Takeda Award 2002 Achievement Facts Sheet

Techno-Entrepreneurial Achievements for Social/Economic Well-Being

Technical Achievement:
The Development of Blue Light Emitting Semiconductor Devices
- Development of the blue light emitting diode and laser diode is the final link in completing the light spectrum for semiconductor devices.

Awardees:
Isamu Akasaki (Meijo University) and Hiroshi Amano (Meijo University)
Shuji Nakamura (University of California, Santa Barbara)

One half of the prize is awarded jointly to Isamu Akasaki and Hiroshi Amano, and the other half is awarded to Shuji Nakamura.

(Awardees are listed in alphabetical order.)

Executive summary

The achievement honored is the development of blue light emitting semiconductor devices, the blue light emitting diode (LED) and the blue laser diode (LD), which complete the light spectrum for semiconductor devices.

Light emitting devices have many useful applications in daily life. The completion of the light spectrum, enabled by the invention of the blue LED, has resulted in the development of large, outdoor, full-color displays on building walls, which are clear and easy to see even in bright sunlight. Traffic signals have also been developed using semiconductor LEDs. Compared to conventional incandescent bulb-based traffic signals, these new traffic signals have improved visibility, lower electric power consumption and a longer lifespan. Thirdly, combining the blue LED with fluorescent material has resulted in the creation of white LEDs, which have been used to backlight displays in mobile phones. In the future, it is believed that the white LED will replace conventional lamps for lighting homes and offices. The replacement of conventional lamps with LEDs is expected to reduce electric power consumption, due to the high efficiency of LED light emission, and to reduce waste because of the longer lifespan of LEDs.

The blue LD will also enable a new generation of DVDs and CDs with more than a six-fold increase in optical storage capacity. Photographs and moving pictures, including
animation, can be easily recorded, manipulated, stored, and carried as digital data using blue LD-based devices. Image projectors using laser light are also expected.

The development of blue light emitting semiconductor devices involved many challenges. Akasaki and Amano, and Nakamura, respectively, succeeded in overcoming these challenges through highly creative approaches within the same time frame.

Akasaki was confident that gallium nitride (GaN) would be the best material with which to make blue light emitting semiconductor devices, although the majority of researchers were using zinc selenide (ZnSe). Akasaki and Amano fabricated GaN film with good crystal quality and uniform thickness on an aluminum nitride (AlN) buffer layer grown on a sapphire substrate. Then, they fabricated a p-type GaN layer with high conductivity by using electron beam irradiation on a p-type impurity doped GaN thin film. In 1989, they developed a p-n junction blue LED in their laboratory. Regarding blue LD development, they observed emission of strong light with a narrow bandwidth, and it was reported in November 1995.

Nakamura also chose to work with GaN, rather than with ZnSe used by the majority of researchers. He invented film deposition equipment with a unique two-flow method, and fabricated GaN film with good crystal quality and uniform thickness. He achieved high conductivity, p-type GaN by heat treatment in the atmosphere without hydrogen. He developed a high brightness blue LED of double heterostructure using In-doped GaN layer in 1993. Regarding blue LD development, he succeeded in achieving laser oscillation, introducing a multi-quantum well structure with several tens of In GaN layers and a blocking layer, and reported this in January 1996. Based on these achievements, the blue LED was introduced in 1993 and the blue LD was introduced in 1999. Those were the first blue LED and blue LD commercialized products in the world.

Akasaki and Amano and Nakamura overcame difficult challenges that required a creative approach. Moreover, they contributed to the commercialization of blue LD and LED-based devices. The Takeda Award 2002 for Social/Economic Well-Being is awarded to this techno-entrepreneurial achievement that embodies engineering intellect and knowledge, and enables many applications in human life that will expand the wealth, richness, and happiness of people.
Achievement and creativity

1. Role of light emitting semiconductor device

The amount of digital data used in homes and in offices is increasing in our rapidly developing information society. Light emitting semiconductor devices are key devices for information display and information storage. A “light emitting semiconductor device” is a generic term that refers to electronic devices that use both light emitting diodes (LED) and laser diodes (LD).

Red and green light emitting semiconductor devices were developed in the 1970s. For example, red and green LEDs were used as indicator lights for displays in various electronic products and in signs. The red LD was widely used for optical fiber networks and light pickup in optical storage devices. By completing the light spectrum with the development of blue light emitting semiconductor devices, new applications such as bright, full-color displays and brighter, higher energy efficiency lighting became possible. The blue LD has a short wavelength and can emit a narrow light beam. Applications in high-density optical storage devices, such as the next generation of CDs/DVDs, are expected. The development of blue light emitting devices was highly desired by many electronics companies.

2. Light emitting devices

2.1. Light emitting diode

A LED is a compound semiconductor device that emits visible or infrared light when an electric current passes through it. It contains a p-n junction with a direct transition band structure. Injected electrons and holes recombine near the p-n junction area to emit light. In order to increase light emitting efficiency, a double heterojunction is widely used, which is made by covering an active layer by p-type and n-type semiconductor layers that have larger band gaps. The wavelength of the emitted light is determined by the band gap of the semiconductor material. Gallium arsenide (GaAs) based material and gallium phosphide (GaP) based material are used to produce red and green LEDs. In the 1970s, red and green LEDs were commercialized and used for a variety of applications, such as indicators and signs.

2.2. Laser diode

A laser diode is a light emitting diode that uses an optical cavity to amplify the light emitted from the energy band gap that exists in semiconductors. For a laser to work, there needs to be a population inversion between the conduction and valence band of the semiconductors,
which is created by applying an electrical current to the p-n junction. First generation LDs used homogenous p-n junctions and the same material as LEDs. These were relatively inefficient and required a high current density to achieve lasing. Laser operation was limited only to pulse mode at low temperatures. Today, double heterojunction lasers are widely used. They produce light more efficiently, have a lower current density threshold for operation, and can provide stable and continuous laser oscillation at room temperature. Since the 1980s, red and infrared LD have been used for optical fiber communications, as read/write devices for optical storage, and in other applications.

2.3. Blue light emitting semiconductor device

Silicon carbide (SiC), zinc selenide (ZnSe) and gallium nitride (GaN) were all potential semiconductor materials for the development of blue light emitting semiconductor devices, based on the width of their energy band gaps. However, SiC has an indirect transition band structure, so it was not a suitable candidate. ZnSe was considered a good candidate and active research was undertaken from the 1970s. Although LEDs and LDs using ZnSe were developed in the laboratory, commercial products could not be developed because of the short lifespan caused by, for example, the electrode metal migration to the light emitting semiconductor layer.

Research on GaN also started in the 1970s. There were no good substrates possessing a lattice constant near that of GaN. A sapphire substrate with a 16 percent different lattice constant had to be used, and it was almost impossible to fabricate a GaN thin film with good crystal quality and uniform thickness. Even if such a high quality GaN thin film could be fabricated, it was also almost impossible to make p-type GaN film. Considering these challenges, many researchers thought it was impossible to develop a blue light-emitting device using GaN and they abandoned this line of research.

3. GaN-based blue light emitting device development by Akasaki and Amano

3.1. Selection of GaN

Akasaki undertook research to develop blue light emitting devices using GaN in the 1970s. He adopted a molecular beam epitaxy (MBE) method that can bond each molecule to the right place, in addition to the previously used hydride vapor phase epitaxy (HVPE) method. He succeeded in fabricating a GaN single crystal film using MBE method. In 1981, he achieved a light emission from a metal-insulator semiconductor structure with 0.12% efficiency using HVPE method. However, he could not develop a thin film with good thickness uniformity. Also, he could not produce a p-type GaN film. Even on this macroscopically inferior quality crystal, he noticed a strong light emission from the very small spot of film that had thickness...
uniformity. This observation convinced him that GaN was, indeed, the material with which to develop a blue light-emitting device, despite the fact that many other researchers had abandoned GaN-based research. In 1981, Akasaki moved to Nagoya University to continue his research. In 1982, Amano joined Akasaki’s laboratory and began his research work.

3.2. GaN thin film formation with superior uniformity

Akasaki had recognized the limitations of the HVPE method and the MBE method for GaN thin film fabrication. The problem with the HVPE method is that the crystallinity of thin film is bad under high film fabrication speed. On the other hand, the problems associated with the MBE method include slow film fabrication speed and difficulty in attaining a stoichiometric composition because nitrogen is easily pulled out by its high vapor pressure in an ultra-high vacuum ambient atmosphere. He decided to use a metal organic chemical vapor deposition method (MOCVD method) that brought an appropriate film fabrication speed in the same temperature region for each film. He selected a sapphire substrate, which can be used at a GaN film fabrication temperature of over 1,000 °C in MOCVD method, and has a lattice symmetry near to that of GaN. However, because the lattice constant difference was 16 percent, it was a difficult challenge to fabricate hetero epitaxial thin film with good crystal quality and thickness uniformity.

Akasaki and Amano designed and developed the MOCVD equipment by themselves, because MOCVD fabrication equipment that could be used for GaN was not available at that time. They tried to identify the best fabrication conditions repeating experiments with various combinations of substrate temperature, vacuum pressure, material gas feed rate, inactive gas feed rate, fabrication time length, and so on. They performed over 1,500 experiments in two years, but they could not fabricate thin film with good crystal quality and thickness uniformity.

They conceived of locating a buffer layer fabricated at low temperature between the sapphire substrate and the GaN thin film after thorough investigation of their experimental results. They selected GaN, aluminum nitride (AlN), SiC, and zinc oxide as the candidates. They succeeded in fabricating GaN thin film with good crystal quality and thickness uniformity using AlN as a buffer layer in 1986\(^2\). Good crystal quality of the film was confirmed using photoluminescence, x-ray diffraction, Hall effect measurement, transmission electron microscope and so on. From the result of Hall effect measurement, the electron mobility at room temperature was calculated to be improved from 50cm\(^2\)/V\(\cdot\)s to 450cm\(^2\)/V\(\cdot\)s\(^3\). Moreover, they succeeded in realizing the n-type thin film with thickness uniformity and good conductivity\(^4\).

This success was achieved not just through one experiment, as above, but instead required a tremendous number of experiments to find the optimized conditions that made it a great breakthrough.
3.3. P-type GaN layer fabrication

GaN thin films usually became n-type semiconductors, because they contained smaller than stoichiometric numbers of nitrogen. Attempts were made to make a p-type layer using acceptor-doping material. This was also a big hurdle to making a GaN light emitting device. Akasaki and Amano initially used zinc as an acceptor dopant and tried to fabricate p-type GaN, but they did not achieve success. After they used magnesium as a dopant because of its higher electron affinity, they still did not achieve success. In 1988, Amano found that cathode luminescence light intensity increases with electron beam irradiation in other experiments to measure cathode luminescence of acceptor doped GaN. He thought from this result that the electrical and optical characteristics of acceptor doped GaN thin film were changed by electron beam irradiation. Based on this, they measured the electrical characteristics of 10 kV electron irradiated Mg doped GaN thin film and found that the resistivity decreased by a factor of 10,000 to 35 Ohm-cm, and confirmed that GaN film is clearly p-type by its Hall effect measurement. Its hole mobility was 8 cm$^2$/V·s. They made a p-n junction at the same time and measured current rising in the forward direction on the current-voltage curve and light emission -- both are characteristics of p-n junction. This p-type layer fabrication method by electron beam irradiation was the historic first fabrication of p-type GaN. They made an important step toward developing a blue light emitting semiconductor device by inventing p-type GaN fabrication and GaN layer fabrication method.

3.4. Development of the blue LED

From 1987, Akasaki and Amano led blue LED development at Toyoda Gosei Co. Ltd., which received funding from the Japan Science and Technology Corporation. In 1992, they succeeded in developing a bright blue LED with 1 percent light emitting efficiency. Toyoda Gosei announced commercial production of blue LEDs in 1995.

3.5. Development of the blue LD

Akasaki and Amano also undertook the challenges of developing a blue LD. In 1990, they succeeded in measuring narrow bandwidth 374nm stimulated emission at room temperature with high density electron-hole pairs created by radiating 337nm nitrogen laser beam on a GaN thin film, which was fabricated on a low temperature buffer layer with good thickness uniformity. Then they fabricated a multi-quantum well structure device and, decreasing the threshold power density, succeeded in measuring strong emission of 3nm half bandwidth on 1.0 kA/cm injection current density, which they reported in Japanese Journal of Applied Physics in
November 1995\(^6\). They reported achieving a laser oscillation of 405nm wavelength in June 1996\(^9\).

4. GaN-based blue light emitting device development by Nakamura

4.1. Selection of GaN

Shuji Nakamura began development of a blue light emitting semiconductor device at Nichia Chemical Industries, Ltd., in 1988. In order to undertake this research, he had to obtain the consent of the president of Nichia Chemical Industries through direct negotiation, something that was extraordinary for a researcher in a Japanese company to do. Material selection was an important issue. Since many researchers were pursuing ZnSe-based blue LED research and would patent their manufacturing technology processes, Nakamura decided it would be difficult to develop a novel technology. Therefore, he selected GaN as the focus of his research, which was only being pursued by a minority of researchers. Nakamura had the confidence and engineering creativity to solve the technical problems stemming from GaN-based research.

4.2. GaN thin film formation with superior uniformity

Nakamura began his experiment to make GaN thin film using standard equipment with a well-known substrate material, sapphire, and a well-known method, MOCVD. However, GaN thin film with good uniformity had yet to be produced. Nakamura thought that the gas flow scheme was the key issue to improving film uniformity. This approach is different from that taken by Akasaki and Amano, who introduced the low-temperature buffer layer. Nakamura thought over many kinds of gas flow schemes that were different from the conventional layer flow. In the conventional gas flow, material gases, including Ga compound and nitrogen, were supplied in parallel to the substrate surface as the layer flow. This conventional scheme was considered to be rational but had achieved only inferior uniformity and less than stoichiometric nitrogen content. Nakamura repeatedly modified the MOCVD equipment by himself to change the gas flow mode, and performed experiments over 500 times in a short period.

Finally, he found the most appropriate method where the main material gases, including Ga compound, flow in parallel to the substrate, and nitrogen and hydrogen are supplied vertically from the upper side to the substrate surface. In 1991, he succeeded in fabricating a GaN thin film with superior uniformity using this method, which he named the “two-flow method”\(^10\). The electron mobility calculated from the Hall effect measurement with this GaN thin film was 200 cm\(^2\)/V·s, which was much larger than the conventional result of 90 cm\(^2\)/V·s. In an additional experiment, in which the low temperature buffer layer was introduced as well as the two-flow method, the resultant GaN thin film showed 500 cm\(^2\)/V·s, which is a satisfactory figure for
light emitting device fabrication. Moreover, in 1992, he achieved high quality InGaN ternary compound thin film by the two-flow method. This film was necessary to optimize the emitted light wavelength and to improve the emission efficiency\(^{11}\).

Nakamura reexamined the material for the low-temperature buffer layer and found that not only AlN but also GaN itself was suitable\(^{12}\). In the case of GaN, the material gas change is not needed between the buffer layer and the active layer. This is practically important and beneficial for mass production technology.

### 4.3. P-type GaN layer fabrication

In order to fabricate the p-type GaN layer, Nakamura investigated another more practical method than that with electron irradiation studied by Akasaki and Amano. Heat treatment, using a specimen doped with magnesium as the acceptor impurity, had been studied but had proven unsuccessful. Nakamura reexamined the heat treatment effect and found that a hydrogen-free atmosphere was essential to activate the doped magnesium and get the p-type layer. He clarified the mechanism of p-type formation, proposing a model based on the connection between hydrogen atoms and acceptor impurities\(^{13}\).

Using this method, in 1992, the specific resistance of heat-treated GaN thin film was improved to 2 Ohm-cm – smaller than usual by more than a factor of 100,000 – and the superior p-type layer was successfully obtained where the hole mobility by Hall measurement was 10 cm\(^2\) V\(^{-1}\) s\(^{-1}\)\(^{14}\).

### 4.4. Development of blue LED

Using the above-mentioned technologies, Nakamura fabricated p-n homo-junction blue LEDs with 0.18 % light emitting efficiency and the double heterostructure type with 0.22 % light emitting efficiency in 1992. The latter one was improved to 2.7 % light emitting efficiency using the above-mentioned InGaN thin film as the light emitting layer, in 1993\(^{15}\). Based on these results, Nichia Chemical Industries delivered the world’s first blue LEDs in 1993. Moreover, the quantum-well structure was adopted to develop the high brightness blue LED with 9.2 % light emitting efficiency\(^{16}\).

Nakamura investigated the InGaN light emitting layer in detail. Before that time, to achieve a long light emission lifespan, the maximum allowable number of crystal defects was considered to be under 10\(^3\) cm\(^{-2}\). Although there were many crystal defects – about 10\(^{10}\) cm\(^{-2}\) – in the InGaN film of LED, the lifespan of this LED was longer than 100,000 hours. The reason for this long lifespan with that level of defects is as follows. The distribution of indium atoms doped in GaN film is not uniform but fluctuates slightly, and consequently the potential for
electrons in the crystal is not uniform but varies locally. Injected electrons are trapped in this localized potential and recombined with holes to emit the light without being captured by the crystal defect region. This kind of localized potential was a thoroughly new phenomenon not found in any previous crystal material and found first in this ternary mixed crystal InGaN. The research project then started intentionally to design and to make this kind of localization in the crystal.

4.5. Development of blue LD

Based on these technologies for GaN-based blue LEDs, Nakamura succeeded in achieving high power pulse oscillation of the GaN-based blue LD with an InGaN multi-quantum well structure and clad layers. This was reported in the January 1996 issue of *Japanese Journal of Applied Physics*<sup>17</sup>. This paper attracted remarkable attention from researchers all over the world, and the number of resulting citations was nearly 160 by 1997.

The key step for achieving this LD oscillation was introducing InGaN twenty cycles multi-quantum well structure for the light emitting layer and AlGaN film for blocking layer<sup>18</sup>.

In the case of LDs, compared to LEDs, the number of defects had to be decreased still more to obtain a long lifespan, since the number of injected electrons in LDs is larger. Nakamura adopted the Epitaxial Lateral Overgrowth (ELOG) method<sup>19</sup>. In 1998, he succeeded in decreasing the defects from $10^{10} \text{cm}^{-2}$ to $10^7 \text{cm}^{-2}$ and achieved 290 hours of continuous oscillation at room temperature, from which the oscillating lifespan was estimated to be 10 thousand hours. In 1999, Nichia Chemical Industries delivered the world’s first blue LD<sup>20</sup>.

5. Repercussion effects

Completing the light spectrum for semiconductor devices has been accomplished through the development of the blue LED and LD, which, together with red and green devices, enables various kinds of applications.

GaN is now used as a key material for producing ultraviolet, blue, green and yellow light emitting devices by changing the indium doping content. Of special importance, green LEDs with higher purity and higher efficiency than that of GaP-based LEDs have already been developed as commercial products using indium doped GaN.

The development of the blue LD has enabled the development of the next generation of large-capacity optical storage discs. Nine electronics manufacturing companies from Japan, Korea, and Europe recently announced the specifications for the next generation of CDs/DVDs, known as the “Blu-ray Disc,” in February 2002. The blue LD will enable 12 cm discs to record, rewrite, and play back 27 gigabytes (GB) of data — six times more information than on a
conventional disc. This will allow for the recording of all kinds of digital content (audio, video, television, photographs, etc.) delivered to homes and offices through digital broadcasting and broadband communication. The recording of two hours of digital high-definition video and thirteen hours of standard TV broadcasting will be possible with the Blu-ray Disc. In addition, double-sided discs or the enlargement of the size of the disc is also possible, enabling one disc to store 50 to 100 GB data. One terabyte recording becomes possible using multiple discs. Large projection displays can be designed using all three colors of LDs.

Using blue LEDs in conjunction with red and green LEDs, large outdoor full-color displays, which are clear and easy to see even in bright sunlight, have been developed and displayed on buildings.

LEDs have also been increasingly used to replace incandescent bulbs in traffic signals. LEDs have many advantages over conventional lamps. First, the light provided by LEDs is clearer, especially in direct sunlight. Second, the lifespan of LEDs is over eight years, much longer than that of incandescent lamps with average lifespans of six months to one year. Thirdly, LEDs reduce electric power consumption by one-fourth. The Department of Transportation in California replaced 200,000 traffic signals with LEDs during the most recent energy crisis. There are about one million traffic signals for cars and 770,000 for pedestrians in Japan. In about 8,000 of these signals, incandescent bulbs have been replaced with LEDs at present. It is estimated that when all traffic signals for pedestrians are replaced with LEDs, electric power consumption will be greatly reduced and carbon dioxide gas generation will fall by 110,000 tons per year\(^2\).\(^1\)

There are two types of white LED. One uses the blue LED combined with a yellow fluorescent substance. The other consists of blue, green and red LEDs. Both types of white LED were enabled by the development of the blue LED\(^2\).\(^2\) In the near future, the efficiency of the white LED should be improved to generate five times better efficiency than the incandescent lamp and the same efficiency as that of fluorescent light (50%). The lifespan of the white LED is over 8 years, which is much longer than the one-year durability of the white incandescent lamp and the fluorescent lamp. When the use of white LED lightning is expanded to all kinds of living spaces, it is expected to contribute greatly to the reduction of electric power consumption through the high efficiency of its light emission, and to the reduction of waste through its longer life. White LED lighting is flexible in terms of shape (flat, curved, spherical) and color (changeable tone by controlling the three colored LEDs respectively). The flexibility of this type of lighting is ideal and contributes to improving living environments.

The total market size of blue and white LEDs was estimated to be 90 billion yen in 2001 and is expected to grow to 230 billion yen in 2006\(^2\).\(^3\) When the white LED lightning replaces the incandescent bulb and the fluorescent lamp, the market size is expected to grow to more
than one trillion yen. As for the blue LD, the market size of the market for blue LDs will benefit greatly from the development and commercialization of the next-generation, high-density CD/DVD, growing to more than several tens of billion yen\textsuperscript{24)}.

6. Conclusion

The development of the blue light emitting diode and blue laser has completed the light spectrum for semiconductor devices. Blue light emitting semiconductor devices are expected to enable a wide range of electronics-related applications (large and bright full-color displays, traffic control signals and lightning lamps). Of special importance, white LEDs are expected to contribute to the reduction of electric power consumption through a high level of light emission efficiency and to the reduction of waste through its longer life.

Akasaki and Amano, and Nakamura, respectively, succeeded in developing blue light emitting semiconductor devices through highly creative approaches, which enable many applications in human life and extend the wealth, richness, and happiness of people.
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1995 International Symposium on Compound Semiconductors (ISCS) Award
1995 Heinrich Welker Gold Medal
1996 IEEE/LEOS Engineering Achievement Award (with Drs. Amano and Nakamura)
1997 Medal with Purple Ribbon, Government of Japan
1998 Laudise Prize, The International Organization for Crystal Growth (IOCG)
1998 C&C Prize (with Dr. Nakamura)
1998 IEEE Jack A. Morton Award (with Dr. Nakamura)
1998 British Rank Prize (with Drs. Nakamura and Amano)
1999 Electrochemical Society (ECS) Solid State Science and Technology Award
1999 Honoris Causa of University of Montpellier
2000 Toray Science and Technology Prize
2001 Asahi Prize (with Dr. Nakamura)
2001 Honoris Causa of Linköping University, Sweden
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He is honored with many other awards.

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Honors:
1994 Optoelectronics Conference Special Award (with Dr. Akasaki)
1996 IEEE/LEOS Engineering Achievement Award (with Drs. Akasaki and Amano)
1998 Japanese Journal of Applied Physics Award for the best review paper (with Dr. Akasaki)
1998 British Rank Prize (with Drs. Nakamura and Akasaki)
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2000-present Professor, University of California at Santa Barbara

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1996 IEEE Lasers and Electro-Optics Society (LEOS) Engineering Achievement Award (with Drs. Akasaki and Amano)
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1998 C&C Award (with Dr. Akasaki)
1998 British Rank Prize (with Drs. Akasaki and Amano)
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