

Let There Be Light—With Gallium Nitride: The 2014 Nobel Prize in Physics

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Electrification has been a hallmark of progress and a center of human culture since the invention of the incandescent bulb. The electrification revolution has created and re-invented a multitude of industries. Ironically, the incandescent light-bulb itself seemed immune to these changes, only recently giving way to the solid state technologies. Indeed, it took a truly major development, the invention of the white light-emitting diode (LED), to displace the iconic symbol of electrification. Recognizing this extraordinary shift, the 2014 Nobel Prize in Physics has been awarded to Isamu Akasaki, Hiroshi Amano, and Shuji Nakamura for their pioneering development of gallium nitride (GaN)-based materials and devices, including the blue LED.

Blue emission (>2.7 eV) was long recognized as the missing piece to white light generation. The first wave of activity aimed at producing blue-emitting devices occurred in the late 1960s and early 1970s with notable work at RCA Laboratories and Bell Laboratories.^[1] Despite the promise of the early work, the poor structural quality of GaN on the foreign substrate of choice, sapphire, proved a major challenge. Additionally, while early GaN films were naturally n-type, researchers were unable to produce p-type GaN, necessary for high-efficiency devices.

As the problems with producing efficient GaN-based blue emitters seemed insurmountable, interest waned and efforts focused on more promising compounds, such as zinc selenide (ZnSe). Only a few groups continued to pursue GaN research at all, among them Professor Akasaki at Nagoya University.

In the late 1970s and 1980s there was significant activity in heteroepitaxy of chemically dissimilar materials and materials with large lattice mismatch, respectively. One of the approaches to improve the quality of heteroepitaxial layers was the use of an intermediate nucleation or buffer layer—indeed this was one approach developed for the growth of GaAs on Si.^[2] Adopting these concepts, S. Yoshida and co-workers included an aluminum nitride (AlN) nucleation layer during molecular beam epitaxy (MBE) growth of GaN.^[3] This improved the GaN quality greatly, but not enough for effective blue emission. Using metal organic chemical vapor deposition (MOCVD), Hiroshi Amano, a young researcher working with Professor Akasaki, developed a two-step

growth process that started with a low-temperature (900–1000 °C) AlN nucleation layer^[4] followed by GaN grown at high temperature (950–1060 °C). These films had markedly lower defect densities than any GaN film ever produced before. Akasaki and Amano's two-step growth, with the use of a low-temperature AlN nucleation layer, was a key step in nitride research.

The next challenge for Akasaki and Amano was the realization of efficient p-type doping. LEDs require n-type and p-type regions, and an “active” region between the n-type and p-type regions where the injected electrons and holes recombine to produce light (Figure 1). Magnesium (Mg) was

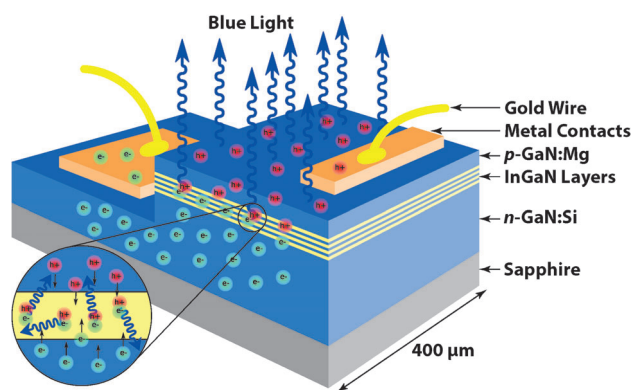


Figure 1. Schematic depicting blue light emission from a GaN-based LED. Carriers are injected from p-type (holes) and n-type (electrons) regions, respectively, and radiatively recombine in the InGaN multi quantum well “active region”.

known to be a possible p-type dopant, but consistently exhibited unacceptably low doping efficiency. In a second major contribution to the development of nitrides, the Nagoya group demonstrated that low energy electron beam irradiation (LEEBI) could “activate” Mg acceptors and effectively render Mg-doped GaN p-type.^[5]

Shuji Nakamura at Nichia Chemical Co. also recognized the potential of GaN for blue emission, despite the prevailing sentiments towards ZnSe as a better candidate. While Akasaki and Amano were working at Nagoya University, he was busy developing and refining a highly original two-flow MOCVD growth system^[6] to allow precise control over GaN growth conditions. Combining his unique growth system with a low-temperature (450–600 °C) GaN buffer layer (a variation

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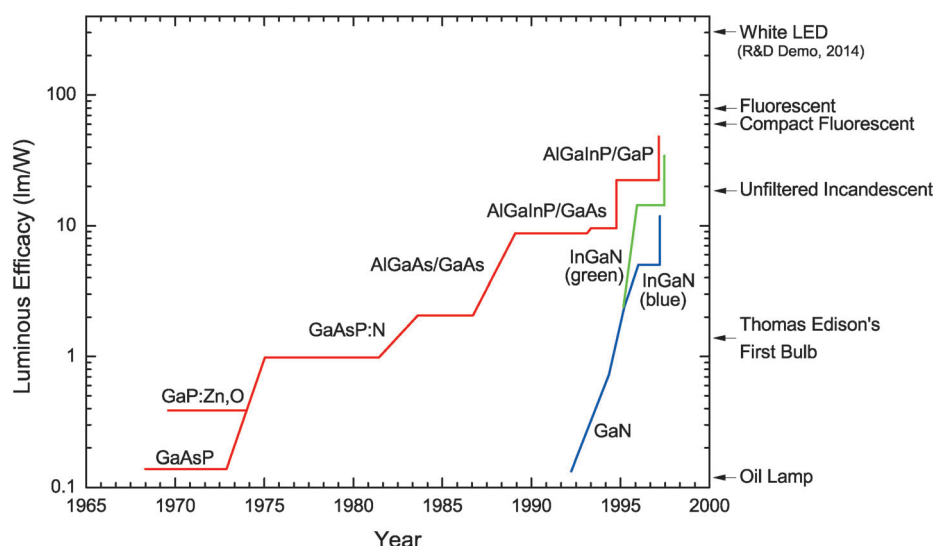


Figure 2. Evolution of luminous efficacy for various technologies. Note the rapid progress in GaN-based blue and green LED performance over the past 20 years.

on the AlN layer used by Akasaki and Amano), Nakamura was able to grow epitaxial GaN of unprecedented quality.^[7] In 1991–92, Nakamura made p-type GaN via a post-growth thermal activation treatment of GaN. Following growth, the Mg-doped material was still highly resistive, but annealing under appropriate conditions caused it to become p-type. This was similar to the LEEBI approach, but more industrially tractable. Remarkably, along with the first report of activating Mg-doped GaN by the Nagoya team, Nakamura also immediately proposed the correct explanation for the effect: hydrogen. Hydrogen was unintentionally incorporated into the GaN lattice during growth, and “passivated” the Mg acceptor atoms. Removing the hydrogen by thermal treatment caused the “activation”. Though novel at the time, low-temperature (GaN and AlN) buffer layers and post-growth thermal Mg dopant activation are now standard industrial processes for GaN-based LED production.

Subsequently, Nakamura demonstrated high brightness p-n homojunction LEDs.^[8] He then developed all individual layer components, particularly those of the alloy of indium nitride (InN) and GaN (referred to as InGaP) necessary for advanced light emission. Armed with these tools, in 1993 he successively demonstrated bright blue and green LEDs.^[9] In 1994, Nakamura led the first research team to produce an efficient white light LED. In late 1995/early 1996, Nakamura demonstrated the first GaN-based laser diode^[10]—which was considered impossible even up to the day of the announcement! Within a decade, violet laser diodes were in mass production for high definition optical disc players.

When Professor Nakamura combined a yellow phosphor with a blue LED to produce white light, he opened the field of solid-state lighting and marked “the beginning of the end of the Edison bulb”. Nowadays, commercial white LEDs have a luminous efficacy in excess of 150 lumen/watt (LPW), much greater than incandescent lamps (12 LPW), or fluorescent lights (80 LPW). Progress has been rapid even with respect to other LED types; in just a few years LPW values for GaN-

based LEDs were on par with those for other, decades-old LED types (Figure 2).

GaN-based LEDs are already ubiquitous in modern technology. They are found as backlights for liquid crystal displays, full color LED displays (using InGaP-based green and blue emitters) and now as the primary light source in solid state lighting. Lighting uses ca. 18% of endpoint electricity use in the U.S. The U.S. Department of Energy estimates that by 2025 solid-state lighting could reduce electricity consumption by 217 TWh (about 6% of the total). New GaN-based lighting technologies include adaptive lighting, laser-based lighting, and communication by optical light, now referred to as “Li-Fi”. Efficient, compact and robust GaN-based LEDs even make it practicable to deploy lighting in the absence of traditional electrification infrastructure by using solar power and batteries in remote locations.

Beyond lighting, nitride-based devices are beginning to penetrate a variety of markets and enabling new applications. Wide electrical bandgaps (> 3.5 eV) created when alloying GaN with AlN can yield emission in the UV range, critical for purification and sterilization. The bandgap can also be tuned for optimum plant growth, or LED-based agriculture. Nitride-based semiconductors also hold great promise for radio frequency (RF) power applications.

Nitrides as a materials class have been vehicles for new solid-state physics. For instance, GaN and the closely related compounds AlN and InN all have the wurzite crystal structure, which is polar. Thus, unlike common semiconductors such as silicon or gallium arsenide, the nitrides have a spontaneous polarization and also show significant strain-induced polarization—referred to as piezoelectric polarization. Nitride-based heterostructures, when grown in the natural *c*-axis orientation, have large fixed polarization charges and internal electric fields. These polarization effects dominate the physics of the nitrides and have opened new areas of research. The growth of high-quality nitride materials coincided with David Vanderbilt’s quantum theory of polar-

ization.^[11] Subsequently, the Nagoya group wrote one of the key early papers that demonstrated the impact of polarization-related electric fields in nitride quantum wells.^[12] Surprisingly, the nitrides, and not classical ferroelectrics, have served as the model system to prove polarization is indeed a bulk property of a solid, and a property that cannot be described classically.

This Nobel Prize especially recognizes the power of perseverance and the fortitude necessary to attempt what many might have judged a hopeless task. No doubt similar was said about the concept of “light without smoke”. Just as the original lightbulb marked the dawn of a new age, the societal and scientific impacts of these new discoveries will likely continue to unfold for some time to come.

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- [1] a) H. P. Maruska, J. J. Tieljen, *Appl. Phys. Lett.* **1969**, *15*, 327–329; b) H. P. Maruska, D. A. Stevenson, J. I. Pankove, *Appl. Phys. Lett.* **1973**, *22*, 303–305; c) M. Ilegems, R. Dingle, R. A. Logan, *J. Appl. Phys.* **1972**, *43*, 3797–3800.
- [2] B. Y. Tsaur, M. W. Geis, J. C. C. Fan, R. P. Gale, *Appl. Phys. Lett.* **1981**, *38*, 779–781.
- [3] S. Yoshida, S. Misawa, S. Gonda, *Appl. Phys. Lett.* **1983**, *42*, 427–429.
- [4] H. Amano, N. Sawaki, I. Akasaki, Y. Toyoda, *Appl. Phys. Lett.* **1986**, *48*, 353–355.
- [5] H. Amano, M. Kito, K. Hiramatsu, I. Akasaki, *Jpn. J. Appl. Phys.* **1989**, *28*, L2112–L2114.
- [6] S. Nakamura, Y. Harada, M. Senoh, *Appl. Phys. Lett.* **1991**, *58*, 2021–2023.
- [7] S. Nakamura, *Jpn. J. Appl. Phys.* **1991**, *30*, L1705–L1707.
- [8] S. Nakamura, T. Mukai, M. Senoh, *Jpn. J. Appl. Phys.* **1991**, *30*, L1998–L2001.
- [9] S. Nakamura, M. Senoh, T. Mukai, *Appl. Phys. Lett.* **1993**, *62*, 2390–2392.
- [10] S. Nakamura, M. Senoh, S. Nagahama, N. Iwasa, T. Yamada, T. Matsushita, H. Kiyoku, Y. Sugimoto, *Jpn. J. Appl. Phys.* **1996**, *35*, L74–L76.
- [11] a) R. D. King-Smith, D. Vanderbilt, *Phys. Rev. B* **1993**, *47*, 1651–1654; b) F. Bernardini, V. Fiorentini, D. Vanderbilt, *Phys. Rev. B* **1997**, *56*, R10024–R10027.
- [12] T. Takeuchi, S. Sota, M. Katsuragawa, M. Komori, H. Takeuchi, H. Amano, I. Akasaki, *Jpn. J. Appl. Phys.* **1997**, *36*, L382–L385.
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