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► **To cite this version:**

Christophe Lécuyer, Takahiro Ueyama. The Logics of Materials Innovation The Case of Gallium Nitride and Blue Light Emitting Diodes. *Historical Studies in the Natural Sciences*, 2013, 43 (3), pp.243-280. 10.1525/hsns.2013.43.3.243 . hal-01362207

**HAL Id: hal-01362207**

**<https://hal.sorbonne-universite.fr/hal-01362207>**

Submitted on 8 Sep 2016

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## The Logics of Materials Innovation: The Case of Gallium Nitride and Blue Light Emitting Diodes

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### ABSTRACT

This article examines the rise of gallium nitride as a major semiconductor material at the center of a new industry, solid-state lighting. It argues that the development of gallium nitride was shaped by the interplay of three contextual logics: material logic (the materiality of substances, tools, and fabrication techniques); market logic (the needs, demands, and interests of intended users); and competitive logic (the competitive tensions among laboratories, firms, and nations). For nearly forty years, chemists, physicists, and engineers in the United States and Japan struggled with the persistent material challenges presented by gallium nitride to meet the needs of potential markets in lighting and consumer electronics. Competition among firms and the technological and economic rivalry between the U.S. and Japan led to significant material developments and to the shaping of gallium nitride into a critical material for the manufacture of light emitting diodes. This article is intended to contribute to recent interest in the history of advanced materials and to the larger question of the determinants of innovation in technoscience.

KEY WORDS: compound semiconductor, gallium nitride, innovation, light emitting diode, logic, material, solid-state lighting

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The following abbreviations are used: ARPA, Advanced Research Projects Agency; CCL, Christophe Lécuycer private collection, Budapest, Hungary; HVPE, hydride vapor phase epitaxy; LED, light emitting diode; MBE, molecular beam epitaxy; MIS, metal-insulator-semiconductor; MITI, Japanese Ministry of International Trade and Industry; MOCVD, metal-organic chemical vapor deposition; MRIT, Matsushita Research Institute Tokyo, Inc.; NASA, National Aeronautics and Space Administration; NTT, Nippon Telegraph & Telephone Corp.; RCA, Radio Corporation of America.

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*Historical Studies in the Natural Sciences*, Vol. 43, Number 3, pps. 243–280. ISSN 1939-1811, electronic ISSN 1939-182X. © 2013 by the Regents of the University of California. All rights reserved. Please direct all requests for permission to photocopy or reproduce article content through the University of California Press's Rights and Permissions website, <http://www.ucpressjournals.com/reprintinfo.asp>. DOI: 10.1525/hsns.2013.43.3.243.

In June 2008, the U.S. Department of Energy announced the Bright Tomorrow Lighting Prize (L Prize), to foster the development of new technologies aimed at replacing incandescent light bulbs. The Department of Energy promised \$10 million in cash and future procurement contracts to the corporation able to manufacture a solid-state device using significantly less energy than incandescent light bulbs. A little more than a year later, Philips Lighting North America, the subsidiary of the Dutch electronic giant Philips, submitted a lamp based on light emitting diode (LED) technology to the Department of Energy. After a year and a half of testing, the DoE concluded that Philips's solid-state lamp met the prize requirements: It consumed less than ten watts and produced a light output that was equivalent to the output of a sixty-watt incandescent bulb. The resulting energy saving was on the order of eighty-three percent. In August 2011, the DoE awarded the L Prize to Philips and thirty utilities serving more than a hundred million customers in the United States and Canada agreed to promote Philips's solid-state lamp in their respective markets.<sup>1</sup>

The lamp submitted by Philips was based on more than forty years of research and development of a little-known material, gallium nitride. Gallium nitride belongs to a class of electronic materials: compound semiconductors. Compound semiconductors, which are man-made (no compound semiconductors exist in nature), are semiconductors made of two or more chemical elements. In the case of gallium nitride, these elements are gallium and nitrogen. In its crystalline form, gallium nitride is very hard and transparent. It also has unique characteristics. As researchers have painstakingly found out over the last four decades, one of the particularities of gallium nitride is that electrons move especially fast within it. Gallium nitride also withstands higher voltages and can emit more light—blue light in this case—than other compound semiconductors.

But gallium nitride is also a recalcitrant material. It is especially difficult to grow single crystals (nearly perfect crystals) of gallium nitride. It is also very difficult to alter the material's electronic and light-emitting properties. Four decades after the creation of the first crystals of gallium nitride, basic techniques for growing them are still in flux. Critical problems related to their fabrication, such as the nature of the substrate on which to grow gallium nitride films, remain to be solved.<sup>2</sup> In spite of these persistent uncertainties,

1. For the L Prize, see C. Edward Christy, "Bright Tomorrow Lighting Competition," *Federal Register* 73:122 (24 Jun 2008), p. 35680, at <http://www.lightingprize.org/> (accessed 20 Oct 2011).

2. The method that is the most commonly used is the deposition of a gallium nitride film on top of a sapphire crystal. Sapphire and gallium nitride do not have the same crystalline structure, however, which leads to the creation of cracks or dislocations within the gallium nitride crystals.

gallium nitride is generally viewed as a critical semiconductor material, second in importance to silicon. It is widely used in light emitting diodes and increasingly so in transistors as well. Some observers predict that gallium nitride may one day replace silicon in the manufacture of microprocessors and other integrated circuits.<sup>3</sup>

How did gallium nitride become a critical semiconductor material? What were the forces that presided over its creation and made its rise possible? In his book *Brilliant!: Shuji Nakamura and the Revolution of Solid State Lighting*, Bob Johnstone pointed to the role of Nakamura, a Japanese engineer, in the development of gallium nitride crystals and the making of blue and white LEDs (gallium nitride and blue LEDs are inextricably linked; improvements in gallium nitride crystals lead to improvements in light emission). According to Johnstone, Nakamura almost single-handedly created gallium nitride crystals and blue LED technology at Nichia Chemical Industries, a medium-sized chemical firm based in Japan. In this heroic account, Nakamura overcame all odds in innovating gallium nitride crystals and blue LEDs, including numerous roadblocks coming from the firm's upper management.<sup>4</sup>

Closer attention to the historical record reveals a much more complex and interesting story. Nakamura was not the only innovator of gallium nitride and blue LED technology—far from it. Others such as Herbert Maruska and Jacques Pankove at RCA and Isamu Akasaki and Hiroshi Amano at Nagoya University played critical roles as well. As a matter of fact, Nakamura's main contribution to gallium nitride technology came from his commercialization of materials and material practices originally created by Maruska and Akasaki. For example, Nakamura transformed laboratory techniques developed at Nagoya University into mass production manufacturing processes. It is interesting to note as well that Nakamura and his collaborators at Nichia were not the only ones to introduce gallium nitride to the market. A group of engineers at Toyoda Gosei, another Japanese firm, commercialized gallium nitride and blue LEDs around the same time and in direct competition with Nakamura and Nichia Chemical. In other words, Nakamura, who is portrayed by Johnstone and others as the inventor of the blue LED, was in fact one of the

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Other technologists currently employ silicon or silicon carbide substrates to fabricate gallium nitride crystals.

3. Robert Service, "Nitrides Race Beyond the Light," *Science* 237 (2010): 1598–99.

4. Bob Johnstone, *Brilliant! Shuji Nakamura and the Revolution of Solid State Lighting* (Amherst, MA: Prometheus Books, 2007).

many engineers who commercialized materials and devices innovated at RCA and Nagoya University.<sup>5</sup>

Thus the story of gallium nitride opens a window into the study of technology transfer and university-industry relations. The case of gallium nitride also throws light onto materials innovation, a form of innovation that has received little attention among historians of technology.<sup>6</sup> One of the authors of this article (Christophe Lécuyer), in collaboration with David C. Brock, recently developed an analytical framework to account for technological innovation, especially innovation in materials and manufacturing processes. This framework distinguishes different types of contextual challenges that shape the creation of new materials and manufacturing processes: the materiality of substances, tools, and fabrication techniques (referred to as “material logic”); the needs, demands, and interests of intended customers (“market logic”); and the competitive tensions among laboratories, firms, and nations (“competitive logic”). These material, market, and competitive logics are not determinative, in the sense that they do not lead to necessary outcomes. But they are particularly stable over time and provide powerful resources and constraints to innovators and their patrons. When combined, they define chains of potential events that are actualized by the will, passions, and efforts of scientists and technologists—in other words, their emotional and professional commitments. The contextual logics are situated within larger shaping fields, such as the Cold War and transnational capitalism. The shaping fields give relevance to the materials themselves and to materials innovation. They also construct the competitive and market pressures that orient innovative activity.<sup>7</sup>

5. Nakamura is generally seen as “the father of the blue LED” because of an energetic public relations campaign mounted by Nichia in the 1990s. The corporation was then embroiled in a bitter patent war with Toyoda Gosei and needed to establish its precedence in the development of blue LEDs. Nakamura later got extra publicity from his legal wrangling with Nichia over rights to the processes he had developed at the firm.

6. For recent exceptions, see Ursula Klein and E. D. Spary, eds., *Materials and Expertise in Early Modern Europe: Between Market and Laboratory* (Chicago: University of Chicago Press, 2010), and Christophe Lécuyer and David C. Brock, “The Materiality of Microelectronics,” *History and Technology* 22 (2006): 301–25. For an insightful discussion of materials innovation, see Dominique Pestre, “Louis Néel, Le Magnétisme et Grenoble: Récit de la création d’un empire physicien dans la province française 1950–1965,” *Cahiers pour l’Histoire du CNRS* 8 (1990): 1–188.

7. For a discussion of the logics of innovation, see Christophe Lécuyer and David C. Brock, *Makers of the Microchip: A Documentary History of Fairchild Semiconductor* (Cambridge, MA: MIT Press, 2010) and David C. Brock and Christophe Lécuyer, “Digital Foundations: The Making of Silicon Gate Manufacturing Technology,” *Technology and Culture* 53 (2012): 561–97. The concept of contextual logics draws on the writings of Andrew Pickering on the reciprocal

The development of gallium nitride was shaped by the interplay of material, market, and competitive logics. Material logic was especially prevalent as chemists, physicists, and electrical engineers struggled for decades to create gallium nitride crystals and improve their electronic and light emitting properties. To transform gallium nitride into a material useful for the manufacture of LEDs, they relied on the full armentarium of semiconductor technologists: epitaxial reactors, ion implanters, high temperature furnaces, and techniques such as the deposition of different crystalline layers on the same crystal substrate.

Engineers and scientists at RCA, Nagoya University, Toyoda Gosei, and Nichia Chemical Industries struggled with the material logic imposed by gallium nitride to meet the needs of potential markets. Remarkably, these intended markets changed very little over this forty-year period. Since the late 1960s, technologists saw a market for gallium nitride-based blue light emitting diodes in LED-based displays, such as flat panel televisions. From the start, they were also interested in producing LEDs for the illumination market. As new types of communication and consumer electronic devices emerged, gallium nitride technologists identified other markets for their diodes such as cell phones and blue lasers employed in compact disks. It was this market logic that drove research on gallium nitride for many decades before the material was commercialized in the first half of the 1990s.

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structuring between human intentions and material systems and on the work of other sociologists who pioneered the “material turn” in science studies. For example, in an effort to reconceptualize sociology and to sketch a new metaphysics, Bruno Latour, Michel Callon, and other actor-network theorists argued that nonhuman entities have agency and that they compose the social world as much as humans do. They also pointed to the constant co-redefinition of knowledge, society, and nature. While building on actor-network theory, the logics of innovation framework is based on a more limited understanding of the agency of nonhuman entities. In the logics of innovation approach, material substances have an inert and ambiguous form of agency. They can be either well understood or opaque to human understanding. They can be enormously plastic and flexible, but they can also be intractable and offer considerable resistance to human action. For the material turn in science studies, see Andrew Pickering, “The Mangle of Practice: Agency and Emergence in the Sociology of Science,” *American Journal of Sociology* 99 (1993): 559–89; Andrew Pickering, *Mangle of Practice* (Chicago: University of Chicago Press, 1995); Andrew Pickering, “Decentering Sociology: Synthetic Dies and Social Theory,” *Perspectives on Science* 13 (2005): 352–405; Bruno Latour, *Les microbes: Guerre et Paix, suivi de Irréductions* (Paris: A. M. Métailié, 1984); Michel Callon, “Éléments pour une sociologie de la traduction: La domestication des coquilles St-Jacques et des marins pêcheurs dans la baie de St. Brieuc,” *L'Année Sociologique* 36 (1986): 169–208; and Michel Callon, “Society in the Making: The Study of Technology as a Tool for Sociological Analysis,” in *The Social Construction of Technological Systems: New Directions in the Sociology and History of Technology*, ed. Wiebe Bijker, Thomas Hughes, and Trevor Pinch (Cambridge, MA: MIT Press, 1987).

Competitive logic also shaped the development of gallium nitride, mostly in the early days of the material and again in the late 1980s and 1990s when Toyoda Gosei and Nichia Chemical competed to bring gallium nitride and blue LEDs to the market. Fierce competition between these firms led to the development of stable production processes and the constant improvement of gallium nitride crystals and blue LEDs. Also important for the development of gallium nitride was competition in semiconductors between Japan and the United States. Since the late 1960s, Japanese semiconductor firms sought to gain a competitive advantage over their American counterparts, especially in silicon integrated circuits. This rivalry led to the semiconductor wars of the late 1970s and 1980s, when Japanese manufacturers of integrated circuits gained the upper hand in global markets for semiconductors. The U.S. government reacted by supporting its domestic semiconductor industry and negotiating a trade agreement opening the Japanese market to American microchips. Competition between the United States and Japan extended to compound semiconductors as well.<sup>8</sup>

The development of gallium nitride was as much about failures and dead-ends as it was about successful innovation. It occurred in three phases. It was at the RCA laboratories and at Stanford University in the late 1960s and early 1970s that Herbert Maruska and other researchers pioneered the first single crystals of gallium nitride, in order to produce blue light emitting diodes for flat panel televisions. They did so with Cold War funding provided by ARPA and NASA. Using this novel material, they made relatively simple and rather dim blue and violet light emitting diodes. But the group at RCA rapidly encountered significant difficulties in growing positively charged gallium nitride, which was critical for the fabrication of a more advanced and brighter form of LED. The logic imposed by gallium nitride and competitive logic in the form of a severe financial crisis at RCA brought about by Japanese competition in consumer electronics persuaded RCA's research managers to terminate the project in 1974.

Isamu Akasaki, a Japanese applied physicist, pursued the further development of gallium nitride at the Matsushita Research Institute Tokyo, Inc. (MRIT) in the 1970s and 1980s. At Matsushita, Akasaki reproduced Maruska's work and re-created essential aspects of RCA's material logic. After joining Nagoya University in 1981, Akasaki made several technological breakthroughs

8. For the semiconductor wars, see Kenneth Flamm, *Mismanaged Trade? Strategic Policy and the Semiconductor Industry* (Washington, DC: Brookings Institution Press, 1996).

in collaboration with his doctoral student Hiroshi Amano. Akasaki and Amano pioneered new techniques for the deposition of crystal films and for altering their electrical properties. These revolutionary techniques enabled them to fabricate the first bright blue LEDs in 1989.

Akasaki's inventions were commercialized by two recent entrants into the LED business: Toyoda Gosei, a Toyota subsidiary manufacturing tires, and Nichia Chemical Industries, a chemical firm that produced phosphors for color televisions. Exploiting Akasaki's work and relying on novel processing techniques developed at NTT and Nagoya University, Nakamura's group at Nichia Chemical took an early lead. They perfected the growth of gallium nitride crystals and designed a low-cost process for altering their electrical and light emission properties. These incremental innovations enabled Nichia and later Toyoda Gosei to "tame" the logic of gallium nitride and to mass produce gallium nitride crystals and blue LEDs. These LEDs and other types of gallium nitride-based diodes were used at first in public displays and traffic signs, before being widely adopted in cell phones and other consumer electronics products. They permitted the production of LED-based flat panel displays. When properly coated with phosphor, they also increasingly displaced incandescent light bulbs.

## CREATING A NEW MATERIAL

Gallium nitride emerged out of the laboratories of the Radio Corporation of America (RCA). RCA was an important center for compound semiconductor research. Starting in the mid-1950s, managers of the firm's research laboratories in Princeton, New Jersey, made significant investments in compound semiconductors. They recruited young PhDs to the compound semiconductor program and reassigned experienced researchers who had previously worked on silicon and germanium. By the late 1960s, RCA had one of the largest research groups on compound semiconductors in the United States with thirty PhD researchers and more than fifty technicians and associate researchers.<sup>9</sup>

RCA's managers saw gallium arsenide and other compound semiconductors as representing the future of semiconductor technology because of their unusual electronic and optical properties. In their view, unlike silicon and

9. Hyungsub Choi, "The Boundaries of Industrial Research: Making Transistors at RCA, 1948–1960," *Technology and Culture* 48 (2007): 758–82.



germanium, these materials would permit the fabrication of new types of transistors and optoelectronic devices. They would also allow the making of novel components such as varactors, laser diodes, and light emitting diodes and open up new applications for semiconductors in communications and consumer electronics. In their view, compound semiconductors had the added advantage of being heavily funded by the Department of Defense, NASA, and other federal agencies (like RCA, these agencies viewed compound semiconductors as offering unique opportunities for the making of high-speed integrated circuits and optoelectronic devices for military and aerospace applications). Plentiful federal funding would support the research enterprise at RCA, at a time when other financial sources were drying up. Since the 1930s, RCA had financed its extensive research activities with the proceeds of the licensing of its intellectual property to other American corporations. But in 1957, the firm agreed to a consent decree with the U.S. Department of Justice obligating it to give royalty-free licenses on a nonexclusive basis to domestic competitors. Federal monies would replace licensing revenues to support research at RCA. The firm's turn to compound semiconductors can also be interpreted as part of a wider movement toward the development of new materials in American industry in the 1950s and 1960s. It was widely believed among research managers that materials were a bottleneck for the development of new technologies. Materials innovations would open up new technological and market opportunities.<sup>10</sup>

The RCA labs quickly emerged as a major center of innovation in compound semiconductor materials. The labs' researchers created several epitaxial techniques for fabricating them (epitaxy was the process whereby one grew a crystalline layer on top of another crystal). For example, Herbert Nelson invented liquid phase epitaxy at RCA. This technique became the preferred technique for the fabrication of most compound semiconductors in the 1960s and the 1970s. Then, in 1965 and 1966 two PhD chemists at RCA, James Tietjen and James Amick, developed a new epitaxial method for making compound semiconductor crystals: hydride vapor phase epitaxy or HVPE. Unlike liquid phase epitaxy which employed liquid precursors, HVPE relied on the chemical reaction of different gases, including hydrides (hydrogen-based compounds). These crystal-growing techniques enabled the firm's researchers to fabricate a large number of compound semiconductor materials. They developed competencies in indium phosphide, gallium

10. *Ibid.*

arsenide, and various gallium arsenide alloys, including gallium arsenide phosphide.<sup>11</sup>

Another section in the RCA labs, mostly composed of physicists, used these crystals to create new devices. For example, a group around Jacques Pankove, a Russian-born physicist and a former germanium transistor specialist, was at the forefront of research on semiconductor lasers. His group was one of the first in the United States to make gallium arsenide-based infrared lasers. Pankove later branched out into heterostructure laser diodes, namely diodes made of layers of different compound semiconductor materials. Henry Kressel, another star compound semiconductor specialist at RCA, pioneered the first semiconductor lasers working at room temperature (the first lasers operated in very cold environments). Other researchers worked on varactor diodes, diodes used for communications purposes. RCA researchers also fabricated red and yellow light emitting diodes.

In the spring of 1968, Tietjen, a hard-driving researcher with managerial ambitions, became interested in using the HVPE method he had co-invented with Amick to grow a new kind of compound semiconductor material—single crystals of gallium nitride (no other laboratory had yet produced these crystals). Tietjen thought that the growth of gallium nitride crystals would make it possible to fabricate blue LEDs. He surmised that gallium nitride, because of the position of gallium and nitrogen in the periodic table of elements, would have one of the widest band gaps among compound semiconductors. This wide band gap would make gallium nitride a valuable semiconductor for blue light emission.<sup>12</sup>

Tietjen further reasoned that gallium nitride-based blue LEDs would have great commercial potential. They would be critical for the making of flat panel televisions. The compound semiconductor group at RCA already knew how to

11. James Tietjen and James Amick, "Preparation and Properties of Vapor-Deposited Epitaxial GaAsP Using Arsine and Phosphine," *Journal of the Electrochemical Society* 113 (1966): 724–28; James Tietjen, interview by Christophe Lécuyer, 14 Jun 2010. Tietjen and Maruska's project was in direct competition with the large research program on liquid crystal displays at RCA. This program, like Tietjen's, aimed at the production of flat panel displays for television sets and other applications. Flat panel TVs were the holy grail at RCA. David Sarnoff, RCA's chairman, had identified flat panel video displays as one of the main technologies to be developed in the labs. Flat panel displays would be lighter and easier to move than cathode ray tubes and they had aesthetic appeal as well. For the history of LCD technology at RCA see Benjamin Gross, "Crystallizing Innovation: The Emergence of the LCD at RCA, 1951–1976" (PhD dissertation, Princeton University, 2011).

12. Tietjen, interview (ref. 11).

make LEDs with two primary colors: red and yellow. Adding blue would enable them to produce the full spectrum of colors and thereby produce attractive flat panel television sets. Tietjen expected the market for flat panel TVs to be very large. Thus, the development of gallium nitride crystals was shaped from the start by the interplay between material and market logic. The material logic of HVPE reactors was essential for the creation of gallium nitride crystals. But it was market logic, intended markets for gallium nitride-based LEDs, that gave meaning to this project and justified significant investments of time and resources in it.<sup>13</sup>

Enthusied by the commercial potential of blue LEDs, Tietjen asked Herbert Maruska, a young trainee in the Princeton laboratories, to use the lab's HVPE technology to grow single crystals of gallium nitride and to make blue PN junction LEDs. PN junction LEDs were LEDs that relied on a junction between a semiconductor layer with an excess of electrons and a layer with an oversupply of electron deficiencies or holes. The recombination of electrons and holes across the junction led to the production of photons and to light emission. PN junction LEDs were known as the form of LEDs that were the most efficient and emitted the most light. A native of New York, Maruska had recently received a master's degree in electrical engineering from New York University and joined the associate technical staff of the RCA laboratories. At RCA, Maruska had at first worked with Pankove on gallium arsenide phosphide lasers. Interested in the growth of the compound semiconductor materials used by Pankove, he had moved to Tietjen's lab where he had familiarized himself with HVPE and grown films of gallium arsenide phosphide, indium phosphide, and indium arsenide. To fund Maruska's high-risk project on gallium nitride, Tietjen used monies coming from a NASA contract as well as internal RCA funds.<sup>14</sup>

It took Maruska more than eight months of constant experimentation to grow single crystals of gallium nitride. Such was the difficulty of fabricating this new material (it was much more difficult to grow than other compound semiconductors). Maruska repurposed the HVPE reactor he had used for growing other types of compound semiconductors to accommodate the gases needed to fabricate single crystals of gallium nitride: ammonia gases and

13. *Ibid.*; Herbert Maruska, interview conducted by Christophe Lécuier, 23 Jun 2010; Herbert Maruska, "The Early History of Gallium Nitride Light-Emitting Diodes," CCL.

14. Tietjen, interview (ref. 11); Maruska, interview (ref. 13); Maruska, "Early History of Gallium Nitride" (ref. 13).

gallium chloride (gallium chloride was the product of the reaction between gallium and hydrochloride). Maruska also faced significant technical uncertainties. Which temperatures should he use to grow gallium nitride crystals? Which crystal substrate should he employ (he needed to grow epitaxial films of gallium nitride on another material with a similar crystalline structure)? Which method and which chemical elements should he adopt to dope crystal films and make PN junction LEDs. Doping was the process whereby one introduced a chemical substance into a semiconductor crystal in order to change its electrical properties.<sup>15</sup>

Maruska experimented with sapphire, quartz, silicon, and silicon carbide, before settling on sapphire as the crystal substrate to grow gallium nitride (this was a prescient choice as the great majority of gallium nitride crystals have been grown on sapphire up to this day). Getting access to these sapphire crystals was easy at the Princeton labs. They were part of the local material logic. There was an abundant supply of sapphire at RCA, as other researchers were engaged in the making of radiation-hardened silicon integrated circuits that used sapphire substrates. These silicon-on-sapphire integrated circuits were directed at military and space applications. Maruska also struggled for many months with the right temperature for growing gallium nitride films. At first, he used low temperatures, in the order of 600°C, to grow the crystals, but he obtained only white powder. It was only by raising the temperature to 900°C, the temperature used to make gallium arsenide, that he succeeded in making single crystals of gallium nitride. This important breakthrough was accomplished in November 1968.<sup>16</sup>

But Maruska did not succeed in making PN junction light emitting diodes. PN junction LEDs were made of a sandwich of N-type and P-type layers. N-type crystal layers were negatively charged. They were characterized by an excess of electrons. In contrast, P-type layers were positively charged. They had an oversupply of electron deficiencies or holes (holes, like electrons, were able to move within the crystal). Single crystals of gallium nitride came naturally as N-type. But it proved exceedingly difficult to make P-type gallium

15. Herbert Maruska, research notebook 14 Mar 1968 to 24 May 1968, courtesy of Alexander Magoun, David Sarnoff Library, Princeton, NJ; Herbert Maruska, research notebook 23 May 1968 to 28 Feb 1969, courtesy of Alexander Magoun, David Sarnoff Library, Princeton, NJ; Tietjen, interview (ref. 11); Maruska, interview (ref. 13); Maruska, "Early History of Gallium Nitride," CCL (ref. 13).

16. Maruska, research notebooks (ref. 15); Tietjen, interview (ref. 11); Maruska, interview (ref. 13); Maruska, "Early History of Gallium Nitride," CCL (ref. 13).

nitride. To fabricate P-type crystals, Maruska tried to dope crystalline films with zinc, a P-type element. He also used other P-type elements such as magnesium, cadmium, and mercury. None of these efforts succeeded, however. Maruska could not make P-type gallium nitride. He had run into a major problem with the material logic of gallium nitride that would plague researchers aiming at making blue LEDs for the next twenty years.<sup>17</sup>

Maruska and Tietjen published their article on single crystals of gallium nitride in *Applied Physics Letters* in November 1969. This article attracted significant attention in the compound semiconductor community. The market potential of gallium nitride was obvious among compound semiconductor researchers at that time. Gallium nitride appeared to be the material that would enable the fabrication of blue PN junction LEDs, and the market potential of these devices seemed to be very large. LEDs were then starting to be used in significant quantities in the numeric displays of electronic watches and scientific calculators such as those produced by Hewlett-Packard. Blue LEDs would find applications in scientific calculators and other digital devices as well. They would make possible the manufacture of LED-based flat panel TVs. Finally, it was generally believed that blue LEDs would pave the way to the generation of white light and open up very large markets in illumination.<sup>18</sup>

These commercial opportunities encouraged several electronics corporations to establish research groups focusing on gallium nitride in the late 1960s. Among these firms were large U.S. corporations with significant research operations such as North American-Rockwell and the Bell Telephone Laboratories. N.V. Philips, the Dutch consumer electronics giant, also moved into gallium nitride research at that time. So did Japanese corporations such as Hitachi and Toshiba. Most researchers working on gallium nitride at these firms followed Maruska's example and grew their films of gallium nitride with HVPE.<sup>19</sup>

Maruska's breakthrough also convinced Tietjen, who had by then ascended in the lab hierarchy, as well as other RCA managers to invest more in gallium nitride. Their goal was to make blue PN junction LEDs. In early 1970, Pankove, who had recently returned from a sabbatical at UC Berkeley where he had written a textbook on optical processes in semiconductors, joined the

17. Maruska, interview (ref. 13); Maruska, "Early History of Gallium Nitride" (ref. 13).

18. Herbert Maruska and James Tietjen, "The Preparation and Properties of Vapor-Deposited Single-Crystalline GaN," *Applied Physics Letters* 15 (1969): 327–29; Herbert Maruska, "Gallium Nitride Light Emitting Diodes" (PhD dissertation, Stanford University, 1973), Stanford University Archives and Special Collections, Stanford, CA.

19. E. Fred Schubert, *Light-Emitting Diodes* (New York: Cambridge University Press, 2006).

research group on gallium nitride. So did Edward Miller, a research chemist. At the same time, Maruska, who had developed a fascination and a deep emotional attachment to the material he had created, joined the doctoral program in materials science and engineering at Stanford University. At Stanford, he continued working on gallium nitride and blue LEDs with RCA funding and in close collaboration with Pankove. Maruska's work at Stanford was also financed by an ARPA contract. ARPA was then interested in pushing semiconductor development beyond silicon and supported the development of compound semiconductor materials such as gallium arsenide and gallium arsenide phosphide for military applications.<sup>20</sup>

In the next few years, Pankove, Miller, and Maruska grappled with the material logic of gallium nitride. They focused on improving the quality of gallium nitride crystals and on doping them with P-type elements. For example, Miller sought to optimize the properties of gallium nitride crystals by modifying the temperature and the flow rates at which these crystals were grown. This required detailed studies of gas flows within the HVPE reactor to determine the best positioning of the substrate facing the gas flow and to obtain good crystals. Miller sought also to dope gallium nitride crystals with zinc, a P-type dopant. At Stanford, Maruska developed a parallel approach to P-type doping by using magnesium, instead of zinc. But in spite of considerable efforts, neither Maruska nor Miller succeeded in making P-type gallium nitride crystals.<sup>21</sup>

As it proved to be extremely difficult to make PN junction LEDs, in 1971 Pankove, the device physicist, redirected the group's activities toward the fabrication of another type of LEDs, metal-insulator-semiconductor (MIS) LEDs. These LEDs did not require P-type gallium nitride crystals. Pankove's idea was to grow a thin film of insulating gallium nitride, heavily doped with zinc, on top of a thicker film of undoped N-type gallium nitride. By applying a voltage to the device, one forced electrons in the N-type base to collide into the P-type atoms in the insulating layer, thereby knocking off electrons stored on the dopant atoms. These electrons would then recombine with holes and emit light. Miller, the group's chemist, made the first gallium nitride MIS LED in mid-1971. It emitted green light. Soon, he fabricated blue LEDs as well. Using the same basic device structure, Maruska made green, blue, and violet

20. Maruska, "Gallium Nitride Light Emitting Diodes" (ref. 18); Edward Miller, "The Early History of Gallium Nitride Research," at <http://www.compoundsemi.com/documents/articles/cldoc/5022.html> (accessed 1 Jun 2010).

21. Maruska, "Gallium Nitride Light Emitting Diodes" (ref. 18); Miller, "Gallium Nitride Research" (ref. 20).

MIS LEDs at Stanford in 1972 and 1973. But these devices, like Miller's LEDs, had low efficiency and remained very dim. It was clear that they were far from being ready for commercial production.<sup>22</sup>

Persistent difficulties with the material logic persuaded Tietjen to terminate the project in 1974. He laid off Maruska, who had recently returned from Stanford with his doctorate. Miller was let go as well and Pankove was reassigned to other projects within the lab (he later moved to the University of Colorado). This decision to close down the gallium nitride project was part of a major reduction in the size of the compound semiconductor program at RCA. In the first half of the 1970s, RCA encountered enormous competitive pressures in consumer electronics from Japanese firms. It lost significant market share in television sets and other products to Japanese competitors. Experiencing serious financial difficulties, RCA downsized its research laboratories in Princeton and refocused them toward television technology. One of the main targets for budget cuts was research in compound semiconductors. In 1974 and 1975, the management of the laboratories phased out most compound semiconductor research projects, with a few exceptions such as the making of indium phosphide detectors for CAT scanners (medical devices remained an area of strategic importance for RCA). Interestingly, many other firms, including RCA's Japanese competitors, left gallium nitride research around the same time. Unable to make PN junction LEDs, the Bell Labs were among the first to abandon the field. Other corporations followed suit, such as Philips, Hitachi, and Toshiba. A consensus grew among compound semiconductor researchers that gallium nitride was an intractable material and that working on it was a loss of time and resources.<sup>23</sup>

## CONTENDING WITH MATERIAL LOGIC

When most research groups focusing on gallium nitride were disbanding, Isamu Akasaki, a Japanese applied physicist, took on the challenge of growing

22. Jacques Pankove, "Electroluminescent Semiconductor Device of GaN," U.S. Patent 3,683,240, filed 22 Jul 1971 and issued 8 Aug 1972; J. Pankove, E. Miller, and J. Berkeyheiser, "GaN Blue Light Emitting Diodes," *Journal of Luminescence* 5 (1972): 84–86; Maruska, "Gallium Nitride Light Emitting Diodes" (ref. 19); Maruska, "Early History of Gallium Nitride," CCL (ref. 13); Miller, "Gallium Nitride Research" (ref. 20).

23. Tietjen, interview (ref. 11); Maruska, interview (ref. 13); Maruska, "Early History of Gallium Nitride," CCL (ref. 13); Miller, "Gallium Nitride Research" (ref. 20); oral communication from Maurice Bernard to Christophe Lécuier, 28 Mar 2013.

high-quality gallium nitride crystals and making PN junctions LEDs at the Matsushita Research Institute in Tokyo. Akasaki was an experienced researcher. After obtaining an undergraduate degree in chemistry from Kyoto University, he had started his research career at Kobe Engineering Co. (currently known as Fujitsu Corporation), where he had worked on television tubes and had become fascinated with luminescent materials. He had later received a doctorate in applied physics from the department of electronics at Nagoya University. He had also taught as an assistant professor at the same university. In 1964, he joined the Matsushita Research Institute at the request of his mentor at Nagoya University (such was the power of Japanese senior scientists in the 1960s that they could assign junior faculty to positions in other organizations without consulting their younger colleagues). The Matsushita Research Institute Tokyo (MRIT), established four years earlier, was the fundamental research division of Matsushita Electric Industrial Co., the large Japanese electronics firm (now known as Panasonic). The MRIT had been conceived by Yujiro Koike, a leading figure in Japanese electronics and a professor at Tohoku University, as an academic institute doing basic research within a manufacturing organization. Koike's goal for the new institute was to do research that would be of similar quality as the one performed at the University of Tokyo, the Bell Telephone Laboratories, and other major corporate laboratories in the United States. All laboratory directors at the Matsushita Research Institute came from national universities (the top public universities in Japan), where they had taught as assistant or associate professors.<sup>24</sup>

Akasaki, who had worked on germanium crystals at Nagoya University, chose to do research on compound semiconductors when he joined the Matsushita Research Institute. Akasaki, who had long been interested in luminescent crystals, saw his specializing in compound semiconductors as a way to grow and investigate these crystals. In 1964, he became the head of one of MRIT's fundamental research laboratories and later the general manager of its

24. Isamu Akasaki, oral history interview by Takahiro Ueyama, 2 Nov and 10 Dec 2010. See also Isamu Akasaki, "Renaissance and Progress in Nitride Semiconductors: My Personal History of Nitride Research," *Material Research Society Symposium Proceedings* 639 (2001): G8.1.1–12; Isamu Akasaki, "Aoiro Kakko Daiodo O Motomete," *Oyo Butsuri* 76 (2007): 892–98; Isamu Akasaki, "Yume No Kakko Debaisu No Jitsugen O Kataru," *Oyo Butsuri* 73 (2004): 1060–67; Isamu Akasaki, "Aoiro Kakko Daiodo Wa Ikani Tsukuraretaka," *Denki Tsushin Joho Gakkaishi* 9 (2008): 1009–23; Isamu Akasaki, "Aoiro Hakko Daiodo O Motomete: Watashi No Tikkabutsu Kenkyuu," *Oyo Butsuri* 71 (2002): 814–17.



semiconductor division. His laboratory at Matsushita was large, with more than ten researchers. Akasaki and his group grew crystals of gallium arsenide and its alloys. For example, in 1968, using vapor phase epitaxy, Akasaki and his group made high-quality gallium arsenide crystals that had the highest electron velocity up to that time. Akasaki also experimented with a new crystal-growing method, the Liquid Encapsulated Czochralski (LEC) method, to grow high-purity epitaxial layers of gallium phosphide. This breakthrough enabled him and his group to make red and green LEDs.<sup>25</sup>

Pursuing his interest in luminescence, Akasaki increasingly looked to gallium nitride as a way of fabricating blue LEDs.<sup>26</sup> He met Pankove at a conference in Boulder, Colorado, in October 1972 and learned about his work on gallium nitride. He later visited him in his laboratory at RCA, where he heard about blue MIS LEDs and gallium nitride crystals.<sup>27</sup> This visit prompted Akasaki to start a gallium nitride project (funded by MRIT) in parallel with his other projects on gallium arsenide and gallium phosphide. At first, he re-created essential aspects of RCA's material logic. His group built an HVPE reactor and they grew single crystals of gallium nitride. They reproduced the results obtained by Maruska and Pankove at RCA and Stanford University. But it became increasingly clear to Akasaki that HVPE had significant downsides for the fabrication of gallium nitride crystals. The growth rate in HVPE reactors was too rapid, which made it difficult to carefully control the fabrication of the crystal. Hydrogen trichloride corroded the equipment. Akasaki also suspected that the HVPE process introduced contaminants into the crystal (contaminants were chemical elements that impaired the electrical and luminescent properties of semiconductor crystals).<sup>28</sup>

25. Akasaki, interview (ref. 24); I. Akasaki and T. Hara, "Non-Ohmic Properties and Negative Resistance in n-Type GaAs," *Proceedings of the Ninth ICPS Conference 2* (1968): 787–92; Isamu Akasaki, "Process of Fabricating Electroluminescent Element," U.S. Patent 3,649,383, filed 17 Mar 1969 and issued 14 Mar 1972.

26. Akasaki claims to have thought of gallium nitride as the appropriate material for blue LEDs independently of the work of Maruska and Pankove. Akasaki, interview (ref. 24).

27. Akasaki became familiar with Maruska and Pankove's work mostly through the literature. According to him, he did not learn much from his meeting with Pankove. He also did not have any contact with researchers working at RCA's laboratory in Tokyo, who knew of Pankove and Maruska's work on gallium nitride. Akasaki, interview (ref. 24).

28. *Ibid.*; Isamu Akasaki, "Insight, Challenge, and Success: The Development of GaN Blue Light Emitting Devices," at <http://www.takeda-foundation.jp/en/award/takeda/2002/forum/oza.html> (accessed 20 Oct 2011); Isamu Akasaki, "Progress in Crystal Growth of Nitride Semiconductors," *Journal of Crystal Growth* 221 (2000): 231–39; Isamu Akasaki and Hiroshi Amano, "Breakthroughs in Improving the Crystal Quality of GaN and Invention of the PN Junction

This assessment led Akasaki to add new elements to the material logic. He explored the use of molecular beam epitaxy (MBE), another method used for growing compound semiconductor crystals, to make single crystals of gallium nitride. Unlike HVPE, which was a chemical method for growing crystals, molecular beam epitaxy was a physical process. MBE, which had been developed at the Bell Telephone Laboratories in the late 1960s, enabled the growth of semiconductor crystals in a more controlled fashion than HVPE.<sup>29</sup> In addition, Akasaki was interested in using ion implantation to dope gallium nitride with P-type elements. Ion implantation, a technique derived from nuclear physics research, had been recently used to introduce dopants into silicon and gallium arsenide crystals to make transistors, integrated circuits, and photovoltaic cells.<sup>30</sup> In order to fund this ambitious research program whereby he would apply these new tools and techniques to the making of gallium nitride and blue LEDs, Akasaki turned to MITI, the Japanese Ministry of International Trade and Industry, which saw compound semiconductors as an area of strategic importance for Japanese industry and funded research in this area. The Ministry granted Akasaki a small grant for his project on “Creation of Blue Light Emitting Devices by Using Ion Implantation Technique in Gallium Nitride Single Crystals” in 1974. The following year, Akasaki received a much larger three-year grant (“Applied Study of Blue Light Emitters”) from MITI jointly with other corporate researchers. This project financed Akasaki’s laboratory as well as the work of Izuo Hayashi, a leading researcher at NEC who had made the first heterostructure laser (composed of a sandwich of two different compound semiconductor materials) at the Bell Telephone Laboratories.<sup>31</sup>

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Blue-Light-Emitting Diode,” *Japanese Journal of Applied Physics* 45 (2006): 9001–10; Isamu Akasaki, “Key Inventions in the History of Nitride-based Blue LED and LD,” *Journal of Crystal Growth* 300 (2007): 2–10; Isamu Akasaki, “Dramatic Improvement in Gallium Nitride Crystal Quality and Realization of PN Junction Blue LEDs,” paper presented at *Nitride Semiconductors and Their Device Applications: Current Status and Future Prospects*, 2009 Kyoto Prize Workshop in Advanced Technology, Inamori Foundation, 12 Nov 2009, at [http://www.inamori-f.or.jp/laureates/k25\\_a\\_isamu/img/wks\\_e.pdf](http://www.inamori-f.or.jp/laureates/k25_a_isamu/img/wks_e.pdf) (accessed 20 Oct 2011).

29. For the history of molecular beam epitaxy, see W. Patrick McCray, “MBE Deserves a Place in the History Books,” *Nature Nanotechnology* 2 (2007): 259–61.

30. For the history of ion implantation, see Christophe Lécuyer and David C. Brock, “From Nuclear Physics to Semiconductor Manufacturing: The Making of Ion Implantation,” *History and Technology* 25, no. 3 (2009): 193–217; Richard Fair, “History of Some Early Developments in Ion-Implantation Technology Leading to Transistor Manufacturing,” *Proceedings of the IEEE* 86 (1998): III–37.

31. Akasaki, interview (ref. 24); Akasaki, “Insight, Challenge, and Success” (ref. 28); Akasaki, “Progress in Crystal Growth” (ref. 28); Akasaki and Amano, “Breakthroughs” (ref. 28); Akasaki,

With MITI funding, Akasaki purchased an expensive MBE machine and adapted it to the growth of gallium nitride crystals. In 1974, Akasaki and H. Kobayashi succeeded in growing films of gallium nitride with MBE, but they discovered that the crystals they had created were not appreciably better than those made with HVPE (they nearly had as many dislocations or cracks as those made with hydride vapor phase epitaxy). They also discovered that ion implantation did not dope gallium nitride with P-type elements. The material logic of gallium nitride and blue light emitting diodes remained as intractable as ever.<sup>32</sup>

These disappointing results led Akasaki, like Pankove several years earlier, to reorient his group toward the making of MIS LEDs. The main production problem with MIS LEDs was that it was very difficult to attach the electrodes to the gallium nitride films. This challenge led Akasaki to invent a new LED structure whereby one of the electrodes (a pillar of heavily doped gallium nitride on top of a layer of silicon oxide) would be incorporated into the gallium nitride crystal. Making this device structure required the acquisition of new material capabilities. Over a three-year period, Akasaki's group learned to grow strongly doped N-type gallium nitride on top of silicon oxide with HVPE. These capabilities enabled the group to make MIS LEDs that were easier to manufacture than those fabricated by Miller and Maruska. The LEDs were also more efficient and more luminous. These results encouraged the managers of the MRIT to bring Akasaki's MIS LEDs to pilot production. Matsushita Electric produced roughly 10,000 of these devices. But the firm encountered significant yield problems (yield being the proportion of good devices coming out the manufacturing line). Yields of blue LEDs were low due to numerous cracks and pits in the gallium nitride crystals and to the difficulty of controlling the thickness of the insulating layers, and hence the LEDs' operating voltage. As a result, Matsushita promptly ceased producing the blue MIS LEDs and did not commercialize them.<sup>33</sup>

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"Key Inventions" (ref. 28); Izuo Hayashi, "Heterostructure Lasers," *IEEE Transactions on Electron Devices* 31 (1984): 1630–42.

32. Akasaki, interview (ref. 24); Akasaki, "Insight, Challenge, and Success" (ref. 28); Akasaki, "Progress in Crystal Growth" (ref. 28); Akasaki and Amano, "Breakthroughs" (ref. 28); Akasaki, "Key Inventions" (ref. 28).

33. Yoshimasa Ohki, Yukio Toyoda, Hiroyuki Kobayashi, and Isamu Akasaki, "Gallium Nitride Light-Emitting Elements and Method of Manufacturing the Same," U.S. Patent 4,396,929, filed 20 Oct 1980 and issued 2 Aug 1983; Akasaki, interview (ref. 24); Akasaki, "Insight, Challenge, and Success" (ref. 28); Akasaki, "Progress in Crystal Growth" (ref. 28); Akasaki and Amano, "Breakthroughs" (ref. 28); Akasaki, "Key Inventions" (ref. 28).

After five years of a long struggle with the material logic of gallium nitride, Akasaki had very little to show for his efforts. But he had become obsessed with gallium nitride and was convinced that in spite of his lack of success and gallium nitride's reputation as a recalcitrant material, it was possible to grow high-quality crystals of gallium nitride and to make PN junction LEDs with them. This conviction came from observations that Akasaki had made of crystals of gallium nitride in 1978. Using fluorescence microscopy, a technique mostly employed in biomedicine, he had seen high-quality micro-crystals in tiny parts of much larger crystals. Members of his group had also noticed that clusters of needle-like crystals left in the HVPE reactor had good luminescent properties. As Akasaki later reported, these observations gave him "a gut feeling for the inherent quality of gallium nitride."<sup>34</sup>

The challenge was to make large crystals with the same quality as the micro-crystals he had noticed in his lab. Akasaki tackled this problem at his new post at Nagoya University. In 1979, Nagoya University asked him to return as a senior faculty member in the department of electrical engineering and to build a research group in compound semiconductors. Akasaki joined the university two years later.<sup>35</sup> Moving to a university environment was not without challenges. The university did not have adequate facilities to do compound semiconductor research. Akasaki had to convince the university to build a clean room and he had to design the clean room himself. Akasaki also needed to obtain external funds to support research on compound semiconductors (in his last years at Matsushita, his gallium phosphide red LED project had been funded by Konosuke Matsushita, the founder of Matsushita Electric, through his special budget). To finance research on gallium nitride, Akasaki secured grants from the Ministry of Education to do research on other compound semiconductors such as gallium arsenide and its alloys, and he diverted some of these funds to finance the gallium nitride project. It was impossible to secure funding for gallium nitride research, as no Japanese governmental agency wanted to support research on such a hopeless material.<sup>36</sup>

34. Akasaki, "Insight, Challenge, and Success" (ref. 28); Akasaki, interview (ref. 24).

35. According to Akasaki, Matsushita's managers wanted him to stay at the company. MITI was then organizing a large research program on photonics on the model of its previous research program on VLSI (very-large-scale integration). Losing Akasaki meant that Matsushita would not be able to participate in the new program piloted by MITI. Akasaki, interview (ref. 24).

36. Akasaki, interview (ref. 24); Akasaki, "Insight, Challenge, and Success" (ref. 28); Akasaki, "Progress in Crystal Growth" (ref. 28); Akasaki and Amano, "Breakthroughs" (ref. 28); Akasaki, "Key Inventions" (ref. 28).

Since HVPE and MBE did not seem conducive to the production of good gallium nitride single crystals, Akasaki added yet another element to the material logic. He turned to a new crystal-growing technique, metal-organic chemical vapor deposition (MOCVD), to make these crystals.<sup>37</sup> MOCVD was an epitaxial technique closely related to HVPE. Like HVPE, it was based on the deposition of chemical vapors. But the chemistry of the gases was different. In MOCVD, they were metal-organic gases, whereas they were hydrogen-based in HVPE. MOCVD had been pioneered by Harold Manasevit, a chemist, at North American-Rockwell in the late 1960s. Manasevit had also been the first researcher to use MOCVD to make single crystals of gallium nitride (in 1971). His crystals, however, were of poor quality. Since Manasevit's pioneering work, MOCVD reactors and techniques had been perfected by researchers at Philips and by Manasevit's colleagues at Rockwell. In the late 1970s, Japanese researchers increasingly adopted MOCVD to grow compound semiconductor crystals. For example, a laboratory at Sony pioneered its use for the making of laser diodes for compact disk players in 1977. Other firms such as Toshiba, NEC, Hitachi, Sharp, and Mitsubishi Electric also built MOCVD capabilities in the late 1970s. Thus by 1981, MOCVD had become an established technique in the Japanese compound semiconductor community. But no researcher in Japan had yet used MOCVD to grow gallium nitride.<sup>38</sup>

In Akasaki's view, MOCVD had several advantages over HVPE and MBE for the growth of single crystals of gallium nitride. MOCVD was faster than MBE and it was much more controllable than HVPE. Akasaki also felt that MOCVD was better at depositing films of gallium nitride on a material with a different crystalline structure such as sapphire. Moving to organometallic gases also solved the problem of contaminants introduced by hydride gases in HVPE reactors. Switching to MOCVD did not prove easy, however. At first,

37. Akasaki claims that he made the decision to move to MOCVD in 1979, when he was still at Matsushita. See Akasaki, interview (ref. 24). MOCVD is also known as MOVPE, or OMVPE, organometallic vapor phase epitaxy.

38. H. M. Manasevit, F. M. Erdmann, and W. I. Simpson, "Use of Metalorganics in the Preparation of Semiconducting Materials: IV. The Nitrides of Aluminum and Gallium," *Journal of the Electrochemical Society* 118 (1971): 1864; H. M. Manasevit, "Recollections and Reflections of MO-CVD," *Journal of Crystal Growth* 55 (1981): 1–9; Russell Dupuis, "Harold M. Manasevit (1927–2008)," *Electrochemical Society: Interface* 18, no. 2 (2009): 23–24; Hiroshi Shimizu and Satoshi Kudo, "How Well Does Knowledge Travel? The Transition from Energy to Commercial Application of Laser Diode Fabrication Technology," paper presented at the 2011 Business History Conference, St. Louis, MO, at <http://www.thebhc.org/publications/BEHOnline/2011/shimizuandkudo.pdf> (accessed 21 Oct 2011).

students in Akasaki's lab tried to transform the HVPE machine that Akasaki had brought from Matsushita into a MOCVD reactor. But as this proved impossible, Yasuo Koide and Hiroshi Amano, two graduate students in the laboratory, built a MOCVD reactor from scratch on the basis of articles they had found in the technical literature. They were assisted in this endeavor by students from Toyohashi University of Technology, a technical university nearby. As they conducted experiments with the new equipment, Akasaki's students made adjustments to it, for example redesigning the gas flows within the reactor to better grow their crystals.<sup>39</sup>

The shift to MOCVD enabled Akasaki and his students to make high-quality crystals of gallium nitride in 1986. The use of MOCVD permitted the introduction of a layer of aluminum nitride between the sapphire crystal and the film of gallium nitride. This layer of aluminum nitride acted as a buffer between the sapphire and the gallium nitride. It accommodated differences in their crystalline structures. As a result, the buffer allowed the growth of gallium crystals with much fewer dislocations (or cracks) than crystals directly grown over sapphire crystals. Divergent accounts have been offered for this critical breakthrough. According to Akasaki, he had thought about creating a buffer layer between the sapphire and the gallium nitride film as early as 1982 (the idea of the buffer layer itself was not original; low-temperature buffers had been used to grow other compound semiconductor crystals since the early 1960s). As the story goes, Akasaki asked Amano to try aluminum nitride for making the buffer.<sup>40</sup>

But Amano, the graduate student who produced the first high-quality crystals in Akasaki's laboratory, has a different account. He recalled that he

39. Hiroshi Amano, interview conducted by Takahiro Ueyama, 8 Dec 2010; Hiroshi Amano, "Passion and Struggle in the Development of Gallium Nitride Light Emitting Devices," 2002, at <http://www.takeda-foundation.jp/en/award/takeda/2002/forum/02a.html> (accessed 21 Oct 2011); Akasaki, "Insight, Challenge, and Success" (ref. 28); Akasaki, "Progress in Crystal Growth" (ref. 28); Akasaki and Amano, "Breakthroughs" (ref. 28); Akasaki, "Key Inventions" (ref. 28).

40. According to Akasaki, he had the idea of inserting a soft, low-temperature buffer layer to relieve the strain due to the mismatch between the gallium nitride epilayer and the sapphire substrate. For the buffer layers, he considered using AlN, ZnO, GaN, or SiC because these materials had similar physical properties to those of gallium nitride and sapphire. In discussions with Amano, Akasaki suggested that the buffer layer should be deposited at a temperature lower than 500°C and be thinner than fifty nanometers. Akasaki, interview (ref. 24). See also H. Amano, N. Sawaki, I. Akasaki, and Y. Toyoda, "Metalorganic Vapor Phase Epitaxial Growth of a High Quality GaN Film Using an AlN Buffer Layer," *Applied Physics Letters* 48 (1986): 353–55; Akasaki, "Insight, Challenge, and Success" (ref. 28); Akasaki, "Progress in Crystal Growth" (ref. 28); Akasaki and Amano, "Breakthroughs" (ref. 28); Akasaki, "Key Inventions" (ref. 28).

fabricated the first buffer layer in a serendipitous way. Because his reactor was not working properly one day (it could only operate at low temperatures), he thought of using these low temperatures to deposit a film of aluminum nitride. When his reactor started to operate normally again, he grew a film of gallium nitride on top of it. To his surprise, the gallium nitride film was so transparent that he thought at first that the experiment had failed and that he had not grown any crystal. But after a second examination, Amano discovered to his amazement that he had made the first high-quality crystal of gallium nitride. In the following months, he worked night and day to find the best conditions to make the aluminum nitride buffer. Akasaki, who had been schooled in the importance of intellectual property protection at Matsushita, patented this work soon thereafter.<sup>41</sup>

Amano and Akasaki's breakthrough attracted significant attention in the compound semiconductor community when it was published in 1986. As it became clear that they had refashioned the material logic of gallium nitride and that it was possible to make high-quality crystals (something which had previously been deemed impossible by many), several compound semiconductor researchers moved into the field, including Shuji Nakamura at Nichia Chemical Industries. These researchers tackled the issue of making P-type gallium nitride. Like Akasaki and his group, they were interested in fabricating PN junction LEDs. The field of gallium nitride research that had been nearly moribund for a decade and a half started to revive. From then on, competitive logic—the competition among laboratories, firms, and nations—played an increasingly important role in shaping the development of gallium nitride and blue LEDs.<sup>42</sup>

At the forefront of the race to P-type gallium nitride and blue PN junction LEDs were Akasaki, Amano, and their co-workers at Nagoya University. They had a significant advantage over other research groups: they knew how to grow high-quality crystals. In the second half of the 1980s, Akasaki and Amano solved the problem of P-type gallium nitride by combining several techniques. The first technique was the use of beams of electrons, a well-known technology

41. Amano, interview (ref. 39); Amano, "Passion and Struggle" (ref. 39). According to Akasaki, Amano did not understand at first that the aluminum nitride buffer enabled the making of high-quality gallium nitride crystals. But Akasaki, who had worked before with this material, quickly realized that this was the case. Akasaki, interview (ref. 24).

42. Amano, interview (ref. 39); Amano, "Passion and Struggle" (ref. 39); Akasaki, "Insight, Challenge, and Success" (ref. 28); Akasaki, "Progress in Crystal Growth" (ref. 28); Akasaki and Amano, "Breakthroughs" (ref. 28); Akasaki, "Key Inventions" (ref. 28).

in the material logic of compound semiconductor technologists. Since the early 1960s, electron beams had been used to improve the luminescence of various compound semiconductor materials, including gallium nitride. Compound semiconductor researchers had also employed electron beams to measure the luminescence of their crystals. In 1987, Akasaki sent Amano to measure the luminescence of gallium nitride crystals doped with zinc, a P-type dopant, in the well-equipped laboratory of NTT's Musashino Telecommunication Laboratory. There, Amano discovered to his great surprise that the more he shot electrons at his crystal samples, the more luminescent they became. But he also found that electron beams did not suffice by themselves to make positively charged gallium nitride.<sup>43</sup>

To fabricate P-type gallium nitride, one had (as the Akasaki lab discovered) to combine electron beams with the use of a different P-type dopant: magnesium.<sup>44</sup> Inspired by the results of a research group at Philips and by the work of Maruska at Stanford who had used magnesium for his doctoral work, Amano and other graduate students in the Akasaki lab experimented with a magnesium compound, bis-cyclopentadienyl magnesium. This was an organometallic compound compatible with MOCVD. With these new elements added to the material logic—magnesium compounds and electron beams—Akasaki, Amano, and their group fabricated P-type gallium nitride for the first time in 1988. Now that they knew how to make P-type gallium nitride, it was relatively easy to make a PN junction LED, a LED made of N-type and P-type layers of gallium nitride. This LED was much more efficient than the ones produced up until then. It also improved light emission by nearly two orders of magnitude. Akasaki and his students announced the PN junction LED in 1989. It had taken Akasaki sixteen years to realize his goal of the blue junction LED.<sup>45</sup>

This breakthrough thoroughly refashioned the material and competitive logics of gallium nitride. It received considerable attention in the compound

43. Amano, interview (ref. 39); Amano, "Passion and Struggle" (ref. 39); Akasaki, "Insight, Challenge, and Success" (ref. 28); Akasaki, "Progress in Crystal Growth" (ref. 28); Akasaki and Amano, "Breakthroughs" (ref. 28); Akasaki, "Key Inventions" (ref. 28).

44. In the late 1980s, Akasaki and Amano did not understand why e-beams worked. It was years later that researchers found that electron beams generated heat that eliminated hydrogen from single crystals of gallium nitride.

45. Amano, interview (ref. 39); Amano, "Passion and Struggle" (ref. 39); Akasaki, "Insight, Challenge, and Success" (ref. 28); Akasaki, "Progress in Crystal Growth" (ref. 28); Akasaki and Amano, "Breakthroughs" (ref. 28); Akasaki, "Key Inventions" (ref. 28).



semiconductor community and was widely seen as paving the way to LED-based displays and to blue lasers for consumer electronics. It was at that time that gallium nitride grew into a major research field, attracting hundreds of researchers working in industry. For example, Maruska, who had pioneered single crystals of gallium nitride, returned to the material he had created and worked on gallium nitride at Spire, a Massachusetts-based start-up in the early 1990s. Many academic research groups moved into gallium research as well. According to a project director in the Pentagon's Office of Naval Research, three quarters of the university researchers who had worked on gallium arsenide in the 1980s had switched to gallium nitride by 1992.<sup>46</sup>

One can ask why Akasaki succeeded in growing high-quality gallium nitride crystals and making blue PN junction LEDs, whereas many others had tried and failed to do so during the previous twenty years. Akasaki's success in "taming" the material logic of gallium nitride crystals had a lot to do with his tenacity, his obsession with gallium nitride, and his staying power. Akasaki was a high-status researcher within the Japanese compound semiconductor community, which enabled him to get access to critical resources. He garnered financial support from Matsushita's management and from the firm's founder. Grants from MITI and the Ministry of Education were also essential for building a competency in the growth of gallium nitride crystals. Through his contacts, Akasaki could use state-of-the-art equipment located at other firms and universities. He surrounded himself with talented researchers such as Hiroshi Amano. Also important was his constant diversion of financial resources from his larger projects on other compound semiconductors and their redirection toward high-risk studies on gallium nitride at times when there was no outside funding for research on this material.

To solve the riddles of gallium nitride, Akasaki and his collaborators used all the armentarium of compound semiconductor researchers: buffer layers, ion implantation, electron beams, HVPE, MBE, and MOCVD (these techniques had been developed for the growth and doping of other crystals, including silicon, gallium arsenide, and gallium arsenide phosphide). It was by "throwing" these well-understood techniques at gallium nitride that Akasaki and his group overcame key obstacles in their quest for blue PN junction

46. For the public announcement of the blue PN junction diode in the United States, see "Diode a Step Toward 'Flat TV,'" *Tulsa World*, 29 Nov 1989; "TVs May Soon Hang on Walls Like Paintings," *San Diego Union-Tribune*, 11 Dec 1989; Peter Spotts, "New Semiconductors Lie at the End of the Rainbow: Blue LEDs and Lasers May Revolutionize Lighting and Video Displays," *Christian Science Monitor*, 28 Feb 1995.

LEDs. Through a process of trial and error, they found a complex of techniques and materials—MOCVD, aluminum nitride buffers, electron beams, and magnesium-based organometallic compounds—that produced very clear crystals and P-type gallium nitride.

## THE COMMERCIALIZATION OF GALLIUM NITRIDE

The research of Akasaki, Amano, and their group was commercialized by two Japanese corporations, Nichia Chemical Industries and Toyoda Gosei Co., in the early and mid-1990s. These firms were unlikely candidates for the commercialization of gallium nitride and blue LEDs. They operated in very different technologies and industries. Nichia Chemical, founded in 1956, was a medium-sized chemical corporation producing phosphors for fluorescent lamps and color televisions. The company was located in a rural region in the far-off island of Shikoku. Similarly, Toyoda Gosei had very little prior experience with LEDs and compound semiconductors. It was a manufacturer of automotive components. Established as Nagoya Rubber Co. immediately after World War II, Toyoda Gosei was a subsidiary of the Toyota Motor Corporation. It produced tires and plastic components such as car door handles for the automotive giant.<sup>47</sup>

Competition between Nichia and Toyoda Gosei, along with material and market logics, shaped the further development of gallium nitride over the next fifteen years. The firms approached the engineering of gallium nitride crystals and the commercialization of blue LEDs in strikingly different ways. Toyoda Gosei's technologists collaborated closely with Akasaki and Amano and obtained funding from an agency of the Japanese government to finance technology transfer from Nagoya University. They also focused at first on the production of MIS diodes. In contrast, Nichia aggressively appropriated the techniques developed by Akasaki and his group—without any formal agreement with the university and in direct competition with it and its corporate partner. The engineering group at Nichia was also more aggressive technically and concentrated from the start on the mass production of PN junction LEDs.

It was Toyoda Gosei that first entered the field of gallium nitride and blue LEDs and it did so in close collaboration with Akasaki and his laboratory. In 1985, Akasaki gave a talk at the Nagoya Chamber of Commerce and Industry,

47. Koichi Ota, interview by Takahiro Ueyama, 10 Dec 2010.

an organization promoting the industrial growth of the Chubu region around Nagoya. In his talk, Akasaki discussed the development of the School of Engineering at Nagoya University and its research program on silicon-based integrated circuits. In the last five minutes of his talk, Akasaki discussed his own research on gallium nitride crystals and the great potential that these crystals held for the making of blue LEDs (Akasaki and Amano were then working on the low-temperature buffer of aluminum nitride to fabricate very clear gallium nitride crystals). This short discussion piqued the interest of Toyoda Gosei researchers who attended Akasaki's talk. They soon approached Akasaki, asking him to initiate a research collaboration on gallium nitride and blue LEDs with their firm. Working for an automotive component supplier, they were well aware of the market logic of light emitting diodes. Toyota used red LEDs in its cars. Toyoda Gosei engineers also understood the commercial potential of high-quality blue LEDs. The news of Akasaki's talk quickly spread to other Japanese firms, some of which made similar entreaties to Akasaki and proposed to work with him on blue LEDs.<sup>48</sup>

After nearly a year of discussions and negotiations, Akasaki, who was keenly interested in commercializing his research, settled on Toyoda Gosei as his exclusive partner.<sup>49</sup> But Toyoda Gosei itself was far from being united on the question of whether to expand into blue LEDs. The upper managers of Toyoda Gosei were interested in diversifying into other markets and industries, as were many other Japanese firms at that time. The mid-1980s were the height of the Japanese "bubble economy." Many Japanese corporations diversified then into new businesses to grow their sales and maximize their profits. Before the discussions with Akasaki, Toyoda Gosei's managers had identified LEDs as one of the thirty fields into which their corporation could diversify. But they were concerned that Toyoda Gosei did not have prior experience in semiconductors and that investments in gallium nitride research would be very large. Diversifying into gallium nitride and blue LEDs required that the firm recruit

48. Akasaki, interview (ref. 24); Ota, interview (ref. 47).

49. It is not known why Isamu Akasaki did not associate himself with Matsushita, his former employer, to commercialize gallium nitride. It is clear, however, that Akasaki maintained good relations with Matsushita after his departure to Nagoya University. For instance, he chose his successor at Matsushita Research Institute Tokyo and was on good terms with Konosheke Matsushita, the firm's founder. One can speculate that, after the failed commercialization of MIS LEDs, managers at Matsushita doubted that Akasaki's work at the University of Nagoya would ever lead to luminous PN junction LEDs. One can also note that MRIT was an "academic" organization with limited interest in the commercialization of research. Akasaki, interview (ref. 24).

compound semiconductor specialists, build a large clean room, and purchase expensive MOCVD reactors.<sup>50</sup>

Seeing the reluctance of Toyoda Gosei's upper managers to commit their own funds to the project, Akasaki persuaded them to jointly apply for financing from the Research Development Corporation of Japan. The Research Development Corporation (now called the Japan Science and Technology Agency) funded joint projects between academia and industry, with the goal of transferring university inventions to the private sector. It had supported technology transfer in a wide range of areas, including red LEDs, batteries, gas sensors, and magnetic materials. In March 1987, Akasaki and Toyoda Gosei obtained a large grant from the Research Development Corporation in the amount of 550 million yens (roughly \$6 million). This grant was initially for three years and was then extended for a fourth one. The grant was for the development of the manufacturing technology for gallium nitride blue LEDs. In case Toyoda Gosei succeeded in introducing these LEDs to the market and making a profit, it would have to repay the full amount of the grant to the Research Development Corporation. If the project was unsuccessful, the Research Development Corporation would write-off its investment. In other words, the financial risk for Toyoda Gosei was nil. But the technological risks were great as Akasaki and Amano had not yet made PN junction LEDs.<sup>51</sup>

The grant from the Research Development Corporation financed the building of a capability in gallium nitride crystals at Toyoda Gosei. It also enabled Akasaki to purchase a second MOCVD reactor for his lab and it supported part of his research on P-type gallium nitride. Toyoda Gosei recruited compound semiconductor specialists from Nagoya University and other universities. These staff familiarized themselves with the art and science of gallium nitride by working with Akasaki's original MOCVD reactor and by participating in seminars in the Akasaki laboratory.<sup>52</sup>

50. Ota, interview (ref. 47); Akasaki, interview (ref. 24).

51. Before making the decision to move into gallium nitride LEDs, Toyoda Gosei did a patent search that showed that no patent on gallium nitride had been filed since the early 1980s. This finding reassured Toyoda Gosei's management and encouraged them to move into gallium nitride. Ota, interview (ref. 47); Akasaki, interview (ref. 24); Japan Science and Technology Agency, "Contract Development since 1958," n.d., at <http://www.jst.go.jp/itaku/result50then.pdf> (accessed 27 Oct 2011).

52. Upper managers of Toyoda Gosei later surmised that the company would have never gone into gallium nitride had compound semiconductor researchers worked at the firm before the agreement with Akasaki. These researchers would have strongly advised against such a risky venture. Ota, interview (ref. 47); Akasaki, interview (ref. 24); Amano, interview (ref. 39).

Toyoda Gosei's engineers soon encountered significant difficulties in making high-quality crystals of gallium nitride. These difficulties led Toyoda Gosei's engineering managers to focus their gallium nitride program on the production of MIS light emitting diodes (even after Akasaki and Amano had succeeded in fabricating junction LEDs). They wanted to develop manufacturing technologies for MIS LEDs, before addressing the more difficult problem of PN junction LED fabrication. In their view, expertise acquired with MIS LEDs would enable them to produce PN junction light emitting diodes. This conservative approach to innovation seems to have led to tensions with Akasaki, Amano, and their group, who wanted to take more risks and develop a manufacturing process for the volume production of PN junction LEDs. Toyoda Gosei's managers prevailed and Akasaki and Amano collaborated with the firm's engineers to design a new device structure and fabrication process for MIS LEDs that would take advantage of recent advances in the growth of gallium nitride crystals.<sup>53</sup>

The MIS LED designed by Akasaki, Amano, and Toyoda Gosei's engineers was reminiscent of Akasaki's earlier work at Matsushita. They incorporated the electrodes inside the semiconductor crystal. To form these electrodes, they used the same materials (including silicon oxide) as the ones Akasaki had previously employed at Matsushita. But, unlike prior art at Matsushita, Akasaki and the Toyoda Gosei team used the crystal growing techniques recently developed at Nagoya University to make MIS LEDs. This creative redesign of the Matsushita MIS LED and the process for making it enabled the Toyoda Gosei engineers to make luminous MIS LEDs (with one fifth of a candela, the unit of luminous intensity). By October 1993, the development of the blue MIS LED and the related production processes had advanced to the point where Toyoda Gosei announced in the trade press that it would bring it to volume production the following year. The firm and Nagoya University patented this work, filing more than ten patents on MIS LEDs and related buffer technology.<sup>54</sup>

But before Toyoda Gosei could introduce its MIS LED to the market, Nichia Chemical, the phosphor-making company based in Shikoku, made

53. Akasaki, interview (ref. 24); Amano, interview (ref. 39); Ota, interview (ref. 47).

54. Akasaki, interview (ref. 24); Ota, interview (ref. 47); Katsuhide Manabe, Nobuo Okazaki, Isamu Akasaki, Kazumasa Hiramatsu, and Hiroshi Amano, "Gallium Nitride Group Semiconductor and Light Emitting Diode Comprising It and the Process of Producing the Same," U.S. Patent 5,218,216, filed 20 Dec 1991 and issued 8 Jun 1993; "Toyoda Gosei to Mass Produce Gallium Nitride Diodes," *Japan Chemical Week*, 28 Feb 1991; "Japanese Beat Blue Obstacle: Toyoda Gosei," *Electronics Times*, 28 Oct 1993.

a startling announcement. At a press conference in November 1993, it announced that it was producing and marketing a blue PN junction LED, which was five times more luminous than the LED recently publicized by Toyoda Gosei.<sup>55</sup> This announcement took everybody by surprise.<sup>56</sup> It also forced Toyoda Gosei to abandon their MIS LED project, focusing all efforts on PN junction light emitting diodes. Nichia's surprise commercialization of blue LEDs was based on four years of feverish work by Shuji Nakamura and his collaborators, where they had appropriated inventions made by the Akasaki laboratory and other research groups in Japan (without much concern for intellectual property) and turned them into stable manufacturing processes.<sup>57</sup>

Nakamura had joined Nichia's development laboratory upon graduation with a master's degree from the University of Tokushima, a provincial university. At the request of Nobuo Ogawa, the firm's founder, who was interested in diversifying from phosphors into light emitting diodes, Nakamura had worked for nearly ten years on the growth of various compound semiconductor crystals. He had also made red and yellow LEDs. Aware of the development of high-quality gallium nitride crystals at Nagoya University, Nakamura convinced Ogawa to reorient the firm's compound semiconductor program toward gallium nitride. This represented a risky gamble, given the complexities of gallium nitride technology. To master the material logic of gallium nitride crystals, in 1988 Nichia purchased an expensive MOVCD reactor, and sent Nakamura to the University of Florida to learn how to use it.<sup>58</sup>

Upon his return from Florida, Nakamura, who in the words of an associate "had fallen in love with gallium nitride," sought to outcompete the Akasaki laboratory and Toyoda Gosei by improving upon their laboratory techniques

55. The original price of blue LEDs was \$18.75 a piece, whereas most red and yellow LEDs cost pennies at the time. The price rapidly fell to \$5 and then to \$1 per LED.

56. The group at Toyoda Gosei was aware that Nichia was doing research on gallium nitride and blue LEDs, but it was not aware that it was so close to the goal of making PN junction LEDs. Amano, interview (ref. 47).

57. Miller Freeman, "Blue Lights Up Record: Nichia Chemical," *Electronics Times*, 9 Dec 1993. In 1986, Nakamura attended a seminar, where Akasaki presented his work on low-temperature buffer technology. Akasaki, interview (ref. 24).

58. Shuji Nakamura, "Breakthrough in Commercialization of Gallium Nitride Blue Light Emitting Devices," at <http://www.takeda-foundation.jp/en/award/takeda/2002/forum/03a.html> (accessed 21 Oct 2011); Johnstone, *Brilliant* (ref. 4); Shuji Nakamura, "Aoiro Hakko Daiodo No Kaihatsu," *Electrochemistry* 63 (1995): 897–901; Shuji Nakamura, "InGaN/AlGaIn Hakko Daiodo No Genjo To Seinou Kajo," *Kogaku* 24 (1994): 701–08; Shuji Nakamura, "InGaN Blue-Light-Emitting Diodes [in Japanese]," *Journal of the Institute of Electronics, Information and Communication Engineers* 76, no. 9 (1993): 913–17.

and transforming them into mass production processes.<sup>59</sup> He did so with internal funds and by working in relative isolation. Over a three-year period, Nakamura and his collaborators transformed the material logic of gallium nitride by developing a manufacturing process for blue LEDs. This process rested on three innovations: the development of a novel MOCVD reactor design; a new method for doping gallium nitride with P-type elements; and the deposition of a film of indium gallium nitride between gallium nitride layers. Inspired by the development of new MOCVD techniques for the deposition of aluminum nitride by Kazuo Tsubouchi, a researcher at Nagoya University, Nakamura designed the two-flow MOCVD reactor to grow single crystals of gallium nitride in the summer of 1990. In traditional MOCVD reactors (such as those used at Toyoda Gosei), gases carrying the reacting chemicals flowed perpendicular or diagonally to the sapphire substrate where the gallium nitride films were grown. In Nakamura's design, the reactor had two gas flows: a flow of reactant gases parallel to the sapphire substrate; and a perpendicular flow of inert gases that forced the chemicals of the reactant gases to flow in close contact to the substrate. The sapphire substrate was also placed on a rotating disk, which evened out exposure to the reactant gases over the whole surface. The two-flow MOCVD reactor enabled the growth of films of gallium nitride that were more continuous, uniform, and defect-free than those deposited in regular MOCVD reactors. These films also had superior electrical and luminescent characteristics.<sup>60</sup>

In conjunction with the development of improved crystal-growing techniques, Nakamura and his group invented a simpler and more economical method for P-type doping than the electron beams used by Akasaki and Amano. It was too slow and expensive to shoot electrons beams at gallium nitride crystals to alter their conductivity. The technique was appropriate in a laboratory setting, but it was not practical for volume production. By chance, Nakamura and his collaborators found that heating gallium nitride crystals in a nitrogen atmosphere at low temperatures (annealing in technical parlance)

59. Gerhard Fasol, "Room-temperature Blue Gallium Nitride Laser Diode," *Science*, 21 Jun 1996.

60. Shuji Nakamura, Yasuhiro Harada, and Masayuki Seno, "Novel Metalorganic Chemical Vapor Deposition System for GaN Growth," *Applied Physics Letters* 58 (1991): 2021–23; Shuji Nakamura, "Method of Vapor-growing Semiconductor Crystal and Apparatus for Growing the Same," European Patent Application 91118176.6, filed 29 Apr 1992; Nakamura, "Breakthrough in Commercialization" (ref. 58); Akasaki, interview (ref. 24); Amano, interview (ref. 39); Johnstone, *Brilliant!* (ref. 4).

had the same results as shooting e-beams on them. They could make positively charged gallium nitride and do so at low cost.<sup>61</sup>

The third important production method developed by Nakamura and his group was the growing of indium gallium nitride, an alloy of indium nitride and gallium nitride, on top of a gallium nitride layer (to create a sandwich of a film of indium gallium nitride between two layers of gallium nitride). The development of this process at Nichia was inspired by the work of Takashi Matsuoka, a researcher at Nippon Telegraph & Telephone Corp. (NTT)<sup>62</sup> who had grown indium gallium nitride in his laboratory and had given several talks about it that Nakamura had attended, including one at Nichia. Building on Matsuoka's techniques, Nakamura and his group developed a process for manufacturing heterostructure LEDs composed of different layers of gallium nitride and indium gallium nitride. The process, patented by Nichia, yielded bright LEDs that emitted a pure blue. It was these LEDs that Nichia brought to the market in late 1993.<sup>63</sup>

The commercialization of high-quality LEDs reconfigured the market and competitive logics surrounding gallium nitride. Nichia quickly reaped the commercial benefits of its product introduction and became a high-volume producer of gallium nitride crystals and blue LEDs. Markets for blue LEDs

61. Shuji Nakamura, Nahurito Iwasa, and Mayasuki Senoh, "Method of Manufacturing P-type Compound Semiconductor," European Patent Application 923110132.3, filed 5 Nov 1992. Interestingly, the idea of annealing wide band gap materials such as gallium nitride to improve their luminescent properties was not original to Nakamura and his group. It had been proposed a few years earlier by Gertrude Neumark Rothschild, a faculty member at Columbia University (she received a patent for this work in 1993). In her patent, Neumark Rothschild, a physicist who had spent much of her career in industry at Sylvania and then at Philips, argued that removing hydrogen from wide band gap materials, by heating them up, would lead to the fabrication of good PN junctions and make the fabrication of high-quality blue, green, and violet LEDs possible. See Gertrude Neumark Rothschild, "Wide Band-Gap Semiconductors Having Low Bipolar Resistivity and Method of Formation," U.S. Patent 5,252,499, filed 15 Aug 1988 and issued 12 Oct 1993; William Grimes, "Gertrude Rothschild, Dies at 83; Advanced LEDs," *New York Times*, 17 Nov 2010.

62. T. Nagatomo at Shibaura Institute of Technology had started to use this process before Matsuoka. Akasaki, interview (ref. 24).

63. Up until late 1993, Nichia had protected its intellectual property through trade secrets. It started filing patents with Nakamura's work on gallium nitride. See Shuji Nakamura, Takashi Mukai, and Naruhito Iwasa, "Light-emitting Gallium Nitride-based Compound Semiconductor Device," European Patent Application 98100538.2, filed 19 Nov 1993; Shuji Nakamura, Takashi Mukai, and Nahurito Iwasa, "Light-emitting Gallium Nitride-based Compound Semiconductor Device," U.S. Patent 5,880,486, filed 30 Aug 1996 and issued 9 Mar 1999; Akasaki, interview (ref. 24); Amano, interview (ref. 39); Johnstone, *Brilliant!* (ref. 4).



emerged among manufacturers of billboards and outdoor displays, at first in the United States and Europe, and then in Japan.<sup>64</sup> Nichia's LEDs were also increasingly used in traffic lights and highway signs. To meet growing demand, the corporation built a new factory and hired hundreds of operators, technicians, and engineers. These investments and tight control of manufacturing processes enabled Nichia to produce blue LEDs in large quantities. Its output reached two million blue LEDs a month in the spring of 1995. By that time, Nichia's sales of LEDs were on the order of \$22 million a year.<sup>65</sup>

Nichia's monopoly on high-quality blue LEDs was soon broken by Toyoda Gosei. After Nichia's product announcement in 1993, Toyoda Gosei's managers reoriented their resources toward the manufacture of junction LEDs. With the technical support of Akasaki and Amano, the firm's engineers developed a new process that resembled Nichia's. In a few years, they mastered the mass production of PN junction diodes. In 1995, the corporation introduced its own blue PN junction LEDs to the market. Fierce competition soon ensued between the two firms. To gain the upper hand in the fast growing gallium nitride business, engineers at Nichia and Toyoda Gosei extended their firms' product lines. They developed and commercialized green and blue-green LEDs.<sup>66</sup> Nichia soon produced white LEDs as well, by coating its blue LEDs with phosphor. In addition, Nichia and Toyoda Gosei raced to the production of brighter LEDs. For example, in 1995 Nichia doubled the luminosity of its blue LEDs, matching the brightness of the light emitting diodes recently introduced to the market by Toyoda Gosei. Two years later, Nichia announced that it was boosting luminosity by another fifty percent. Toyoda Gosei soon followed suit. By 1999, Nichia was the market leader in gallium

64. The first customers for Nichia's blue LEDs were European and American firms, such as Sylvania and Philips. Japanese companies refused at first to purchase LEDs from Nichia, because it was an untried LED maker. See "The Man Who Broke the Light Bulb," *Independent on Sunday*, 10 Oct 1999.

65. Nichia obtained very high manufacturing yields (eighty percent) in 1994. "Vendors Target Elusive Blue LEDs," *Electronic News*, 7 Mar 1994; Yoshiko Hara, "Bright Blue LED Could Enable Color Displays," *Electronic Engineering Times*, 21 Mar 1994; Robert Service, "Structure Meets Function at Materials Gathering," *Science*, 22 Apr 1994; Kathy Rogers, "Future for Blue LEDs Is Shining Brightly," *Electronic Engineering Times*, 7 Aug 1995; Dennis Normile, "The Future Looks Bright for Blue LEDs," *Popular Science*, 1 Oct 1995; Andrew Pollack, "The Little Lights that Could," *New York Times*, 29 Apr 1996; Johnstone, *Brilliant!* (ref. 4).

66. Engineers at Nichia and Toyoda Gosei fabricated green and blue-green LEDs by changing the quantity of indium in the crystals.

nitride-based LEDs with \$287 million in sales. But Toyoda Gosei was a close second. Its sales of LEDs reached \$168 million that same year.<sup>67</sup>

Competition between Nichia and Toyoda Gosei extended to the courtroom. Determined to drive Toyoda Gosei out of the market, Nichia sued its competitor for patent infringement and demanded that it stop shipping blue LEDs in the summer of 1996. Nichia's management was confident that they held a much stronger intellectual property position than Toyoda Gosei, as most of Toyoda Gosei's patents were related to MIS LEDs, an obsolete technology. At first, Toyoda Gosei attempted to reach a settlement and proposed a cross-licensing agreement with Nichia (which was the standard practice in Japan at that time). But as Nichia refused, Toyoda Gosei countersued, claiming infringement of their patents. Toyoda Gosei also asked the courts to ban Nichia from manufacturing and selling gallium nitride LEDs.<sup>68</sup>

A bitter legal battle between the two firms ensued, involving tens of lawsuits over a six-year period. The patent war was so fierce that Toyoda Gosei sued the Japanese Patent Office for granting patents on gallium nitride to Nichia (Toyoda Gosei won these cases). By 2002, neither Nichia nor Toyoda Gosei had succeeded in driving their rival out the gallium nitride business. This

67. Profit margins on gallium nitride blue and green LEDs were high at Nichia—on the order of fifty percent. Toyoda Gosei started to turn a profit in 1998. “Man Who Broke” (ref. 64); Ota, interview (ref. 47); Japan Science and Technology, “Contract Development” (ref. 51); “Pure-Green LED Built Using CVD,” *Electronic Engineering Times*, 28 Aug 1995. Nichia and Toyoda Gosei also competed in the development of blue laser diodes. Blue laser diodes offered great commercial opportunities. They were widely viewed as having the potential to increase the memory capacity of compact disks by a factor of four and thereby opening up a large market in consumer electronics. Developing these diodes was particularly challenging as research groups needed to learn how to direct the light generated by LEDs into a single beam. In the second half of the 1990s, Toyoda Gosei's engineers in collaboration with Akasaki's laboratory and with funding from the Research Development Corporation tackled this problem, as did Nakamura's group at Nichia. It was again the Nichia group that brought the first gallium nitride-based laser to the market. “Japan's Nichia Chem Unveils Purple Chip Laser for DVD Players,” *Asia Pulse*, 20 Jun 2000; “GaN Semiconductor Laser Emits Blue-violet Beam,” *Japan Chemical Week*, 17 May 2001.

68. Nichia's aggressive protection of its intellectual property did not drive Toyoda Gosei out of the market. But it prevented other Japanese firms from entering the gallium nitride LED business in the 1990s. Ota, interview (ref. 47); “Man Who Broke” (ref. 64); Yoshiko Hara, “Laser, LED Developer Dreams in Color,” *Electronic Engineering Times*, 24 Apr 2001; “Nichia Kagaku To Toyoda Gosei Ga Wakai: Aoire LED No Senjo Wa Hotei Kara Sijo E” *Nikkei Electronics* 832 (2002): 59–66; “Aoire Hakkou Daiodo No Tokkyo Funso,” *Nikkei Electronics* 779 (2000): 49–54; “Hakko Daiodo: Nichia Kagaku To Toyoda Gosei No Tokkyo Funso,” *Nikkei Electronics* 736 (1999): 47–52.

realization led the firms to settle their patent dispute and to cross-license their technology. They were also encouraged to do so by growing competitive pressures coming from other corporations. By that time, no less than 180 firms worldwide were engaged in gallium nitride development and production and more than 290 universities and public research centers performed research on the material. Among Nichia and Toyoda Gosei's main industrial competitors were Osram Opto Semiconductors, a German corporation; Cree, a North Carolina-based manufacturer of LEDs; and Lumileds, a joint venture of Philips and Hewlett-Packard (now called Philips Lumileds, the parent company of Philips Lighting North America). American firms that had been late to understand the potential of gallium nitride were particularly aggressive in acquiring competency in it. With funding from DARPA, they launched massive R&D programs on the new material, re-creating the manufacturing processes used at Nichia and Toyoda Gosei. Gallium nitride had become a large business indeed, and Nichia and Toyoda Gosei needed to invest all their resources on product development and manufacturing to maintain their position in it.<sup>69</sup>

Benefiting from the settlement between Nichia and Toyoda Gosei and the rapid growth of the gallium nitride business was Nagoya University. After the Japanese parliament enacted its version of the Bayh-Dole Act in 2000, universities gained ownership of patents obtained from research projects funded by the Japanese government. As a result, the patents on the fabrication of high-quality gallium nitride filed by Akasaki and his laboratory came into the possession of the university. Added to these patents were patents filed jointly with Toyoda Gosei. As more and more firms licensed the Akasaki patents, Nagoya University received a growing stream of royalties.<sup>70</sup> For example, in 2003, it received \$5 million. This represented nearly ninety percent of all royalties received by national universities during that year. Royalties from the

69. Ota, interview (ref. 47); "Japan's Nichia Chem Wins LED Patent Case against Toyoda Gosei," *Asia Pulse*, 1 Sep 2000; "Japan's Toyoda Gosei Wins 4th Blue LED Patent Case," *Asia Pulse*, 10 Oct 2001; "Toyoda Gosei Files Damages Suits against Nichia over LED," *Japan Weekly Monitor*, 22 Oct 2001; Edmund Klamann, "Nichia, Toyoda Gosei Aim to End Blue Laser Spat," *Reuters News*, 9 Aug 2002; Yoshiko Hara, "Blue-LED Rivals Call Truce as Third Parties Wage Competitive Assault," *Electronic Engineering Times*, 2 Sep 2002; "The Future's Blue: Four Years Ago, Gallium Nitride Was a Mere Curiosity—Now Everybody Wants a Piece of It," *The Guardian*, 31 Jul 2003.

70. Many firms, including Toyoda Gosei, also signed a licensing agreement with Gertrude Neumark Rothschild of Columbia University for her patent on annealing. Grimes, "Gertrude Rothschild" (ref. 61); Ota, interview (ref. 47).

gallium nitride patents supported the creation of a new semiconductor research institute founded in Akasaki's honor at Nagoya University. They also financed numerous research projects in other disciplines within the university. Thus, a material technology originally developed at RCA and advanced at Nagoya University had become the center of a large and dynamic industry and had generated funding streams that fed back into the university and supported research on semiconductors and other materials.<sup>71</sup>

## CONCLUSION

Materials innovation is often a lengthy process. But it was particularly long and protracted in the case of gallium nitride. The development of single crystals of gallium nitride took nearly four decades from Maruska's early work in the late 1960s to the worldwide production of gallium nitride-based LEDs in the first decade of the twenty-first century. Maruska and Pankove pioneered the development of single crystals of gallium nitride and blue LEDs at RCA. They did so within a Cold War context. They funded their research with grants from the DoD and NASA and relied on material technologies such as sapphire substrates that had been specifically engineered for military applications. In the mid-1970s, the locus of innovation shifted to Japan. At the Matsushita Research Institute and later at Nagoya University, Akasaki and his colleagues developed new crystal-growing techniques and a method for doping these crystals. Capitalizing on these material advances, they fabricated PN junction blue LEDs. Nichia Chemical and Toyoda Gosei commercialized Akasaki's breakthroughs and introduced high-quality blue LEDs to the market in the mid-1990s—more than twenty-five years after the growth of the first single crystals of gallium nitride at RCA. Gallium nitride, which had been developed by a small group of people, became a worldwide industry as Japanese, American, European, and Taiwanese corporations entered the field. Continuous development of gallium nitride by these organizations enabled the engineering of solid-state lamps and their widespread adoption in the first decade of the twenty-first century.

71. The Japanese government benefited financially from its investment in research at Nagoya University and Toyoda Gosei. The government received \$46 million in royalties for a patent jointly filed by Akasaki and Toyoda Gosei. This represented ninety percent of all royalties that the Japanese government received in the 1990s. Ota, interview (ref. 47).

Several lessons can be drawn from this examination of gallium nitride for the understanding of materials innovation. First, the engineering of new materials has an emotional dimension. Along with market logic, the emotional attachment and fascination of a few scientists and engineers for gallium nitride helped sustain the development of this material over the first twenty-five years of its existence. Maruska felt a deep bond to gallium nitride. Akasaki and Amano were obsessed with it and, according to a close associate, Nakamura “fell in love” with the material. The importance of emotional commitments in the history of gallium nitride is not specific to this material. Similar phenomena occurred in the creation and use of other materials as well. For example, Thomas E. Edison was deeply attached to carbon and used it in many of his inventions including electric lights, telephones, and telegraphic devices. Similarly, in the early history of semiconductors, Gordon Teal, a metallurgist working at the Bell Telephone Laboratories, developed what he called a “sentimental attachment” to germanium. In the late 1940s, Teal fabricated single crystals of germanium which permitted the fabrication of junction transistors and enabled the early growth of the semiconductor industry.<sup>72</sup>

Second, the history of gallium nitride shows the role of the constant interplay among several contextual logics—material logic, market logic, and competitive logic—in materials innovation. The history of gallium nitride is to a large degree a story about Maruska, Akasaki, and Nakamura’s contention with material logic. Over a forty-year period, they consistently faced the same challenges: make very clear, high-quality single crystals; change the conductivity of these crystals; and boost their light emitting properties. To solve these difficult problems, they used material capabilities—tools, instruments, and techniques—engineered for other compound semiconductors. The development of gallium nitride was in part the search for the right combination of tools, instruments, and techniques to “tame” this material technology and make it yield bright light emitting diodes.

But the story of gallium nitride is also a story about market logic. The primary impulse behind Maruska, Akasaki, Amano, and Nakamura’s work was a commercial one. Along with their supporters in industry and government, these men saw gallium nitride and blue LEDs as the key to the opening of large markets in consumer electronics and the lighting industry. In their view, the making of blue light emitting diodes would enable the fabrication of

72. Lécuyer and Brock, “Materiality of Microelectronics” (ref. 6); Paul Israel, *Edison: A Life of Invention* (New York: John Wiley and Sons, 1998).

flat panel displays. When properly coated with phosphors, blue LEDs would compete with established lighting technologies. They would also be critical to a new type of laser, the blue laser, and open vast technological and commercial opportunities in compact discs and other recording technologies. Along with the innovators' deep emotional attachment to the material, it was market logic that carried the development of gallium nitride across the whole period.

Competitive logic—the competition among laboratories, firms, and nations—also shaped gallium nitride technology. The war waged by Nichia Chemical and Toyoda Gosei accelerated material advancements and led to the fabrication of increasingly brighter blue LEDs and to the engineering of white, green, and violet light emitting diodes as well. Competition between the United States and Japan also informed the history of gallium nitride. If RCA abandoned the material it had created, it was partially due to competition with Japanese firms that robbed it of its markets in consumer electronics. Similarly, the adoption of gallium nitride at Matsushita was part of a larger movement whereby Japanese firms took American inventions, improved upon them, and brought them to mass production. Governmental agencies such as MITI fostered and supported these efforts, waging war on the U.S. semiconductor industry. Conversely, in the mid-1990s, American firms and universities went aggressively into gallium nitride technology. They brought significant competitive pressures on Nichia Chemical and Toyoda Gosei and forced these firms to settle their disputes and refocus their resources on the reinforcement of their own material competencies.

The development of gallium nitride was shaped by several longstanding regularities. One of these regularities was the emotional power of materials which drew a small cadre of talented researchers to gallium nitride. Another regularity was the complex of challenges, constraints, and opportunities that scientists and engineers faced in three contexts: materials, material competencies, and their obduracy; users and markets; and the competitive tensions among laboratories, firms, and nations. It was at the convergence point of the emotional spell of materials and these contextual logics that novel material configurations emerged, evolved, and found new uses and new users.

## ACKNOWLEDGMENTS

Christophe Lécuyer and Takahiro Ueyama would like to thank the Collegium de Lyon, the Japan Society for the Promotion of Science, the Chemical Heritage Foundation,

and the Institute for Advanced Study at Central European University for supporting their research on the history of gallium nitride and the writing of this article. Their thanks also go to David C. Brock, Hyungsub Choi, Martin Collins, David Hounshell, Paul Israel, Ann Johnson, Patrick McCray, Cyrus Mody, and Carsten Reinhardt for their insightful comments on earlier versions of this article.