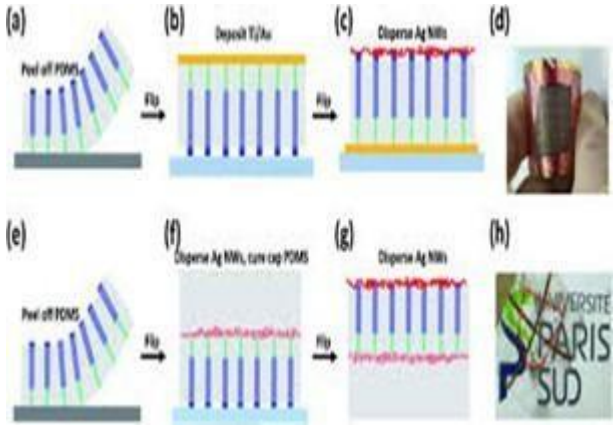

Fig. 2. shows the fabrication process of flexible LEDs based on vertical nitride nanowire arrays. Panels (a–c) outline the steps for making semitransparent LEDs using Ti/Au as the back contact and Ag nanowires as the top contact, with panel (d) showing the resulting device. Panels (e–g) illustrate the fabrication of fully transparent LEDs that use Ag nanowires for both contacts, and panel (h) presents a photograph of the transparent LED. Adapted with permission from the referenced source.

An interesting example of this idea was introduced by Lieber et al. who produced an in-plane ultraviolet LED in crossed n-GaN/p-Si nanowires deposited on a plastic substrate through fluid-directed assembly using fluid-directed align- ment of nanowires on the substrate. The resulting device was capable of having steady emission intensity following repeated bending, which demonstrated mechanical strength of nanowire-based flexible LEDs. This paper has pointed out the possibility of using hybrid nanowire heterojunctions as high performance bendable optoelectronic devices.

Nevertheless, in spite of these good developments, there are still a number of challenges. Making in-plane nanowire LEDs requires very precise spatial orientation of single nanowires or in aligned arrays and this can be challenging and time- consuming

in large-scale applications. Also, the further en- gineering is complicated by the possibility of incorporation of transferred nanowires with multilayer stacks, complex device architectures, and non-flat substrates. The mentioned constraints are the main factors that restrict the general ap- plication of in-plane nanowire LEDs in commercial flexible optoelectronic systems.

However, the current advancements in nanoscale assembly technology, printing technology and control of the alignment process is gradually enhancing the scalability and reliability of nanowire transfer technology, which is a strong contender in the future of flexible LED technology.

V. FABRICATION

One of the most popular approaches to the production of flexible nanowire LEDs is a multi-step procedure that is focused on the integration of vertically aligned nanowire arrays into a polydimethylsiloxane (PDMS) substrate. The first step involves the growth of high quality vertical nanowires on a firm substrate followed by spin-coating or drop-casting a layer of PDMS which completely encases the nanowires. When the polymer has cured, plasma etching is applied whereby it is selectively used to etch away a thin section of the PDMS to reveal the nanowire tips. The given etching step is also important, because it provides both efficient electrical contact and mechanical stability of the nanowire-polymer composite.

The PDMS film is then etched before being peeled off and it forms a free-standing flexible membrane which maintains the exact vertical orientation of the nanowires. This is a free- standing nanowire-PDMS structure on which two different devices are based: semitransparent LEDs and fully transparent LEDs.

In the case of semitransparent flexible LEDs, it is the mem- brane that is inverted, with the exposed ends of the nanowire sticking out vertically. Patterned bottom metal contacts are next evaporation or sputtered on the backside. The structure is bonded to flexible materials to support it mechanically including PET, PEN, and metal foils. An Ag nanowire network is then deposited as the top transparent electrode, that is highly conductive, and has high optical transmittance.

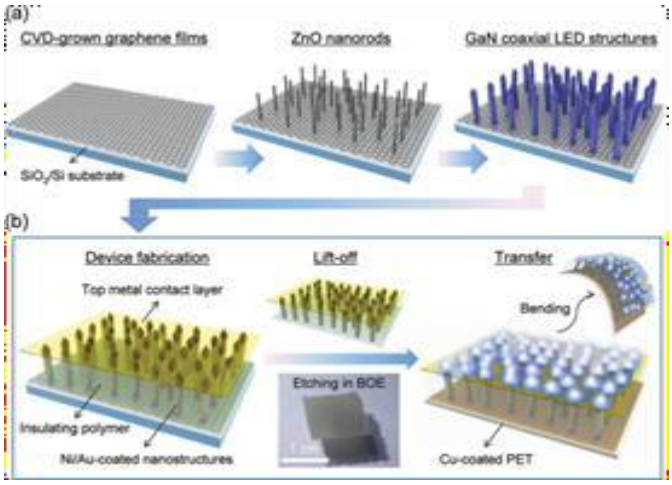

Fig. 3. Schematic illustration of the process used to fabricate flexible LEDs using GaN/ZnO coaxial nanorod heterostructures grown on graphene films. Adapted with permission from ref. 61.

Ag-nanowire networks can be the top and bottom elec- trode in fully transparent nanowire LEDs. This two-pronged electrode structure is the most optimal to enable optical transparency in that the devices are almost invisible except the few metal connection points. A second layer of PDMS is then applied to strengthen the mechanical properties and improve the electrical insulation, fully entrap the nanowire LED structure, and create a powerful, flexible, and transparent light-emitting panel.

The functionality of PDMS based processing also makes it easy to add new functionality. Through adjusting the poly- mer backbone, overprinting new layers or adding wavelength converting materials it is possible to adjust the properties of a device to meet the specialized requirements. An interesting case is the incorporation of the YAG:Ce phosphor particles into the PDMS that made possible the production of the flexible white nanowire LEDs. This design converts the blue light of InGaN nanowires to yellow light using the phosphor, and a combination of blue and yellow light is produced to provide a wide band of white light.

Also, this strategy creates opportunities to explore innova- tive concepts of devices like multicolor emission, stretchable lighting, curved transparent displays and biocompatible opto- electronics since PDMS is soft and transparent and can be used in contact with the skin. The applications of flexible LED can be extended to a broad range of applications by manipulation of phosphor composition, nanowire materials, or polymer stiffness.

VI. BLUE AND GREEN FLEXIBLE NANOWIRE LEDS AND CHARACTERIZATION

There are several studies that have shown the use of light emitting devices that were designed with polymer embedded nanowires whereby the active nanowires are initially placed in a flexible polymer film, and the nanowires are then lifted off of their substrate. Majority of them are based on the nitride materials with the polymer merely acting as mechanical support but the p n junction being all in the nanowires. The initial ones were GaN p n microrods and p-GaN n ZnO coaxial nanorod heterostructures grown on graphene films which also served as back contacts. The nanorods that have grown on graphene-coated Si/SiO 2 are embedded in an insulating polymer and released by wet etching the underlying substrate, and the whole membrane is transferred to a metal- coated flexible substrate as shown in Fig. 4. A semi-transparent top electrode of Ni/Au was employed in

these designs, but its optical transmittance is low and therefore does not allow the efficient extraction of light.

Ag-nanowire transparent contacts were used to analyze the optical, electrical as well as mechanical characteristics of the flexible nanowire LEDs. The films of Ag nanowires had a transmittance above 80 percent in the visible regime after thermal annealing. The complete transmission of the LED was a little less at short wavelength because of absorption in the InGaN/GaN nanowires but at the same time, it remained more than 60 percent transmitted at the green wavelength. The I V characteristics were obvious diode characteristics with a turn- on voltage of approximately 3 V, at which electroluminescence also started characteristics. EL spectra of a voltage-dependent EL at low bias showed a broad peak of the axial quantum wells at 447 nm whereas at higher injection a secondary peak at 415 nm of the radial quantum wells emerged. During mechanical tests, the blue emission was stable during flat, outward and inward bending, as far as 2.5 mm radius and there was no degradation in bending as well as after several bending cycles or storage in ambient conditions. These findings prove that flexible nanowire LEDs have high transparency, electrical sta- bility, mechanical robustness and long-term stability without encapsulation, and are even better than the usual OLED-based flexible devices.

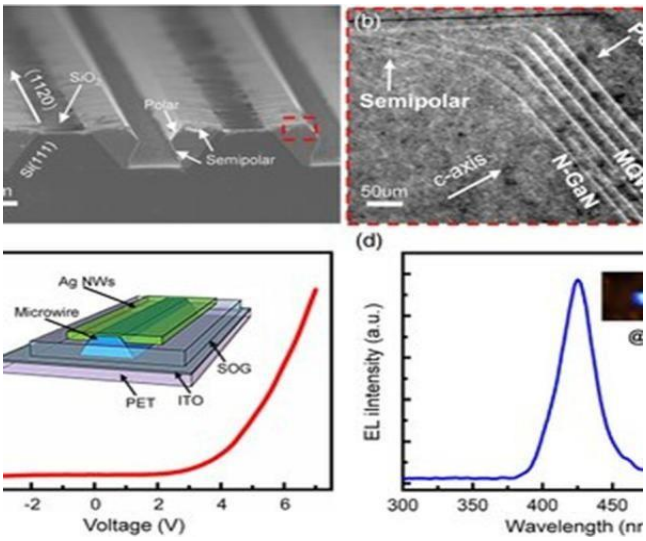

Fig. 4. shows the mechanical and electrical performance of a flexible LED based on vertical nanowire arrays. Panels (a–c) display the device emitting blue light under different bending radii: flat (∞), 3.5 mm, and –2.5 mm. Panel (d) presents the I–V characteristics normalized to the number of contacted nanowires and to the device area, with the inset showing room-temperature EL spectra measured at applied biases from 4 to 8 V. Adapted with permission from Ref. 58.

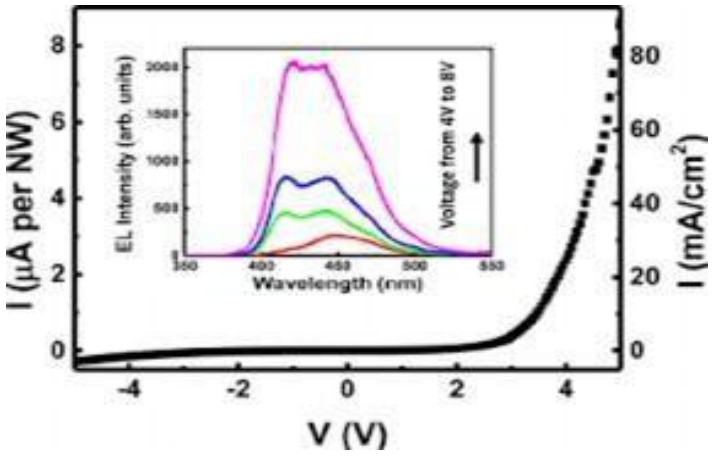

Fig. 5. The SI–V characteristic of the flexible LED is normalized to the number of contacted nanowires (left scale) and to the device surface (right scale). The inset presents the room-temperature EL spectra at various applied biases from 4 to 8 V.

VII. CONCLUSIONS AND PERSPECTIVES

Finally, addressing the weaknesses of OLEDs, namely insta- bility, low efficiency and short lifetime has motivated a robust interest in inorganic flexible LEDs, such as micro-transfer printing of thin-film and micro-pyramid devices. The bottom- up nanowire LEDs is a more elegant alternative that integrates high performance of III V based semiconductors and the mechanical flexibility of nanowires and polymer substrates. A variety of device structures have been investigated, and the ver- tically transferred nanowire structure is the one which is most promising. Through this method, successful demonstrations of blue, green, white and bi-color flexible LEDs have been made and all of them show excellent flexibility, reliability, and long-term stability. The versatility of freestanding nanowire- polymer membranes is that diverse materials may be incorpo- rated, which opens new possibilities of high-efficiency flexi- ble displays and other nanowire versions of flexible devices including photodetectors, solar cells, piezoelectric generators, and so forth

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