

Cutting-edge nano-LED technology

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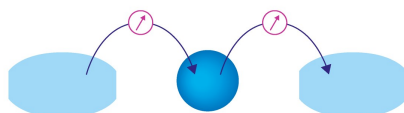
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ABSTRACT

In this Perspective, we will introduce possible future developments on group III-nitride nano-LEDs, which are based on current achievements in this rapidly arising research-technological field. First, the challenges facing their fabrication and their characteristics will be reported. These developments will be set in a broader context with primary applications in lighting, display technology, biology, and sensing. In the following, we will center on advanced applications in microscopy, lithography, communication, and optical computing. We will discuss unconventional device applications and prospects for emerging photon source-based technologies. Beyond conventional and current achievements in optoelectronics, we will present hybrid nano-LED architectures. Novel device concepts potentially could play an essential role in future photon source developments and serve as a key component for optical computing. Therefore, forefront fully photon operated logic circuits, photon-based computational processors, and photon driving memories will be discussed. All these developments will play a significant role in a future highly secure, low energy consuming green IT. Besides today's environmentally friendly terrestrial industrial and information technologies, an enormous potential of nano-LED technology for a large range of applications especially in the next stage of space research is envisaged.

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I. INTRODUCTION

During the last few decades, the development of semiconductor materials and device architectures suitable for a large range of electronic and optoelectronic applications was driven predominantly by efforts to improve device performance. The main attention was focused on their miniaturization and the increase in device integration density. Nevertheless, efficiency limits of conventional material and device concepts seem to be already reached and, therefore, further performance improvement of nano-LED devices can be obtained only by alternative material and device architectures. Hence, unconventional solutions with respect to the increased device functionality, lifetime, and reliability have to play a central role from the early stage of device design. These efforts cannot be successful without a close interdisciplinary exchange between the physics and chemistry of materials with respect to efficient device engineering and technology. Therefore, here, in our Perspective, we will try to introduce current and possible future nano-LED's developments in the interdisciplinary context suitable for a broad scientific and industrial readership.

II. III-NITRIDE NANO-LED TECHNOLOGY

As it is presented in Fig. 1, the nano-LED's development has been driven primarily by arising practical requirements in lighting, displays, biology, and sensing. Besides III-nitride nanostructures for solid-state lighting,^{1–6} there has been a strong need for ultimate light sources serving in the large range of microdisplay applications for more than a decade.^{7–19} The trend is clearly set from micrometer sized LEDs down to nanometer scale devices. When targeting at III-nitride nano-LEDs, we must be aware of the special challenges that must be met in the material system. Due to the lack of large area commercially available group III nitride native substrates, group III nitrides are deposited on foreign substrates such as conventionally used c-plane sapphire, 6H-SiC, or (111) Si, which unfortunately have a large lattice mismatch of 16%, 3.5%, and –16.9% as well as a large thermal mismatch of –34%, 25%, and 54% to GaN, respectively.²⁰ This gives rise to dislocation densities in the range of up to 10^{10} cm^{-2} . Only after extensive studies and developments especially with respect to the crystal quality and p-doping in GaN,^{21–23} III-nitride based LED structures,^{24,25} the developments for which Akasaki, Amano, and Nakamura were

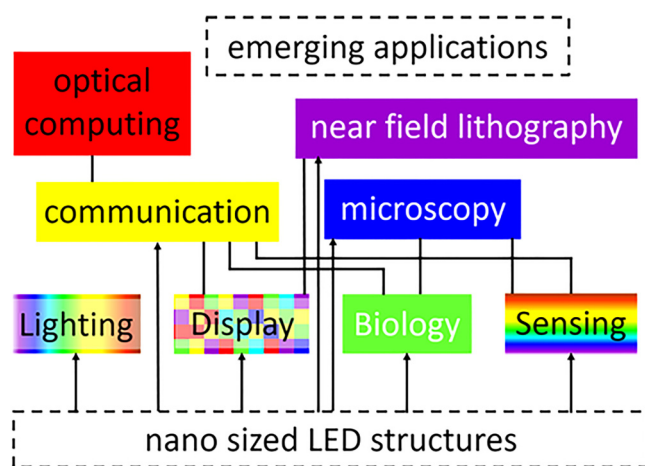


FIG. 1. Current and possible future developments for nano-LED applications.

awarded the Nobel Prize in 2014, could be deposited and LEDs of high brightness could be achieved. The special characteristics of group III nitride based LED structures must be taken into account when developing nano-LED technology in this material system.

Different technological strategies were developed and are now pursued for their preparation. In general, there are three different technological approaches for the production of III-nitride nano-LEDs. The approaches are divided conventionally into two main categories: bottom-up^{26–30} and top-down approaches.^{31–35} Furthermore, colloidal/solution-phase routes^{36–40} and advanced implantation techniques^{41,42} can potentially be useful to reach the mesoscopic (nano crystal LEDs) and down to the atomic scale—single point photon emitting sources. Here, in this Perspective, we will report only on the most representative recent and current approaches and achievements.

A. Bottom-up (epitaxial growth)

The two most common bottom-up techniques will be presented in the following. The first technique was reported in the 1960s:⁴³ nanostructure (usually nanowire) growth is induced by a particle—very often gold. The particle serves as a solvent for the elements the nanostructure is composed of. A eutectic with the particle element is formed. The diameter of the particle affects the nanostructure's dimension and its position determines its location.⁴⁴ Chemical vapor deposition methods such as MOVPE (metalorganic vapor phase epitaxy) is the method of choice for growth since the particle induces the decomposition reactions at lower temperatures than for layer growth. However, the unintentional incorporation of Au in the nanowires⁴⁵ as a non-radiative recombination center is detrimental to carrier mobility and to optical properties of the nanostructures.

The second technique of the two bottom-up approaches is based on selective area growth (SAG)^{28–30,46} and again involves MOVPE. Here, the substrate surface is covered with a masking material such as SiO₂ or Si₃N₄. In contrast to the substrate, the

masking material should not participate actively (catalytically) in the decomposition process of the precursors involved. By preparing holes/windows in the mask, in appropriate size, growth can be induced selectively only in the apertures of the mask, and micro- and nano structures⁴⁷ are prepared. We believe that this second technique of the two bottom-up approaches has a larger potential for future nano-LED applications. Therefore, in this Perspective, we will focus on this bottom-up technique and present it in more detail (see Fig. 2).

Different types of nano-LED structures can be deposited and were successfully demonstrated, e.g., vertically grown/bottom-up p-GaN/In_xGa_{1-x}N nano-dot/n-GaN(LED) and p-GaN/MQW/n-GaN

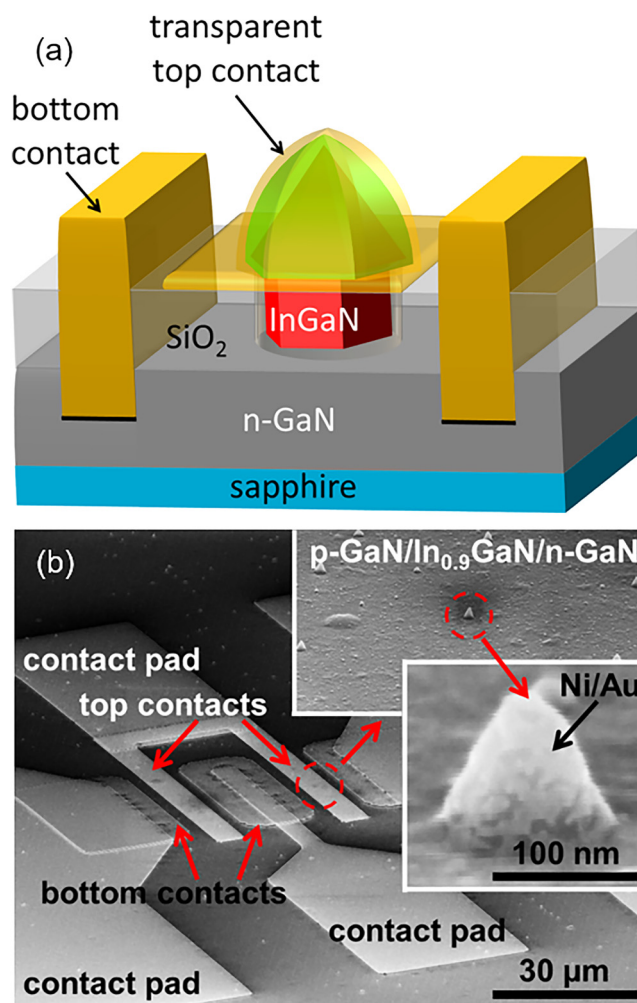


FIG. 2. Schematics of the integrated p-GaN/InGaN nanopillar mesoscopic structure grown on an n-GaN/sapphire template (a). SEM images with different magnifications of the final device with the hexagonally arranged structures and a detail of a single nanopillar with Ni/Au top contact (b). Reproduced with permission from Mikulics *et al.*, Appl. Phys. Lett. **109**, 041103 (2016). Copyright 2016 AIP Publishing LLC.

junctions as well as nano-LEDs consisting of a bottom-up deposited GaN wire core overgrown by a heterojunction shell.³ The dimension of the LED on the nanoscale affects the emission wavelength.⁴⁸ Here, in this section, we will shortly present the most important steps toward vertically integrated III-nitride based nano-LEDs, which were designed and fabricated for operation in the telecommunication wavelength range in the (p-GaN/InGaN/n-GaN/sapphire) material system.⁴⁹ The fabrication process started with the growth of n-GaN on sapphire substrates by MOVPE followed by the preparation of hexagonally arranged “hole” structures in the SiO₂ mask thereon. SAG of InGaN nano-pyramids was then carried out followed by the deposition of the p-GaN at last. The growth procedure allows for strain relaxation in the nano-pyramid heterostructure. The details are reported in Refs. 49 and 50. Figure 2(a) presents schematically the device structure of vertically integrated p-GaN/InGaN nano-pyramids grown on n-GaN/sapphire. After preparing bottom and bottom top contacts to the structures integrating them in a device circuit [Fig. 2(b)], they are suitable for future operation in the telecommunication wavelength range.⁴⁹

The choice of the substrate is a further variable in this bottom-up approach. Conventionally, c-plane sapphire, (111) Si, or 6H-SiC is chosen, depending on the application envisaged. It has been shown that the employment of a metallic substrate could be advantageous for device concepts and device performance. Besides being the growth substrate, it can simultaneously serve as the bottom electric contact.⁵² Furthermore, it can effectively dissipate heat during operation⁵³ as was demonstrated for HFET devices⁵¹ presented in Fig. 3. Therefore, a metallic substrate could also improve the long-term reliability of LEDs. In addition, the functionality of LEDs and detectors can be enhanced by the metallic substrates since they can act as efficient mirrors. Up to now, metallic substrates were used for planar heterostructure growth.^{51,54,55} They may have an even larger potential for nanostructure SAG

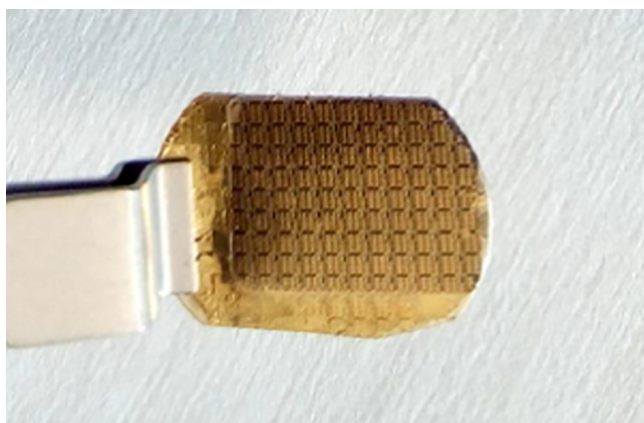


FIG. 3. First demonstration of successful growth and fabrication of III-nitride heterostructure field effect transistor (HFET) devices (more than 70% of tested HFETs achieved the expected device parameters) on metallic silver substrate, which exhibits a high thermal conductivity coefficient of $\sim 430 \text{ W m}^{-1} \text{ K}^{-1}$ at 300 K. To this end, the development of a novel combined MOVPE and MBE procedure was reported. More details can be found in Ref. 51.

since differences in thermal expansion coefficients and lattice constants between epilayers and metal play a minor role for nanostructures.

B. Top-down (etching)

In general, there are two top-down etching approaches for nano-LEDs, both of which are based on an epitaxial (planar) LED layer structure deposited on the respective substrate. In the first step, nano-LEDs are defined with the help of a masking material that must withstand the following dry etching procedure. The masking material is patterned using e-beam lithography. The pattern is then transferred into the LED layer structure by dry etching.^{35,56} The definition of nanostructures leads to strain relaxation which, in turn, affects the wavelength of the emitted light.^{35,57–60} As the structure size will further decrease in the future, the proportion of the surface area of the nanostructures will increase as well. This surface exhibits etching related damage, which is detrimental to the nano-LED's optical properties. Therefore, it will become increasingly essential to develop processes, which improve the etched LED optical properties as the structure size decreases further.

It has been demonstrated that a careful local laser micro annealing process (LMA)⁶¹ increases the radiative recombination, the long-term stability, and reduces the work temperature of the nano-LEDs under operation. Basically, the LMA procedure, reported in Ref. 61, affects both the Ohmic contact and structural MQW properties individually for every single nano-LED device or the ensemble of nano-LEDs patterned in large area arrays. A significant effect of LMA on the nano-LEDs' optical properties was contributed to the reduction of “etching” defects caused during the reactive ion etching (RIE) process employed for the nano-LED formation. Figure 4 presents a scanning electron micrograph (SEM) of a single nano-LED with its nickel mask after the RIE procedure. The nano-LED's side “walls” exhibit a defective surface morphology, indicating that also deeper regions of the MQW, as well as p-GaN and n-GaN regions, are affected. Figure 5 presents schematically in a simplified form the effect of RIE on nano-LEDs with

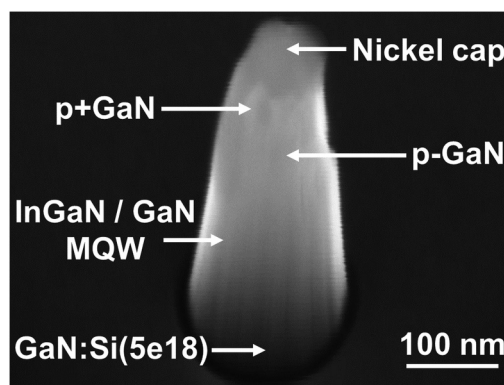


FIG. 4. SEM image of a single nano-LED with its nickel mask after the RIE process. Reproduced with permission from Mikulics *et al.*, Appl. Phys. Lett. **118**, 043101 (2021). Copyright 2021 AIP Publishing LLC.

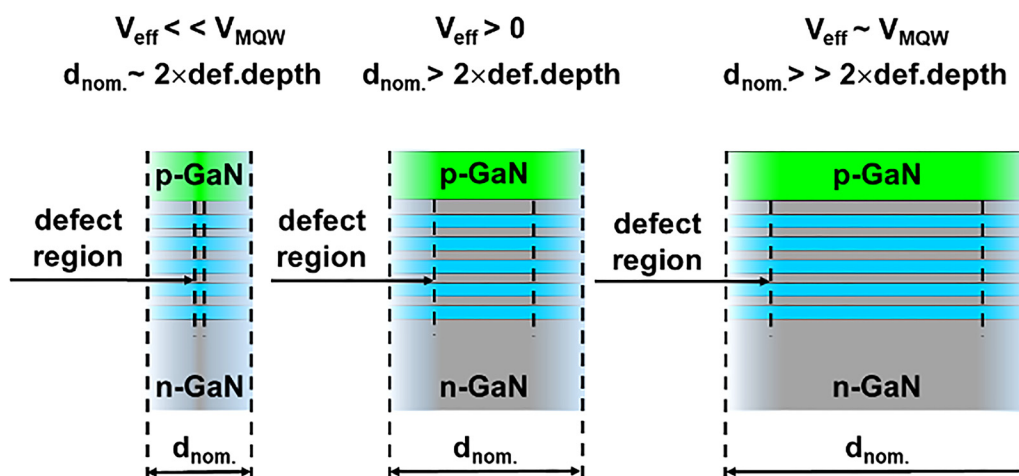


FIG. 5. Simple model showing the effect of the defect layer depth on the virtual equivalent of the effective volume (V_{eff}): the larger the effect is, the smaller the nominal diameter $d_{\text{nom.}}$. Reproduced with permission from Mikulics *et al.*, Appl. Phys. Lett. **118**, 043101 (2021). Copyright 2021 AIP Publishing LLC.

different diameters. The LMA process affects the defect region depth, which decreases after optimal laser annealing conditions were applied. This effect was also confirmed by photoluminescence measurements⁶¹ for non-locally annealed and annealed nano-LEDs in the wavelength region where InGaN/GaN MQWs emit. The mechanism behind this effect was explained by the simple model introduced in our previous study.⁶¹ Therefore, it could be expected that a careful tuning of the LMA process could be of benefit for nano-LED's optical properties by further miniaturization efforts.

Nano-LEDs prepared by this top down etching approach in arrays can be applied to future lithography (presented later in Sec. IV B), in the near-field regime or the far field regime which will help greatly simplify structuring processes from the nanoscale to the microscale.⁶²

In the second approach, the structures are formed without a mask in a self-assembled way.^{63–66} Dry etching is carried out and etching takes place preferentially at charged sites such as at threading dislocations. A re-deposition of the SiO₂ cover plate material in the reactive ion etching (RIE) chamber holder toward the nanowire sidewalls occurs protecting them from further etching and simultaneously increasing their length during the etching process.^{64,65} Therefore, highly anisotropic etching occurs that is not positioned but rather random, i.e., self-assembled. Figure 6 presents SEM images of the nanowires formed in three different magnifications, and Fig. 7 shows how the formation is affected by the etching conditions. The high density and the conical shape of the nanostructures produced by this second top-down etching approach correlate with the concentration of threading dislocations in the original planar group III-N layer. Transmission electron microscopy (TEM) studies reveal that the etching procedure removes the defects—the nanowires are defect-free. The nanowires are advantageous for the suppression of incident light reflection. A graduated strain in the nanostructures in the axial direction is responsible for effective photon absorption of more than 95% in a large range from “blue”

to “red.” We call the family of materials—“black” nitrides in analogy to “black” Si.^{67,68} They are of special interest for photovoltaic applications.

C. Hybrid nano-LED architectures

The nano-LEDs prepared by a positioned top-down approach can also be employed in numerous hybrid applications. Current achievements have already demonstrated that III-nitride based nano-LEDs are very promising multifunctional photon emitting devices, which can be used as hybrid photon emitting sources. Figure 8 presents simple schematics with an integrated single nano-LED structure driven electrically and freestanding CdSe nanocrystal. Such hybrid architectures can serve as testing platforms for the investigations of plasmonic effects. This could be of benefit, especially for the sake of field enhancement at the position of a light emitting nanoparticle. A CdSe crystal (or any other nanocrystalline material object) serves as the “host” = secondary photon emitting source. This photon-generated technique was reported as direct electro-optical pumping.⁶⁹ Furthermore, it can be envisaged to be part of a future fully photon operated transistor structure⁷⁰ or used for testing and characterization, in which they are integrated into a platform for simultaneous investigations in a bottom-up geometry. Possible applications will be described later in Secs. IV A and IV D. An example of hybrid nanocrystal/III-nitride nano-LEDs fully integrated in an HF device layout is presented in Fig. 9. This device concept can be principally used for future single photon emitters where, e.g., nano-diamond crystals with N-vacancies^{71–73} serve as the secondary photon emitters. Furthermore, the technology developed is applicable to a large range of biological, analytical, and communication applications and is suitable for the large production scale.⁷⁴ An example of a prototype-integrated hybrid photon emitter ensemble, which can serve as primary electro-optical converters and/or the “interface” between “outer” conventional electronics and

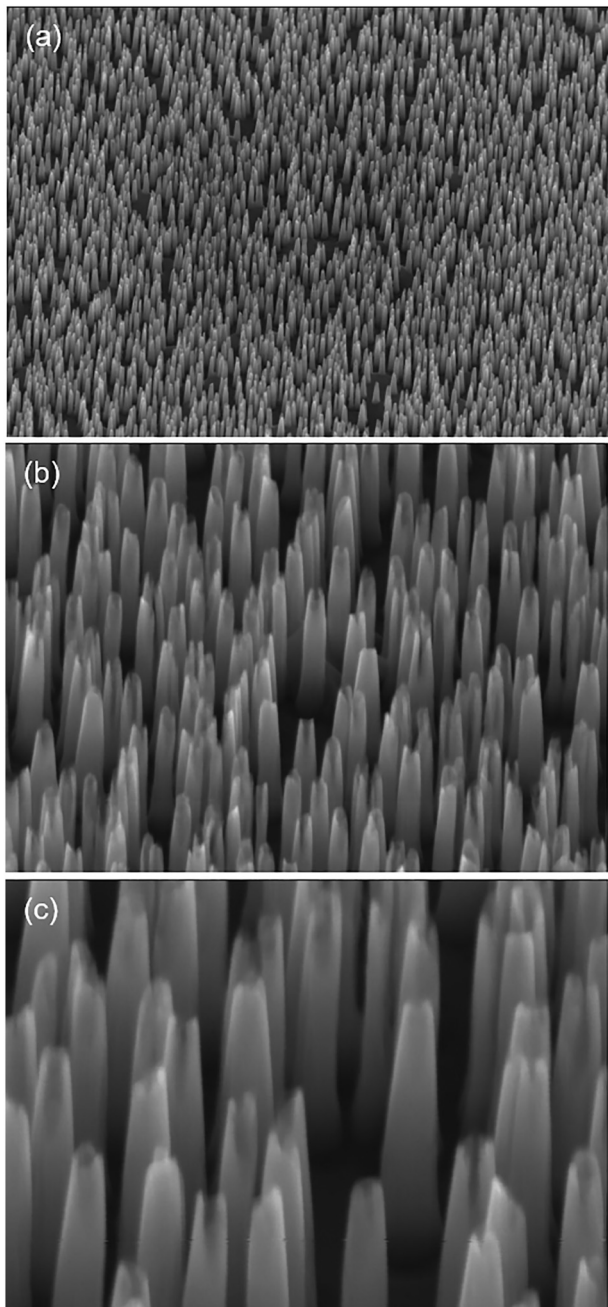


FIG. 6. SEM images of self-assembled nanowires formed by the dry etching process in different magnifications (a)–(c).

an “inner-core” of future optical computing systems, e.g., photon operated processors, is presented in Fig. 10. To this end, the realization of such optical computing systems would require next generation hybrid nano-LED developments. They could be driven and inspired by unconventional material and device architectures. It was

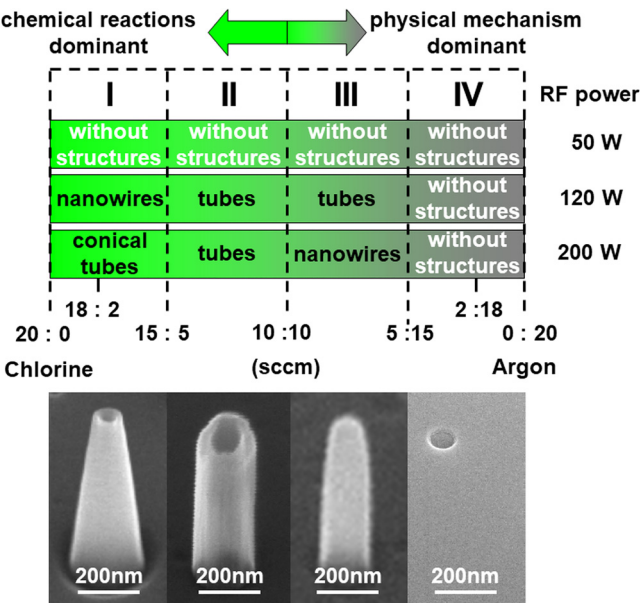


FIG. 7. Overview of nanostructure (GaN) formation obtained for different Cl_2/Ar etching gas mixtures and different RF powers. The ICP power was kept constant at 2500 W. Reproduced with permission from Haab *et al.*, Phys. Status Solidi 209, 443 (2012). Copyright 2012 Wiley-VCH.

already proposed, about a quarter century ago, in pioneering work by Goldman and co-workers⁷⁶ that light emitters can be formed in mixed anion nitride/arsenide alloys. Finally, special attention should be paid also to device concepts that are based on and take advantage of plasmonic materials.^{77–81}

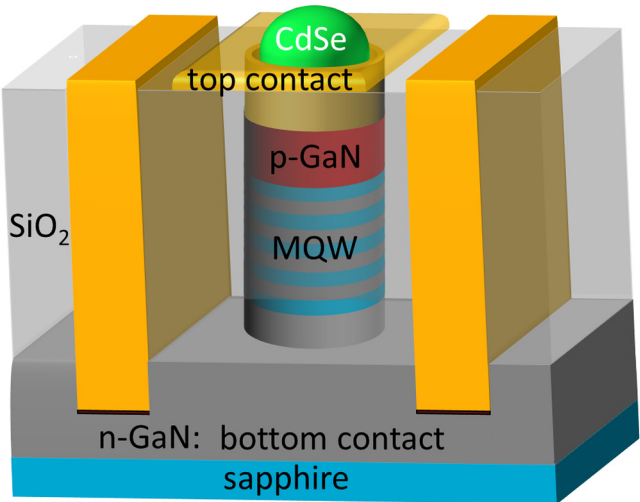


FIG. 8. Principal schematics of the hybrid/III-nitride based nano-LED structure integrated in the vertical device layout.

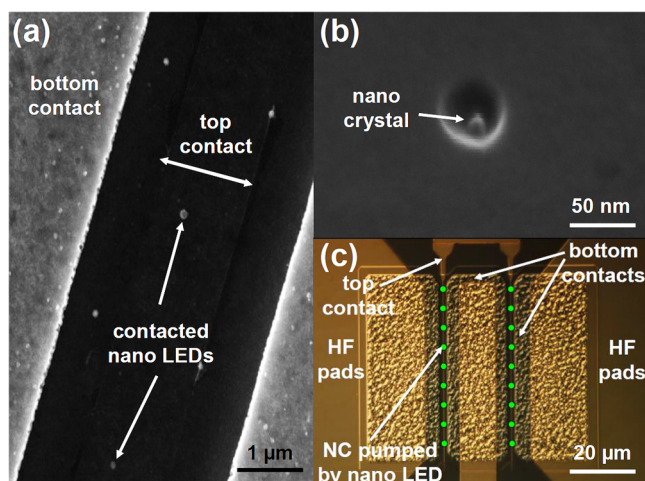


FIG. 9. SEM micrographs: (a) array of nano-LEDs with NCs, a transparent Ni/Au top contact and annealed Ti/Al/Ni/Au bottom contacts. (b) Nanocrystal (NC) positioned in the SiO₂ hole structure. (c) Fully integrated NC/nano-LED structures (green dots as guides to the eye) in the device layout suitable for DC and HF characterization.^{83,75} Reproduced with permission from Mikulics *et al.*, Appl. Phys. Lett. **108**, 061107 (2016). Copyright 2016 AIP Publishing LLC.

III. PRIMARY NANO-LED APPLICATIONS

A. Lighting

Although an enormous amount of work was done during more than a decade on the miniaturization of LEDs toward micro- and nano-sized devices,^{2,3,27,34,82–84} there is still a strong need for

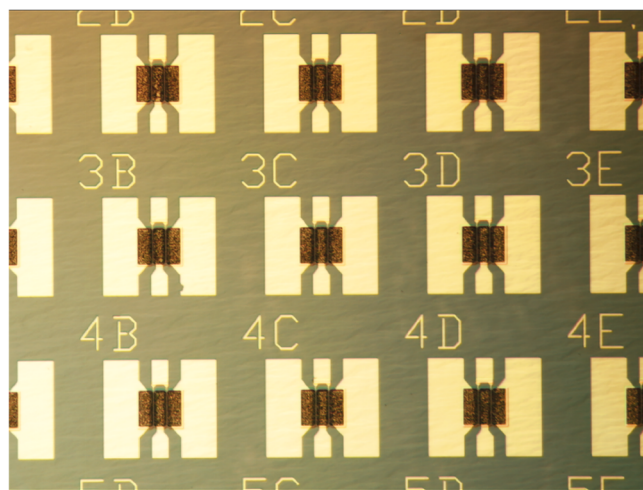


FIG. 10. Ensemble of hybrid photon emitter devices fabricated on a III-nitride material layer system suitable for HF operation. This device prototype can serve in future developed computing architectures as primary electro-optical converters (necessary for the provision of an “interface” between “outer” conventional electronics and an “inner-core” of future transistor based optical processors). More details can be found in Sec. IV D.

further innovations with respect to the significant improvement of their wall-plug efficiency, device lifetime, and reliability. Further important drivers for extensive “lighting” research and especially for “phosphors free” white-light emitting diode/sources are a decrease in production costs and the industrial requirements for the avoidance of “rare” materials. Additionally, advanced LED production technologies have to fulfill strict standards for environmental safety. Therefore, major efforts during the next few decades will be focused on environmentally friendly technologies. The future production lines will be certainly impacted by and confronted with the accessibility of the limited material resources. Hence, the arising future LED products should be designed from the start toward being fully recyclable after reaching their life cycle. The aim of our Perspective is not to give an additional overview of the past and current achievements in the micro- and nano-LEDs research field. We focus our attention rather on unconventional future possible solutions and developments.

In Sec. II, we already mentioned that further technological progress in production and the improvement of nano-LED devices could be reached if metallic instead of conventional substrates would be used for growth. This is indeed not a trivial goal and the realization of such a technological procedure especially for mass production lines would require certainly a decade of extensive research efforts in chemical and physical deposition procedures on metal substrates far beyond today’s general knowledge. Nevertheless, the first step toward the epitaxial growth of III-nitrides on silver substrates was presented by Mikulics and his co-workers,⁵⁴ and diode structures on Ag/AlN/GaN were successfully fabricated and tested. A further milestone in the growth of III-N heterostructures was reached in 2017.⁵¹ Again, HFET devices were fabricated based on the III-nitride layer system, which were also deposited on silver substrates. At this point, it should be noted that the HFET devices presented in those studies exhibited electrical parameters comparable with devices grown on conventional substrates and that the improved thermal heating management increased their device reliability significantly. The application of metallic substrates for III-nitride based devices such as high power diodes, HFETs, and possibly also for nano-LED devices has several advantages. The metallic substrates play a significant role in effective heat dissipation but their role as a “bottom” electrical contact opens new possibilities for device design and the realization of unconventional architectures. Additionally, the positive effect of internal metallic “mirrors” as a part of the III-arsenide layer system for highly efficient and femtosecond photo-detectors⁵⁵ confirms the suitability of this approach for low-cost and large-area fabrication of electronic and ultrafast photonic devices that require a highly effective thermal drain. Therefore, it could be expected that the use of metallic substrates, in the future, for the growth of LED structures will play an essential role in their device performance improvement. The effective heat management inside of such LED ensembles would also significantly contribute to the increase in their device reliability. Last, it has to be noted that the growth of nano-LED structures on metallic substrates (a large amount of published work was already demonstrated on metal seeded “catalyst-assisted growth”^{36–40}) could be beneficial since such structures would be less-affected by strain additionally. These effects on the nano-LED’s optical and electrical properties could bring new insights into LED device physics.

B. From micro- to nano-LED displays

Displays, in general, are the most prominent part of consumer electronics, which surround us in a large range of technical solutions for communication ranging from cell phones to notebooks as well as a large range of monitors/screens as a part of computer techniques and TV electronics, automotive visual interfaces, and many others. All these solutions fulfill a basic function—namely to transfer data/information in visual form—in the most effective way for humans. The large amount of information that we “collect” from our environment is based on the ability of our human eye to acquire information carried by photons. The sensitivity of our eyes has adjusted to the maximum wavelength range of the sun, which we conventionally call the VIS (visible) range, and is the result of a biological evolution process. There is no doubt that displays based on singularly addressable ensembles of organic and inorganic LED devices will play also in the future a dominant role in communication. Therefore, most forefront research efforts will be focused predominantly on the energy efficient generation of pure monochromatic light.^{85–90} Indeed, current achievements confirm that quantum dot based LED solutions⁹¹ deliver unprecedented results in comparison with conventional display technologies. Hence, one would expect that further miniaturization of LED pixels from the micrometer range down to the nanometer scale would increase the “quality” of visual data transfer significantly. This will certainly not be the case for displays used for TV devices since the ability of the human eye for such high pixel resolution has natural limits. On the other hand, the recent development of the “family” of vitreous products with integrated displays confirms that there is still a strong need for further LED miniaturization.

Here, in our Perspective, we will focus our attention rather on possible developments in unconventional applications (Fig. 11) of display technologies after the appropriate nano-LED solutions will reach the development stadium necessary for a consecutive successful mass production. Therefore, research activities toward highly efficient nano-LED devices should also be concerned with hybrid LED architectures.^{69,79} Indeed, this young research field already yielded a large amount of solutions. Nevertheless, unconventional possible-future applications of nano-LED based displays such as for human medicine,⁹² biology,⁹³ and analytical chemistry⁹⁴ would require a paradigm change in device design and material science since the long-term biological-nano-LED device compatibility seems to be a key factor for achieving the required functionality. This is a crucial point, especially in the case of implantable displays.^{11,95} One of the very promising applications of nano-LEDs which is introduced later in Sec. III C is based on the suitability of micro- and possibly nano-LED arrays for the controlled photostimulation of neurons⁹² by means of their light emission. Another important application of nano-LED displays could potentially be to significantly decrease production costs in the semiconductor industry by enormously simplifying device fabrication if a novel maskless lithographical technique^{35,96} would be established in mass production lines. We describe this path in more detail in Sec. IV B. In addition, it was already demonstrated (about four decades ago) that especially “red light” emission is of benefit for healing processes increasing cellular metabolism, etc.^{97–99} Therefore, research in the field of “human-organism” compatible nano-LED displays could be

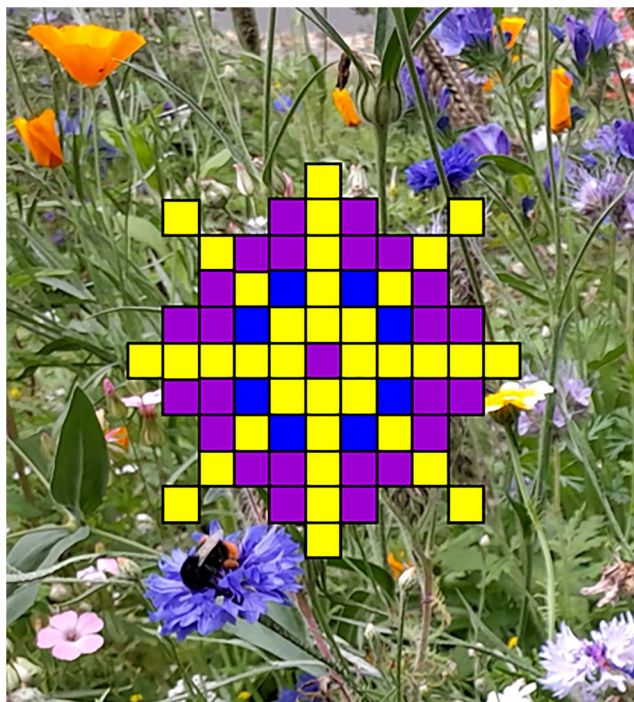


FIG. 11. Future developments in nano-LED display technology can possibly open new horizons in unconventional future display application research fields such as displaying information for insects (attraction, warning, etc.).

essential for the development of novel medical/therapeutic techniques not only for applications outside the human body but rather “inside.” They could be implanted onto human organs or selected organ regions down to the precise targeting of single biological cells. The full impact of such possible-future developments, of course, cannot be estimated in this limited perspective format.

C. Biology: Optogenetic and neuronal stimulation

In this section, we will start with the report on the application of micrometer sized LED structures in medicine for cardiovascular-monitoring first. Although a large amount of work was done during the last few decades in this research field and the results found successful commercial responses in various products, there is still a strong need for further innovations for cardiovascular monitoring and related activities. Today’s commercially available solutions for the estimation of “heart rate” in real time are based on the processing and monitoring of photoplethysmographic (PPG) signals recorded from wearers’ wrists.^{100–104} Current achievements in this research field presented recently by Ye and co-workers^{105,106} demonstrate successfully the application of III-nitride monolithically integrated MQWs with the functions “light emitter” and “detector” devices as a part of an asymmetric optical link in the form of a miniaturized multifunctional chip. This approach based on wireless light “communication” in the “chain” between the micrometer sized light emitting diode (LED) device—the vascular bed—and the detecting unit allows

to monitor information about arterial blood transport. The basic idea behind this concept is based on the mechanism that follows in real time the changes in the vascular bed. They act as modulating retro-reflectors (MRRs). Therefore, MRR monitoring allows obtaining both the person's heart rate and cardiac-related pulse information. It could be expected that further miniaturization of these multifunctional "emitter" and "detector" based III-nitride MQW devices toward the nanoscale in the form of functionally arranged nano-LED arrays could introduce new research possibilities for the local scanning of blood pressure, transport, and distribution in blood vessels down to the human-intercellular and cellular range. The information from such "micro" and "nano" dimensions on the cellular scale and the possible strain changes on this "low" dimensional level would potentially increase information "mining" and help to diagnose the biological effects of medicaments and/or pathological developments in the blood vessels walls. A plethora of applications can be envisaged. The full potential and new future possible developments and discoveries in human medicine based on the employment of nano-LED devices, however, cannot be covered in this Perspective.

Therefore, in the following, we will focus our attention only on the most important further possible developments for nano-LED structures, which are not limited only to cardiovascular-monitoring medicine. More than a decade ago, Edward S. Boyden and his collaborators/co-workers demonstrated a genetically targeted optical control of neural activity on the millisecond time-scale.¹⁰⁷ Optogenetic research in the following years brought a large amount of new insights into the understanding of biological-chemical-physical interaction mechanisms based on neuronal photostimulation.^{108–114} The suitability of micro-LED arrays for the purpose of photo-stimulation of neurons was successfully demonstrated.^{110,113–118} Hence, it could be expected that nano-LEDs could play an important role in future neuroscience research. Because of their dimensions, local and focused low emitting (optical) power⁶² together with the simultaneous low energy consumption, they are predestined to be implanted spatially localized in the future in diverse biological systems and especially in brain regions such as the cortex cerebri, targeting selected neurons. In such a way, the stimulated brain regions responsible for saving information, for example may significantly improve data processing ("recall" of saved information or improvement of the data saving process itself) or serve as an effective tool and/or part of neuro-therapeutical techniques. Indeed, there is potentially a large amount of applications in neuroscience. Nano-LEDs in the form of single devices for stimulated optical pulse generation or in integrated and functionally addressable arrays will open a new era in neuroscience and related medical fields. However, before these applications could be realized open questions regarding the nano-LED's bio-compatibility, the prevention of possible toxic effects, the suppression of corrosion, and others have to be answered.

D. Sensing

In general, it could be expected that nano-LEDs as sources of photon emission for various functional sensing applications are beneficial because of their low energy consumption during operation. Additionally, there is a strong need for the further

miniaturization of nano-LED devices since their use in different sensors could potentially decrease production costs and save material resources significantly. It was already demonstrated and reported that LEDs especially in the form of micro- and nano-sized devices are essential for state of the art and coming sensor developments.^{119–129} Excellent reviews have already been published by Waag and co-workers,^{5,130} which already covered past as well as current achievements and brought a broad overview of sensing applications as well as an outlook for further possible developments. In contrast, in this Perspective, we would rather like to focus our attention on future unconventional trends.

Therefore, we will start with the most important advantages of the use of nano-LEDs in future sensing technologies. They are associated with the generally well-known expectations and requirements for ultimate sensor devices: first, by means of their low dimension, nano-LEDs allow high integration device density. Hence, it could be expected that ensembles of singularly addressable "light emitting and detecting" pairs (units) could reach total device densities up to 10^{10} per inch square (based on, for example, nano-LEDs with a diameter of 50 nm and a device spacing to neighboring devices up to 100 nm). Such device densities would already be reachable for nano-LED device definition and addressing (for contacting bit and word lines) using state of the art conventional e-beam lithography and deep ultraviolet photolithography (DUV) techniques. Novel highly sophisticated, and complex sensor device architectures would allow for singularly addressable and/or programmable arrays of wavelength tunable sources and unprecedented functionalities. In contrast to the conventional LED as well as to micro-sized LED devices, their light emission spreads out across macroscopic regions of the device. Therefore, it is expected that the application of nano-LEDs with dimensions below 100 nm in diameter for the "far" field emission regime (as well as the decrease in their structure pitch) can be essential for improving the localization of the light source. Furthermore, nano-LED devices equipped with appropriate apertures for employment in the "near"-field regime⁶² as is described later in Sec. IV B can be essential for the development of next generation sensing systems. This could unfurl a new era for a family of sensor devices with olfactory functionality for drug and explosive detection (for security reasons), environmental monitoring, pheromone detection, and many others leading to an enormous increase in the sensitivity toward potentially hazardous and toxic molecules in concentrations far below the detection level limits of today's-existing sensors. In addition, chemical and biological analytics as well as human medicine (illness diagnostics) can profit from these developments since analyses and identification of complex "smells" (molecular mixtures) in a large range of concentrations would be possible.

IV. ADVANCED NANO-LED APPLICATIONS

A. Super resolution microscopy

Currently, one very promising application for LED devices covering the micrometer down to the several hundred nanometers scale range is targeted at diverse "lens-free" microscopy techniques.^{131–136} Previous and current efforts in this scientific field were already summarized and described in detail by Wasisto *et al.*⁵ in their excellent review. Significant technological steps

toward super resolution microscopy were presented recently by Bezshlyakh and co-workers¹³⁷ as well as by Kluczyk-Korch and co-workers.^{138,139} Besides others, they presented novel driving architectures for singularly addressable nano-LED structures (arranged in a functional array) with a size and pitch (distance to neighboring emitters) below the emission wavelength. These developments represent a big step forward for this novel super resolution microscopy technique. Such a nano-LED arrangement allows the control of individual LED emissions laterally at the nanoscale, which results in a spatial precision below the wavelength of light emitted and possibly even below the diffraction limit.¹³⁷ Hence, singularly addressable nano-LEDs with a nanoscale pitch would allow the creation of light patterns with unprecedented spatial resolution. Such computer controlled integrated nano-LED ensembles serving as light emitters for illumination systems could potentially be applied in a large range of microscopy and spectroscopy research fields in biology, molecular chemistry, mineralogy, metallurgy, and many others. As we already mentioned in Sec. III, future developments in nano-LED applications will not only be limited to “basic” research fields such as “biology” with its sub-research areas emerging optogenetic and neuronal stimulation, but they could possibly usher in a new era for unconventional display and sensing applications. In particular, these two last mentioned application fields would have a direct impact also on developments of novel emerging super resolution microscopy techniques and vice versa. However, further efforts have to be focused on the improvement of the nano-LEDs’ wall-plug efficiency, the reduction of heat development, the optimization of thermal heating management (possibly by using unconventional “metallic” substrates and novel device architectures), as well as on increasing the nano-LEDs’ device lifetime and their long-term reliability. Special attention should be paid to the multiple functionalities of MQW devices as it was already presented by Wang and co-workers (see, for example, Ref. 140) and is also reported in this Perspective in Sec. IV C. The simultaneous operation of the nano-LEDs in the transmitter and receiver mode could be advantageous also for further developments in super resolution microscopy techniques. The anticipated success in these research fields would play a decisive role in the realization of future emerging nano-LED applications. Therefore, in our opinion, key requirements for the realization of emerging super resolution microscopy techniques cannot be fulfilled without significant progress in “near”-field^{35,96} and hybrid nano-LED solutions,^{69,74} described in Sec. IV B.⁶² They seem to be essential for reaching the ultimate resolution limits in super resolution microscopy.

B. LEDALIT: Nano-LED assisted lithography

Nowadays, the fabrication of an abundance of integrated circuits requires highly sophisticated lithographical techniques. It is well-known that the manufacturing of electronics for different applications (such as customer ICs for information technology, consumer electronics and optoelectronics, automotive and aviation industry, etc.) requires different lithographical techniques ranging from conventional lithography down to emerging EUV lithographical techniques (e.g., primary storage memories, processors, etc.). There is no doubt that the continual increase in demand for new solutions and products targeting especially consumer electronics

drive further developments toward unconventional fabrication techniques. However, the scaling of electronic and photonic devices down to an “a few” nanometer range is confronted with a huge increase in production costs as well as material and energy resources. Therefore, the simplification of existing lithographical techniques could significantly contribute to further progress. One possible solution can be the application of nano-LED devices in the photolithographic process. They can be implemented in singularly addressable arrays (analogous to nano-LED based display technology—introduced in Sec. III) and serve simultaneously as photon sources that could on demand create lithographical patterns flexibly without the need for a mask. The suitability of nano-LEDs for this purpose was already demonstrated in our previous work^{35,62} and patterned structures in a conventional photoresist (using the illumination “far” field regime) with sizes up to several hundreds of nanometers were realized. Additionally, we demonstrated that nano-LED structures with metallic apertures can be used in the near-field regime (the principle presented in Fig. 12). The generated evanescent field initializes locally a photochemical reaction in photosensitive molecular films. In such a way, it was possible to fabricate “hole” structures with a diameter down to ~ 75 nm.⁶² The ultimate limit for the fabrication of “molecularly” sized structures could be reached if single photon sources could be realized in large arrays and could be singularly addressable, which would allow the mass production of molecular sized devices. The structure size limits for this technology are theoretically set to the size of a single photosensitive molecule since one photon of sufficient energy induces the reaction in the ideal case and could alter just one chemical bond. Of course, in a “real” photoresist, a higher yield of incident photons is required for the photochemical reaction. However, before this concept can be realized, suitable single

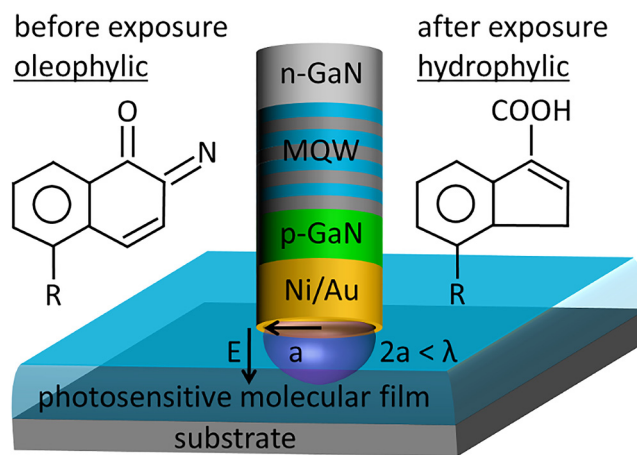


FIG. 12. Principal schematics: nano-LED (n-GaN/MQW/p-GaN) element in the near-field regime to a photosensitive film generating an evanescent field. The fundamental goal of “near-field” LED assisted lithography (LEDALIT) is to create a sub-diffraction limited field distribution in a location favorable for a photochemical reaction.⁶² Reproduced with permission from Mikulics *et al.*, *Nanoscale Adv.* 2, 5421 (2020). Copyright 2020, licensed under a Creative Commons Attribution (CC BY) license.

photon sources operated at room temperature have to be developed. The device architecture introduced as a “hybrid” nano-LED structure in Sec. II can serve as a platform for the next generation of single photon sources.⁷⁴ Indeed, it was already demonstrated that nitrogen vacancies in diamond can emit single photons at room temperature.^{71–73} Therefore, the combination of, e.g., diamond nanocrystals^{71,72} integrated with nano-LED structures could be essential for the realization of single photon lithography.⁹⁶

C. Communication

Recent advances in III-nitride micro-LEDs have also led to the development of monolithic photonic circuits for on-chip communication. These efforts were demonstrated in pioneering work by Wang and his co-workers.^{140–150} Their reports especially on photonic circuits based on a III-nitride MQW layer structure using silicon as the substrate demonstrate multiple functionalities such as a transmitter, modulator, receiver, and a waveguide for transmitting optical pulses. Furthermore, the authors presented for the first time also the successful direct modulation for on-chip data communication performed on monolithically integrated components. The most important message from their studies confirms that III-nitride MQW micro-structures implemented into a functionally arranged device layout provide a spectral overlap between emission and absorption spectra. This allows their use simultaneously in both the emitter and detector regime. Therefore, the presented device architecture is advantageous also for the development of future photonic driving circuits. The further miniaturization of emitter sources down to the nanometer scale will lead to an increase in integration density as well as to a significant decrease in energy consumption. However, for the reduction of LED size from the microscale down to nanoscale, an increase in technological efforts is essential, especially with respect to the suppression of the non-radiative recombination mechanisms. In Sec. II B, we already mentioned that a careful tuning of etching parameters for nano-LED fabrication and a subsequent local annealing process can significantly recover damaged MQW regions inside the nano-LED structure.⁶¹ The further development and optimization of these advanced LMA (laser micro annealing) techniques for conditioning nano-LED structures could also be of benefit for “deep” water and as well as for satellite “short” orbital distance wireless light communication applications. Indeed, strong demand for radiation robust and reliable light emitting sources such as III-nitride based LEDs was reported by several groups.^{151–162} Nano-LED emitting arrays, as well as hybrid single photon sources could certainly play an important role in arising highly secure satellite communication. It could be expected that the potential of nano-LEDs arrays will not be limited only to data communication and metrology but could also play an essential role in sensor developments. This was described in Sec. III D. Highly reliable, long-living, and radiation tolerant nano-LED arrays could play a fundamental role in future satellite navigation, tracking, and autonomous “intelligent” emergency systems for the prevention of satellite collisions. The first steps in these developments were already done,^{151–162} and the proof of principle for wireless light communication applications of micrometer sized MQW devices with their multiple functionalities was demonstrated by several research groups.^{163–169} There is no

doubt that a strong need for environmentally friendly material and device optoelectronics, which exhibit low energy consumption, radiation robustness, and high device reliability, will be the driving force for innovations accelerating developments in this young nano-LED research field.

D. Optical computing

Finally, the tentative last nano-LED’s application discussed in this Perspective focuses on future optical computational techniques. During the last few decades, scientific teams and R&D companies around the whole Globe reached a meaningful progress in diverse research fields targeting quantum photonics and related techniques.^{170,171} The most important developments and current achievements were summarized and reported by Slussarenko and Pryde¹⁷² and Elshaari *et al.*¹⁷³ The detailed “Roadmap on Integrated Quantum Photonics” was introduced by Moody *et al.*¹⁷⁴ Indeed, there are important milestones that have to be reached until the technology matures sufficiently to allow for large scale and mass production suitable for “photon operated computational systems” in “daily life” such as recently reported by Taballione *et al.* in the paper entitled “A universal fully reconfigurable 12-mode quantum photonic processor.”¹⁷⁵

Since single photon emitters seem to be a “key” component for integrated quantum photonics,¹⁷⁶ it could be expected that novel nano-LED hybrid architectures suitable for single photon generation could play an essential role in these developments. On the other hand, the trend toward simple and cheap alternative “optical computational” techniques suitable for mass production, which could possibly replace/substitute the conventional computing electronic based system during the next few decades, call for an increase in research efforts for highly efficient, long-term operated, and reliable optical emitters. In this case, nano-LEDs integrated into sophisticated HF device layouts^{49,61,69} can serve as primary electro-optical convertors necessary for the provision of an “interface” between “outer” conventional electronics and an “inner-core” of photon operated processors. In addition, in the future, simple optical processor architectures analogously to today’s operated “conventional” processors as well as memory/data storage units, etc. could replace also electronic processors in such applications like satellite communication. It is well known that especially the group of III-nitride based electronic and optoelectronic devices were in the past identified as very promising candidates for a harsh and radiative environment. Therefore, it could be expected that nano-LED devices will play a significant role also in the future for integrated circuits in optical computing based on robust transistor units (one of the “youngest” representatives of the large optical switches family).^{70,178} A HRTEM image in Fig. 13 displays the “atomic-scale” detail of a $\text{Ge}_1\text{Sb}_2\text{Te}_4$ nano-membrane structure¹⁷⁷ as a “core” material for such transistor devices. The successful operation in the “two-color” mode was demonstrated. It could be expected that III-nitride based nano-LEDs operating in an appropriate wavelength range could be integrated with the, e.g., $\text{Ge}_1\text{Sb}_2\text{Te}_4$ nano-membrane structures in functional-logic arrays or single units as depicted schematically in Fig. 14. It has to be noted here at this point that in comparison with other semiconducting material systems, group III-nitride alloys can cover a large range of

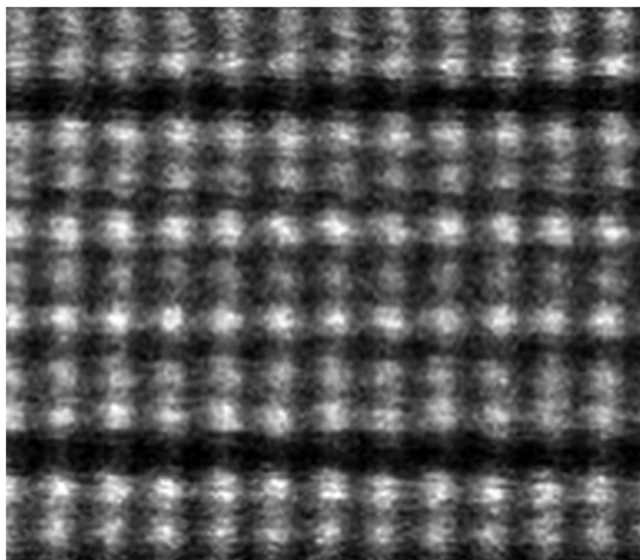


FIG. 13. HRTEM image of the $\text{Ge}_1\text{Sb}_2\text{Te}_4$ nanomembrane used for FPOT devices.^{70,177,178}

emitting wavelengths from UV, through VIS up to the infrared range by appropriate material choice and structural engineering. This is an additional advantage of the group III-nitride alloy system, which can allow the design and combination of nano-LEDs with a large range of optically active media (serving as transistor devices) in addition to their suitability for a radiative and harsh

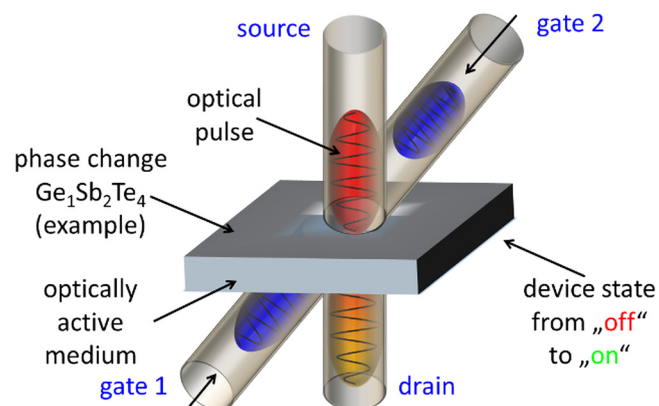


FIG. 14. Schematics of the FPOT device based on the phase-change medium ($\text{Ge}_1\text{Sb}_2\text{Te}_4$ nanomembrane) in the device configuration with two optical gates. Please note that the colored optical pulses are only an artistic illustration and a guide to the eye. The transistor device can be driven by optical pulses from the nano-LED devices (not shown in this picture). Optical pulses (gate input) initiate locally a phase change state manifested as a change in optical transmission from "off" to "on" (opaque vs window) state. To be submitted in 2022 by Mikulics *et al.*

environment. Optical computing units based on such devices would have the potential to significantly increase the stability of satellite communication during solar storms and, hence, decrease losses and reduce operation costs. Besides this application, their usage in a large range of consumer IT as well as for industrial devices and machine products is equally obvious. However, the development of this "new generation" of optical computing architectures requires an intensive interdisciplinary exchange between material device physicists and chemists as well as further progress in correlative characterization methods and tools such as electron microscopy and spectroscopy techniques. Therefore, correlative characterization techniques can open deep insights into material and device science and can help achieve an understanding of new and unique physical effects and phenomena, especially in computational "nano-LED supported" transistor units. The first step, toward a simple optical computational architecture, requiring an "optical input" of nano-LED emission with a spatial, "programmable," singularly addressable optical emission intensity (from an array of nano-LED devices) was already presented.⁶¹ The "programming/conditioning" of the nano-LED's emission intensity itself was achieved/performed with the help of the local laser micro annealing (LMA) process. This serves primarily for the recovery from etching induced defects and in the following for the suppression of the non-radiative mechanism in nano-LED structures [described/mentioned in Sec. II B].⁶¹ There is no doubt that nano-LED structures besides these possible developments described in this section as well as in this whole Perspective study will be a key device component and will play a significant role in the large range of coming and future-emerging applications and technologies.

V. CONCLUSIONS

The last few decades have brought new insights into group III nitride LED structure preparation and rapid development of LED technology down to the nano scale. However, the mass production of nano-LEDs is still challenging and improved, and or alternative technology⁷⁴ are called for and was presented in this Perspective. Due to their unique physical and chemical properties such as chemical inertness, non-toxicity, bio-compatibility, solar and space radiation robustness, long-term device lifetime and reliability, endurance, and high brightness, the range of applications seems to be unlimited.

In this Perspective, future *primary* nano-LED applications were discussed first. Optoelectronic device structures deposited on a metallic substrate were proposed. Such device concepts have the potential to improve the device performance toward higher reliability and unprecedented functionality. By the further miniaturization of the micro-LEDs to the nanoscale, applications in optogenetics and future neuroscience were envisaged, where they could be implanted singularly or in integrated and functional arrays in the, e.g., cortex cerebri to stimulate specific brain regions. Singularly addressable and/or programmable arrays of wavelength tunable sources would allow for novel highly sophisticated and complex sensor device architectures. In addition, it can be expected that the further miniaturization of nano-LEDs for the "far" field emission regime (as well as the decrease in their structure pitch) can be essential for improving the localization of the light sources.

Furthermore, nano-LED devices equipped with appropriate apertures for employment in the “near”-field regime⁶² would play an important role as key components in the development of next generation sensing systems.

Future *advanced* nano-LED applications were presented as well. Computer controlled integrated nano-LED ensembles serving as light emitters for illumination systems in the near-field regime could potentially be applied in emerging super resolution microscopy techniques. Furthermore, a simplification of existing lithographical techniques could significantly contribute to further progress in the scaling of electronic and photonic devices down to the molecular range. A possible solution was introduced based on the application of nano-LED devices in singularly addressable arrays in the near field as well as the far field regime, which could on demand create lithographical patterns flexibly without the need of mask.⁹⁶ Future highly reliable, long-living, and radiation tolerant nano-LED arrays could play an essential role as key components in systems serving for future satellite navigation and autonomous “intelligent” emergency systems for the prevention of satellite collisions. Finally, nano-LEDs are also envisaged to be key components for optical computational techniques, in which nano-LEDs can be combined with a large range of optically active media forming, e.g., transistor devices.¹⁷⁸ Their suitability for a radiative and harsh environment could play a significant role in the large range of coming and future-emerging applications and technologies.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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