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Semiconductor nanowires, what's next II?

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Abstract:

Historically, nanoscience research has been centered around three classes of nanoscale building blocks based on their dimensionalities: C_{60} & quantum dot (0-dimensional, 0D), nanotube and nanowire (1D), graphene and 2D materials (2D). After almost three decades of active research, semiconductor nanowires have been established as one of the key building blocks in nanoscience and technology, with vibrant subfields including nanoelectronics, nanophotonics, thermoelectrics, energy conversion and storage. Here I present some of my personal reflections and perspective in this very active research field with a particular focus on the future development of nanowire nanolasers and nanowire photochemical diodes.

Ten years ago I wrote an article in *Nano Letters* entitled" Semiconductor nanowires, what's next?".[1] 10 years later, I am asking the same questions again, so this is a sequel to my early article, with some reflections and new perspectives.

The last decade has seen two very exciting research frontiers in the semiconductor nanostructure domain, one is halide perovskites. With their promising applications in photovoltaics and light emitting devices, quantum dots and nanowires of halide perovskites have been investigated with extensive efforts, leading to another important sub-class of semiconductor nanowires with unique bonding characters and structural dynamics. The other is the emergence of 2D semiconductors beyond graphene. 2D semiconductor, with its own vast collection of different compositions and electronic structures, becomes a vibrant research frontier just like other building blocks of nanoscience.

After several decades of intensive fundamental research, the last decade started to see some successful commercialization of nanowire-based technology in several important industrial sectors, from thermoelectric waste heat recovery for heat engines to silicon-based anodes for battery energy storage. Broadly defined, another class of inorganic nanowires, metal nanowires such as Ag and Cu, are becoming viable materials as transparent electrodes for the display industry. One would also have to mention that the 2 nm silicon transistor node is now becoming a reality with the significant advancement in the semiconductor industry, albeit

through the highly-integrated top-down nanofabrication process. One should not forget that one of the major inspirations for nanowire research in the early years is to produce highly-integrated computing devices based on synthetic nanowires, however, this did not materialize due to a lack of cost-effective large-scale integration schemes based on these synthetic nanowires.

Back in 2019, I edited a special issue for *Chemical Reviews* on semiconductor nanowires to commemorate 25 years of original nanowire research.[2] This particular collection summarized in more than 400 pages some of the major progress in the subfield of nanowire electronics, photonics, energy storage, photoelectrochemistry, and thermoelectrics. Due to the sheer magnitude of the research scope breath, I will not be able to do justice to cover every aspect of this exciting research field here in this short perspective. Instead, I would like to focus on two research subfields that were initiated here at Berkeley and offer some of my personal opinions on the remaining challenges, scientific questions, and technical bottlenecks.

Nanolasers and Beyond

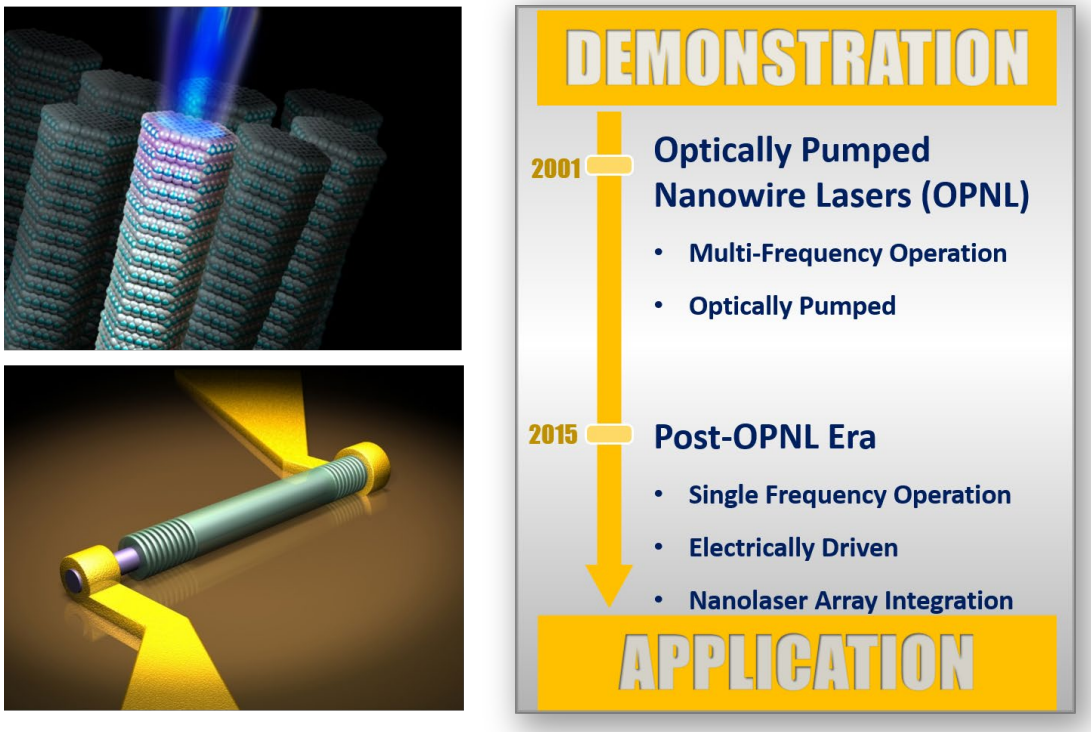

Figure 1. Frome optically pumped nanowire laser to electrically driven nanowire laser.

Research in miniaturized lasers and their applications is paramount for the continued growth of photonic technologies because they can enable new applications and discoveries unattainable by traditional photonics. Of the available

systems, semiconductor nanowires have been identified as potential candidates for nanoscale lasers. The concept offered the possibility to reduce the footprint of optoelectronic devices, and so nanowires have been actively explored to realize compact lasers.

We demonstrated the first semiconductor nanowire laser using ZnO nanowires,[3] which introduced a breakthrough approach to the fabrication of nanoscale lasers. A semiconductor nanowire can fulfill two fundamental requirements for traditional lasers: they act as both the optical gain medium and the optical cavity (Figure 1). These high-quality single-crystalline structures were grown bottom-up without any extended defects that would otherwise lower the luminescent efficiency, a critical parameter for reaching the laser threshold. Most interestingly, we utilized the natural anisotropy of the crystal structure of ZnO to aid in defining the optical cavity. By building the cavity up atom by atom, the crystal planes were used to define the side facets of the waveguide and the mirrors of the resonator. These planes are optically smooth and exhibit low propagation loss. At the time, it was unclear if mirror facets could be so small because they could induce enough scattering loss that the optical cavity would be too inefficient for laser oscillation. In this case, these nanowire lasers shattered expectations by lasing at room temperature.

Since its first demonstration, our approach to designing nanoscale lasers has been adopted by numerous research groups and these nanolasers have been studied extensively.[4,5] It is now understood that these well-faceted nanowires with cross-sectional dimensions on the order of the wavelength of light support Fabry-Perót cavity modes. This understanding has allowed the exploration of numerous new materials, new applications, and nanoscale optical phenomena. The library of nanowire laser materials has expanded enormously, resulting in lasers that emit light in the UV, visible, and near-IR. Optically pumped laser oscillation has been demonstrated for ZnO, GaN, InGaN, CdS, CdSe, GaAs, InGaAs, AlGaAs, ZnS, GaSb, and a collection of halide perovskites with different compositions.[4,5] The compact form-factor of one-dimensional nanoscale lasers has enabled the development of new photonic applications such as nanoscale optical routing, nanoscale electro-optic modulators, and single-cell probes. By introducing novel photonic complexities, researchers have modified the lasing modes of nanowires to achieve single-mode lasing and nanowire ring resonator lasers. Finally, the semiconductor nanowire has been used as a platform to study enhanced light-matter interactions that generate intense electric fields for applications such as ultrafast optical switching in semiconductor lasers. This field has enjoyed tremendous success in the past 20 years. In this particular direction, one powerful route for increasing the capability of semiconductor nanowire lasers is the discovery of new semiconductor materials, e.g. the emerging chalcogenide perovskite semiconductors. For instance, one area that will greatly benefit from the development of new materials is low-cost, compact photonics and the readiness for heterogeneous integration. Low-cost lasers offer the prospect of sensors and point-of-care diagnostics that are

disposable and produced on a large scale. Therefore, fundamental research in novel materials for nanoscale photonics will overcome the current limitations in laser technology and greatly expand their use.

The development of new-generation nanowire lasers with improved functionalities with real-world applications is critical for continuing the success of this field, and their realization can enable a new wave of innovation in nanoscale photonics. One of the principal benefits of semiconductor optoelectronics is their ability to be electrically injected efficiently, as evidenced by the 2014 Nobel Prize in Physics. In fact, its importance is so great in photonic applications that it is generally implied that semiconductor lasers are pumped by electrons. The ability to use metal electrodes instead of an optical pumping scheme truly allows the laser to be integrated into miniature devices. Thus far, there have been limited reports on electrically pumped nanowire lasers in the last ten years. These reports hint at the possibility of achieving stimulated emission. However, the limited correspondence between the theoretical laser design and the experimental results and the limited evidence of lasing oscillation leaves much room for innovation and improvement. With this in mind, it is paramount to develop strong theoretical and experimental design principles to unequivocally demonstrate electrically pumped nanowire lasers on a single-nanowire level.

An electrically pumped nanowire laser will enable the compact integration of nanoscale optical components. Given that the fabrication of silicon-based integrated circuits is beginning to reach its physical limits, large-scale integration of electrically driven nanolaser arrays will represent a significant step toward the realization of a nanoscale photonic integrated circuit that processes information at rates that are orders in magnitude faster than their electrical counterparts. The miniaturization of the laser can also introduce new applications such as nanoscale sensing for biomedical applications in real-world environments. Scientifically, however, it remains an outstanding challenge to deposit metal electrodes on a semiconductor nanowire because metals perturb the cavity mode and induce significant propagation loss.

Lasing from electrical injection in a single nanowire level device is sufficiently different from the bulk semiconductor devices, and requires unique and creative electrical injection configurations. One of the first electrically driven semiconductor nanowire laser was developed based on a CdS nanowire.[6] Later on, electrical injection in *p-n* junction core-shell nanowires and *p-i-n* longitudinal nanowires was investigated using numerical simulation.[7] The longitudinal structure requires higher bias and doping concentration, compare to the core-shell nanowires in electrical driven devices. And later electrically driven metallic cavity nanolasers, based on the structure with InP/InGaAs/InP nano-pillar and SiN layer overcoating, were experimentally demonstrated.[8] These metallic cavity nanolasers can be used as azimuathally polarized beam, driven by electrical injection. Such plasmonic-based

nanolasers, by themselves, belong to a new class of nanolasers due to their different light-matter interaction.

The core-shell structure is an optimum place to start for designing an electrically pumped nanowire laser. III-V semiconductors are well-established material systems to synthesize core-shell nanowires with precise control for light-emitting diodes (LEDs). Specifically, gallium nitride and gallium arsenide-based materials have been used to construct core-shell nanowire LEDs and potentially laser diodes.[4] There is already a hint of nanowire-based nanoLED potentially being integrated into high-definition display devices.

The principal challenge in realizing electrically pumped nanowire lasers is the separation of the metal electrodes from the optical cavity. Metals, although routinely employed as mirrors for macroscopic cavities, are undesirable when in contact with a waveguide because they strongly perturb the cavity mode. Therefore, configurations employed to mitigate this loss usually involve moving the electrodes far away. However, this strategy is in direct conflict with developing compact lasers because it promotes the fabrication of macroscopic devices. Nanoscale systems do not have the luxury to compensate for ohmic losses by adding more gain material.

One way to address the challenges of incorporating metal electrodes into nanowire cavities is by introducing Distributed Bragg Reflectors (DBR) into a semiconductor nanowire.[9] While DBR is routinely adopted in vertical-cavity surface-emitting lasers (VCSELs),[10] its integration on nanowire cavity remains non-trivial. A traditional nanowire laser cavity uses the end facets as mirrors. DBRs are an alternative approach where layers of alternating high and low index of refraction areas are introduced along the axis of propagation. They can be fabricated by layering different materials, or grooves can be defined into the material at regular periods (Figure 1). Because each indentation into the waveguide does not need to cut through the entire nanowire, it is possible to electrically contact the active medium from outside the optical cavity. This type of structure allows the isolation of the optical cavity from the metal electrodes. This approach could allow us to balance the modal gain with electrical transport, which will ensure the necessary control to realize electrically pumped nanowire lasers.

The design of an electrically pumped nanowire laser requires careful consideration of the materials and the device architecture. The device structure must incorporate an electrical diode into an optical cavity without severely degrading the performance of the cavity. The diode should facilitate charge injection along as much of the wire as possible in order to maximize the length of material used for gain, and the free carrier density should be radially uniform across the wire. This consideration raises a natural question of whether it is better to use a core-shell p-n junction or an axial p-n junction. Intuitively, an axial junction would promote recombination only along a short section of the wire. A core-shell structure, doped

correctly, would ensure that recombination would occur over a longer length of the wire, thus maximizing the use of the material gain. A core-shell structure also has the added benefits of allowing for improved material flexibility in the synthesis and for higher injection densities to reach population inversion. Core-shell n-$In_xGa_{1-x}N$/p-GaN, n-GaN/$In_xGa_{1-x}N$ multi-quantum-well/p-GaN, and n-GaN/p-GaN nanowires can now be readily synthesized using MOCVD.[4]

Assuming the electrically-driven nanolaser structures can be fully optimized in the next decade, further systematic investigation is required to fully maximize the device performances such as photon-electron conversion efficiency, lifetime, and reproducibility. And more importantly, additional optical components, e.g. optical coupler, splitter, polarizer, and modulator, should be considered as well as emitter, waveguide, and detector, for all photonic circuit integration.[5,11]

Nanowire Photochemical diodes

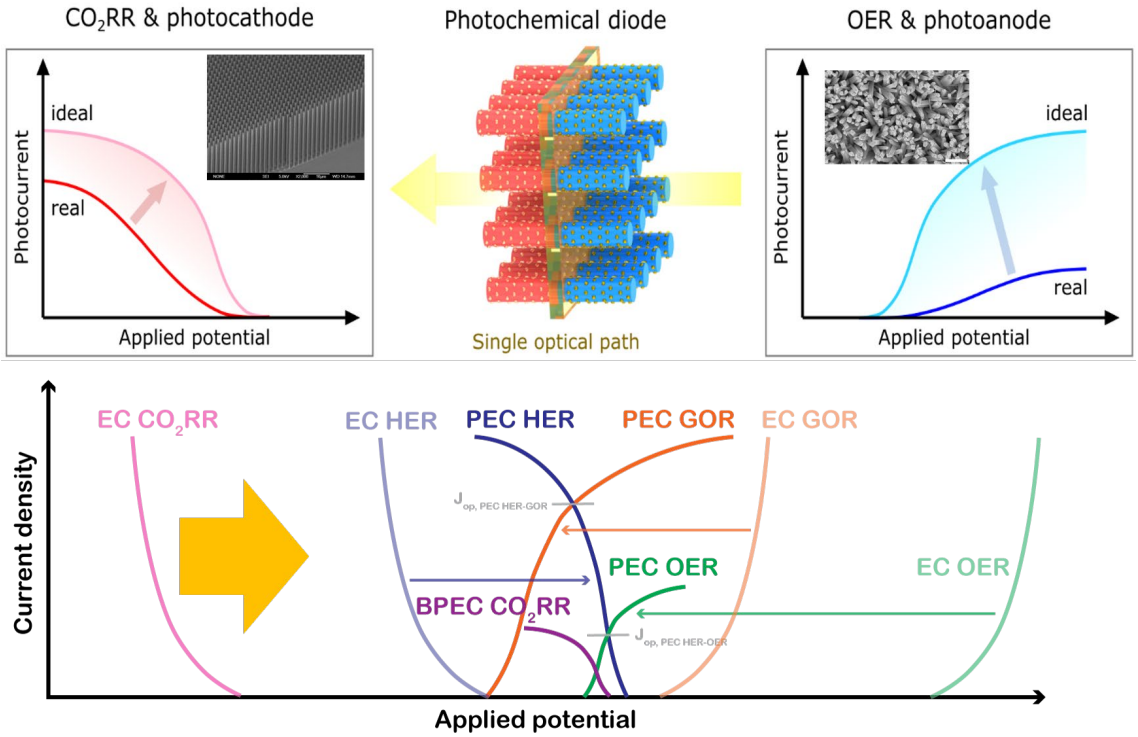

Figure 2. Nanowire photochemical diodes and its overall thermodynamic energy consideration from the photocathode and photoanode for the reduction/oxidation half-reactions. CO2RR: CO2 reduction reaction; OER: Oxygen evolution reaction; HER: hydrogen evolution reaction; GOR: Glycerol oxidation reaction; EC: Electrochemical; PEC: photoelectrochemical; BPEC: bio-photoelectrochemical.

Besides their tremendous potential in nanolasers and nanophotonics, semiconductor nanowires also represent a powerful platform for solar energy

conversion including photovoltaics and artificial photosynthesis due to their strong light absorption and high surface area. The direct conversion of sunlight into energy stored in chemical bonds, termed artificial photosynthesis, mimics the natural photosynthesis process occurring in plants. Artificial photosynthesis is an integrated system that combines multiple different processes, from light absorption, charge separation, to surface molecular activation and transformation (Figure 2).[12,13,14] The energy input requirement for such a process is fundamentally limited by the nature of the chemical reactions involved. Typically, the thermodynamic potential needed to drive the reaction of interest is greater than 1V in order to drive the both oxidation and reduction half-reactions, e.g. HER/OER and CO2RR/OER. For the practical application of solar fuels as a renewable energy resource, high solar-to-fuel conversion efficiency is desirable.

Photochemical diode, was originally proposed back in the late 70s in order to drive solar water splitting, i.e. OER and HER.[15] This photochemical diode concept basically captures the essence of what's happening in natural photosynthesis with two-photon activation. In the original design, two semiconductors were used, one is a p-type semiconductor serving as photocathode, the other side is a n-type semiconductor serving as photoanode. When these electrodes are in contact with the electrolyte, effectively two liquid junction solar cells are established. Each of these two liquid junction solar cells will generate their own photovoltage. The combined photovoltages will provide a sufficient driving force to carry out the water/CO2 reduction and water oxidation. One should note the original concept was based on bulk semiconductors, and naturally, this original design was not a single optical-path device.

In early 2000s, an ambitious effort called Helios Program was launched at Lawrence Berkeley National Laboratory.[16,17] Part of its mission is to develop a functional artificial photosynthetic system that can carry out solar water splitting or fixing CO2 using solar energy without external bias. It was at this time, we modified the original photochemical diodes by introducing semiconductor nanowires as the key photoelectrode components within the design (Figure 2).

Replacing the original bulk semiconductor with semiconductor nanowires has several major advantages and eliminated some of the major scientific bottlenecks in the original bulk design. These single-crystalline semiconductor nanowires typically have great minority carrier mobility, so that the photo-generated minority carriers can be effectively transferred to the nanowire/electrolyte interface over relatively short distance to participate in the oxidation/reduction half-reactions. Nanowire arrays also naturally have large surface area, and enable the stacking of the surface catalysts in the third dimension, and this effectively removed the solar photon flux mismatching issue in the original bulk design.[18] And lastly, these nanowire electrodes can be designed to be a single optical path device considering their own bandgap and absorption coefficient.

In 2013, using Si and TiO_2 nanowires as building blocks, a proof-of-concept solar-to-fuel conversion nanowire-based photochemical diode was demonstrated for direct solar water splitting.[19] As the photochemical diodes are essentially photocathode and photoanode linked in series, the output current from the two electrodes needs to match, otherwise the overall performance will be limited by the electrode producing a lower photocurrent. So far, photochemical diode performance is largely limited by the low current output from the photoanodes, typically large bandgap oxides. So it is not surprising that the first nanowire photochemical diode only delivered solar-to-fuel efficiency at 0.12%. Fundamentally, one can use semiconductors with better photocurrent output in order to increase the overall solar-to-hydrogen conversion efficiency. For example, by using InP and BiVO4 photoelectrode couple, it is expected that the overall efficiency can reach 3%. And more recently some efforts on running the water splitting at a higher temperature using nitride nanowires has already pushed the efficiency to 10%.[20]

This concept of nanowire-based photochemical diodes from the Helios Program was later propagated into the Energy Hub Joint Center of Artificial Photosynthesis, but with a clear focus on running the CO2 reduction on the photocathode. While the overall thermodynamic energy needs for CO2RR/OER and HER/OER are quite similar, the overpotential to drive CO2 reduction towards C2+ products is significantly larger than that for HER. This introduced another significant scientific challenge. Going back to the fundamental physics of the photochemical diode itself, the photovoltage outputs from the two electrodes are limited considering their bandgap as well as the overall solar spectrum. This photovoltage will need to cover the overall thermodynamic voltage needs for the two half-reactions (e.g. CO2RR/OER), the additional overpotential needs to drive these two separate half-reactions, and IR drops and ohmic loss if any (Figure 2).

Without invoking embedded solar junctions, there is simply not enough photovoltage from the two stand-alone semiconductor electrodes with 1 sun irradiation to drive CO2RR and OER under bias-free conditions, as many of the existing CO2RR catalysts (e.g. Cu) still require 700-900 meV overpotential. This is the very reason that so far there is still no experimental demonstration of the photochemical diode for CO2RR/OER using inorganic catalysts.

This led us to the introduction of bio-photochemical diodes back in the early 2010s.[21] The overpotential issue can be largely circumvented by using biological systems operating at very low overpotentials, close to the standard thermodynamic potentials. Taking advantage of an overpotential <200 mV of the bacterium *Sporomusa ovata*, a Si – TiO_2 nanowire biophotochemical diode was able to perform unassisted acetate production at a 0.38% solar-to-chemical efficiency and faradaic efficiency up to 90%. By feeding this CO2-derived acetate to a genetically engineered *Escherichia coli*., one can produce many other value-added chemicals such as *n*-butanol, polyhydroxybutyrate (PHB) polymer, and different isoprenoid natural products. Similarly, the acetate produced by *S. Ovata* can be passed onto the

another microorganism such as *C. basilensis* and *R. Pal* for the purpose of PHB production and N2 fixation at ambient conditions.[22,23] More recently, close packing of *Sporomusa ovata* bacteria onto conductive Si nanowire arrays resulted in a solar-to-chemical conversion efficiency of 3.6% for acetate production, when driving the process by an externally wired, multi-junction Si solar cell. For these biophotochemical diodes, the next step remains to be enhancing the CO2 turnover rates by using adaptive laboratory evolution techniques and/or genetic modification of reaction pathway. By increasing the turnover rates for these microorganisms, one can achieve better overall solar-to-chemical conversion efficiency under bias-free conditions. The other research frontier would be the genetic modification of reaction pathway in order to produce different chemical products, all from CO2 and sunlight.

With a modular design for photochemical diodes, these proof-of-concept devices leave much room for improvement and welcome performance enhancements by updating individual components with the latest improvements. Specifically, the balance between solar-generated electron flux and the turnover frequency of the electrocatalyst demands the development of better electrocatalysts, especially for the OER and CO_2 reduction. Ideally, an overptential of less than 200 mV at 10 mA/cm^2 is desired. This is however currently non-existent for CO2RR electrocatalysts. In this regard, any new development in discovering new CO2RR electrocatalysts with very low overpotential will advance the field of the nanowire photochemical diode significantly.

The requirements for high photocurrent and photovoltage output will rely heavily on photoanode research, which can be further supplemented by the improvement of photovoltage output in the photocathode. At this point, photoanodes that can produce large photocurrent and large photovoltage, and are photoeletrochemically stable should be one of the major research priorities. Theoretically, Photocurrent output should approach beyond 20 mA/cm^2. Assuming all of this photocurrent can be used for reaction, theoretically solar energy conversion efficiency of ~24% can be expected (Figure 2). Whether we will able to reach such ambitious goal, it will largely hinge upon new CO2 electrocatalyst and photoanode discoveries.

Alternatively, one can further expand the definition of photochemical diode by including other type of oxidation that is not that sluggish and requires much lower overpotential. For example, the thermodynamically demanding O_2 evolution ($E_{H2O/O2}$ = 1.23 V vs. RHE) can be replaced by organic oxidations, e.g. glycerol oxidation, which readily occur at lower applied potentials of 0.3-0.6 V vs. RHE (Figure 2). This induces a negative shift in the oxidation curve which improves the overlap operation current of the photocathode and photoanode, resulting in lower overall photovoltage requirement and higher operating photocurrents under no applied bias voltage. This expanded definition of photochemical diodes will no longer closely mimick the natural photosynthesis in green plants by excluding the OER half reaction, it is also interesting to point out that while many autotrophs (plants, algae,

and bacteria) obtain energy and nutrients through solar-powered photosynthesis (photoautotrophs); certain microorganisms can also obtain chemical energy through oxidation (chemoautotrophs) of inorganic/organic sources, i.e. using inorganic/organic substances as electron sources. To this extent, this expanded version of the photochemical diodes follows the basic energy conversion principles for some of these chemoautotrophs. It offers additional benefits of fixing CO_2 into C_{2+} compounds at high operating current density, while simultaneously upgrading biomass into other value-added chemicals, all operating at bias-free one-sun irradiation.

Acknowledgements

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