

Special Article

Development of Holography Electron Microscope with Atomic Resolution

—Triumph over Numerous Difficulties—

Hiroyuki Shinada, Ph.D.

The development of an atomic-resolution holography electron microscope that began in earnest in March 2010 has now been completed with the achievement of world-leading resolution. This article describes the background leading up to the proposal for the microscope's development, the progress of that development with support from the FIRST Program, the features of the instrument, and its future applications. Beset, at first, by a series of unexpected misfortunes and setbacks that included the death by illness of the engineer leading the development shortly after the work commenced, the Great East Japan Earthquake, and the death of Dr. Tonomura, the lead researcher and originator of the project proposal, the development then had to overcome technical difficulties associated with high voltages before it finally succeeded in achieving a world-leading resolution of 43 pm in December 2014. In the future, Hitachi aims to use the microscope to accelerate the development of new functional materials through open innovation.

INTRODUCTION

THE transmission electron microscope was invented in Germany in 1932. It works by directing a beam of electrons through a specimen and then using a lens to enlarge the image formed by their passage. In Japan, development and research into its applications began in 1939, and numerous companies around the world have competed to develop the instruments since Hitachi developed its own first transmission electron microscope, the HU-1, in 1941. Hitachi supplied the first commercial model produced in Japan, the HU-2, to Nagoya Imperial University (as it was then known) in 1942⁽¹⁾. A transmission electron microscope exhibited by Hitachi at the 1958 World's Fair in Brussels won a grand prize⁽²⁾, and since then Japanese instruments, including those made by Hitachi, have competed with the world's best⁽³⁾.

Subsequently, the scanning electron microscope (SEM), which works by scanning a focused electron beam over the surface of the specimen, was successfully commercialized in 1965. The electron microscopes of that time used thermo electron guns, which are based on the thermionic emission of electrons by heating tungsten. Although the field emission effect (whereby electrons are emitted via the tunneling effect by applying a strong electric field to a sharp metal tip) was a known source for obtaining the electrons used by these microscopes, it had not been adopted in practice because it required extreme high vacuum, making it difficult to use in practice.

With assistance from Professor Albert V. Crewe of the University of Chicago, Hitachi successfully implemented a practical electron source, and an electron gun to use it, and incorporated it into an SEM that was released on the market in 1972. From 1984 onwards, this technology became part of a major business dealing with critical dimension-scanning electron microscopes (CD-SEMs) specifically designed for measuring semiconductor patterns, a product category in which Hitachi continues to have the leading share of the global market⁽⁴⁾.

Returning to the subject of transmission electron microscopes, since increasing the energy of the electron beam increases its ability to pass through specimens, the 100-keV energy of early instruments had increased ten-fold to 1 MeV^{(5), (6)} by 1966 to enable the observation of thicker specimens, with the subsequent development of larger models up to 3 MeV^{(7), (8)}. As the history of the electron microscope is a long one that has been well documented elsewhere, this article will now move on to the subject of holography electron microscopes.

DR. TONOMURA AND HOLOGRAPHY ELECTRON MICROSCOPY

Early Years of Holography Electron Microscopy

To many people, the term “holography” will bring to mind the anti-counterfeiting holograms incorporated into credit cards or banknotes. However, the

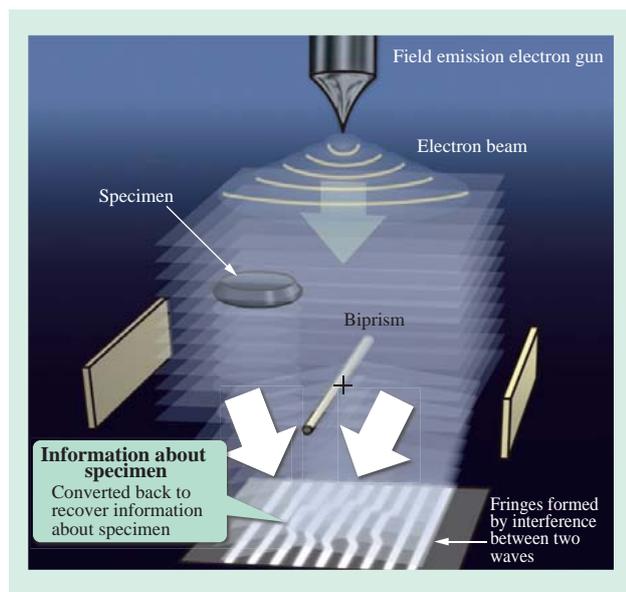


Fig. 1—Principle of Holography Electron Microscope. Information about the electromagnetic fields in the specimen is obtained from interference fringes formed by field emission electrons emitted with high brightness and coherence from the metal tip.

technology was first invented in 1948 by Dennis Gabor as a way of improving the resolution of electron microscopes⁽⁹⁾. The technology was not practically used at the time because the thermo electron guns used back then could only produce electron beams with low brightness and poor coherence. Subsequently, holograms (photographs containing three-dimensional information recorded using the holographic technique) like those of the present day that are able to be viewed under white light became possible following the invention and rise to prominence of the laser in 1960. During this time, research into holography for electron microscopes was making steady progress, with Dr. Akira Tonomura of Hitachi becoming involved in research in this field around 1968. Achieving electron holography in practice requires an electron beam with high brightness, a small source, and high coherence. What made this possible was the successful implementation by Hitachi in 1972 of the field emission electron gun referred to above. While this electron gun immediately entered widespread use thanks to the significant improvement it provided in SEM resolution, it was not adopted on transmission electron microscopes at the same time. Noting that field emission electrons exhibit a high degree of coherence, Dr. Tonomura developed the first practical holography electron microscope in 1978 by fitting a field emission electron gun to a transmission electron microscope^{(10), (11)}.

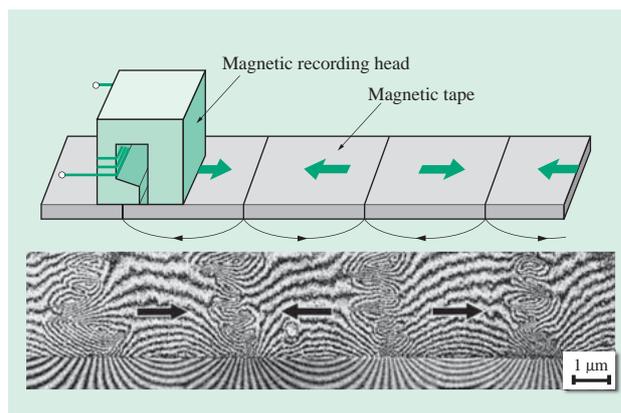


Fig. 2—Magnetic Fields of Recording on Magnetic Tape. It provides clear images of the magnetization inside the tape and the magnetic fields outside the tape.

Electron Holography

This section explains the principles behind electron holography and describes the key work done by Dr. Tonomura and his team.

Part of the electron beam produced by the field emission electron gun is directed through the specimen, with a positively charged conductive filament (diameter: 1 μm or less) called a biprism inserted in the center of the downstream path. The phase of the electron beam is changed by its passage through the specimen. This beam then meets the other half of the electron beam that has traveled through vacuum (bypassing the specimen) at a detector (fluorescent plate or image sensor) where the beams overlap to form interference fringes. In the absence of a specimen, these interference fringes form a uniform pattern. When a specimen is present, on the other hand, the fringes change in ways that carry information about the specimen due to the phase change described above. This specimen information (such as its thickness or information about electrical and magnetic fields) can then be recovered by shining laser light on to the interference fringes (the hologram). In recent years it has become possible to digitize the image and reconstruct it using a computer (see Fig. 1).

Electron holography and the various methods associated with it have enabled such achievements as observing the magnetic fields of recordings on magnetic tape⁽¹²⁾ (see Fig. 2), verifying the Aharonov–Bohm effect that experimentally confirms the presence of vector potentials⁽¹³⁾ (see Fig. 3), and observing a quantum of magnetic flux (minimum unit of magnetic flux, approximately 2.0×10^{-15} Wb)

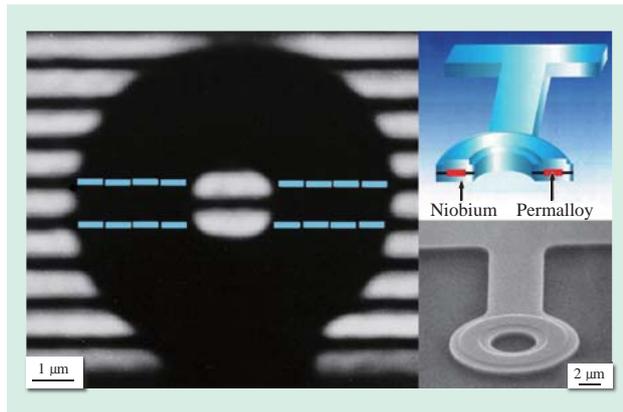


Fig. 3—Experimental Confirmation of Aharonov–Bohm Effect. Permalloy, a ferromagnetic material, was formed into a donut shape and surrounded with niobium, then the temperature was lowered so that the niobium became superconducting, preventing the magnetic field from leaking out of the permalloy into the surrounding space. However, the electron beam interference fringes (phase) are shifted both inside and outside the donut. This occurs because the vector potential (a form of magnetic potential) changes the phase of the electrons, thereby providing experimental confirmation of the Aharonov–Bohm Effect (whereby charged particles such as electrons are influenced by the magnetic potential in space where there is no electromagnetic field).

leaking from superconducting niobium^{(14), (15)}. In the case of the electron-beam version of the double-slit experiment⁽¹⁶⁾, the technique makes it possible to observe how the interference fringes produced by the electron beam after it passes through the two slits form progressively as the cumulative number of electrons increases, starting out as just individual dots (particles) when the number of electrons is still small (see Fig. 4). This experiment was included in “The Ten Most Beautiful Experiments in Science”⁽¹⁷⁾ for the way it gives an intuitive sense of the wave-particle duality

of electrons, and is referred to in numerous physics textbooks both in Japan and overseas.

Development of Ultra-high-voltage Holography Electron Microscope

In 2000, Hitachi and The University of Tokyo jointly developed the world’s first 1-MV ultra-high-voltage holography electron microscope, which was capable of measuring electromagnetic fields in microscopic regions with a point resolution of 120 pm⁽¹⁸⁾. The development was part of the High-resolution Imaging of Phase and Amplitude of Electron Waves project, a Strategic Basic Research Program of the Japan Science and Technology Corporation (since renamed the Japan Science and Technology Agency). The work made a major contribution to elucidating the mechanism of high-temperature oxide superconductors by providing a direct view of magnetic flux in the superconductors. This took advantage of the enhanced ability of ultra-high-voltage electron microscopes to pass a beam through specimens. It also enabled the dynamic observation of fluxon motion that occurs when the magnetic field is varied and which are influenced by crystal defects⁽¹⁹⁾.

DEVELOPMENT OF HOLOGRAPHY ELECTRON MICROSCOPE WITH ATOMIC RESOLUTION

After building the 1-MV holography electron microscope, Dr. Tonomura came up with an idea for the ultimate holography electron microscope. This was in response to the recent development of viable correctors (described below) for the spherical aberrations present in electron microscope lenses⁽²⁰⁾, a technology with the potential to dramatically

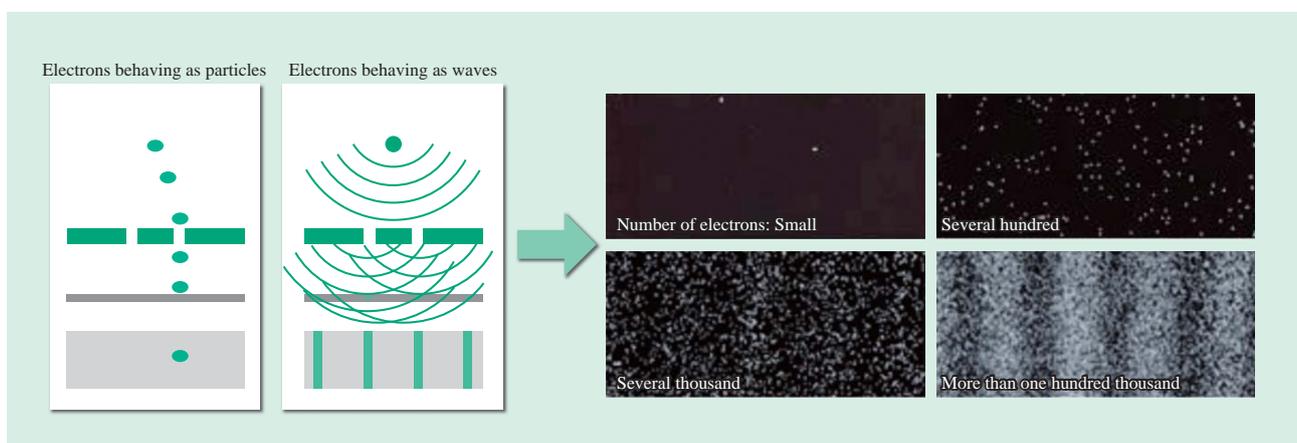


Fig. 4—Electron Double-slit Experiment.

While individual electrons are observed as particles, in large numbers they form the interference fringes characteristic of waves.

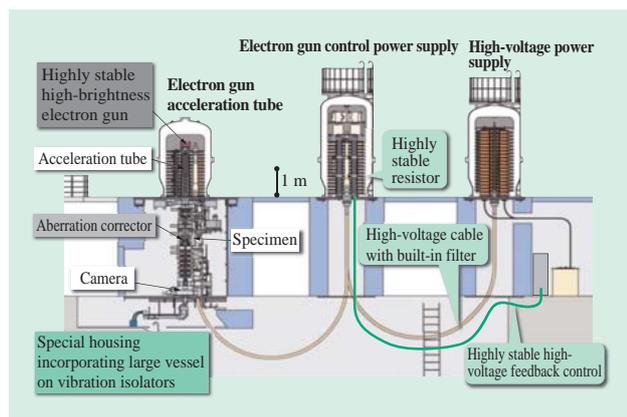


Fig. 5—Overview of Atomic-resolution Holography Electron Microscope.

The 1.2-MV ultra-high-voltage electron microscope has a three-tank design in which the high-voltage power supply is located separate from the electron microscope itself to ensure a high degree of stability. The tanks are connected by high-voltage cables.

improve the resolution of ultra-high-voltage electron microscopes. However, getting the best performance out of an aberration corrector required improving the stability of the electron microscope unit by a factor of more than two. No group in the world had ever previously grappled with such a difficult problem using electron microscopes that were so large in size and worked with such high voltages. Dr. Tonomura decided to take on this challenge of building a holography electron microscope with atomic-scale resolution that would be able to observe magnetic fields at scales in the order of tens of picometers.

At this point it is essential to mention Isao Matsui, the engineer who worked in tandem with Dr. Tonomura on the development of the ultra-high-voltage holography electron microscope. Mr. Matsui, who led the development of Hitachi's ultra-high-voltage electron microscopes and, more recently, the 1-MV holography electron microscope, was a major participant in the development of these instruments. Dr. Tonomura and Mr. Matsui started out in pursuit of this dream in 2003. Their performance targets were resolution in the 40-pm range (the best resolution possible at the time was in the 70-pm range), the ability to use electron holography to detect differences in electron phase equal to one one-thousandth of the wavelength, and the ability to measure electromagnetic fields in three dimensions.

Four Major Obstacles to Overcome

The atomic-resolution holography electron microscope consists of three high-voltage tanks, with the electron



Dr. Akira Tonomura (left) and Isao Matsui (right)

microscope itself mounted on a vessel on vibration isolators (Fig. 5). The 1.2-MV high-voltage generator tank is on the right, the electron gun power supply that generates the control signals for the electron gun is in the middle, and the tank on the left contains the electron gun and the acceleration tube that accelerates the electron beam up to 1.2 MeV and is made up of layers of electrodes and insulators. The electron gun and acceleration tube are maintained in a state of extreme high vacuum with a pressure in the 10^{-9} -Pa range or less. Because these components are at a high negative potential (-1.2 MV), they would discharge if exposed to air. For this reason, these components are all housed in a tank containing sulfur hexafluoride (SF_6) gas, a discharge suppressant, at a pressure of four atmospheres. Because the high-voltage power supply generates acoustic and electrical noise, it is located away from the electron microscope and connected via a high-voltage cable. To prevent external vibrations from reaching the electron microscope, it is mounted on a cylindrical vessel on vibration isolators that is supported by dampers on four corners. The basic design is largely the same as that of the 1-MV ultra-high-voltage holography electron microscope completed in 2000⁽¹⁸⁾. However, there remained four major obstacles to overcome in order to achieve the performance targets set for the atomic-resolution holography electron microscope. The components associated with each of these challenges are highlighted in Fig. 5. This is explained in the following sections.

World's First Use of Spherical Aberration Corrector in Ultra-high-voltage Electron Microscope

To provide an enlarged image of the structure of a specimen, optical microscopes use a combination of convex and concave lenses to eliminate focal blur by correcting for spherical aberrations in the lenses. Because the electron lenses used in electron

microscopes have for a long time not been able to function as concave lenses, spherical aberrations have been an obstacle in the way of improving resolution. While a practical system for correcting spherical aberration was developed in 1995⁽²⁰⁾, the electron microscope in which this spherical aberration corrector is used must be very stable for it to perform well. As a result, the system had not been successfully deployed in large ultra-high-voltage electron microscopes. In response, Hitachi developed the techniques described in the following three sections to make significant improvements in instrument stability, becoming the first in the world to successfully incorporate a spherical aberration corrector into an ultra-high-voltage electron microscope.

1.2-MeV Electron Beam with Reduced Energy Dispersion

Focal blur will occur regardless of the presence of a spherical aberration corrector if the dispersion in the energy of the electron beam is high. This makes it essential that the dispersion used to accelerate the electron beam be extremely stable. A calculation estimated that achieving the target resolution with the high-energy (1.2 MeV) electron beam would require that its dispersion be reduced to 0.54 eV. To improve the stability of the voltage used to accelerate the electron beam, Hitachi developed a number of components, including resistors with low noise and an electrical resistance that does not change significantly with temperature, a high-voltage power supply cable with a built-in noise filter, and a highly stable high-voltage feedback control circuit. These were used to develop a 1.2-MV ultra-high-voltage power supply system with stability of 3×10^{-7} , exceeding that of the previous microscope by about 70%.

Electron Gun Able to Produce Stable Electron Beam for Long Periods of Time

On conventional field emission electron guns like the electron gun in the 1-MV ultra-high-voltage electron microscope, the current of emitted electrons starts to fall immediately after electron emission is initiated by applying an extracting voltage to detach electrons. As a result, the extracting voltage needs to be recalibrated once or twice a day during use. Furthermore, because the trajectory of the electron beam shifts by a small amount each time the extracting voltage is recalibrated, it is not easy to keep the spherical aberration corrector operating with a perfectly aligned beam. Accordingly, to maintain optimal performance without having to recalibrate the spherical aberration corrector settings midway through a day's operation,

Hitachi recognized the need for a field emission electron gun that could emit a stable electron beam for 10 or more hours at a time without calibration. While the Schottky-type electron gun⁽²¹⁾, which uses an electric field to emit electrons from a heated emitter, is very stable, it is not suitable for holography electron microscopes because its energy dispersion is higher than for field emission electrons. This prompted Hitachi to set out to achieve high stability with an electron gun that emits electrons using only an electric field, without having to be heated. This requires an extremely high level of vacuum (3×10^{-10} Pa) at the location in the electron gun where electrons are emitted, approximately 100 times what was possible with previous instruments. Accordingly, they set about developing an electron gun that was able to work in this extreme high vacuum.

Development of Equipment Technology to Eliminate Factors that Degrade Resolution

Making atomic-scale observations requires that external interference with the electron beam or specimen, such as vibration, sound, or magnetic fields, is kept to an absolute minimum. To achieve this, Hitachi built a special reinforced building designed specifically to house the electron microscope, including walls lined with acoustic absorbent material and measures to minimize changes in ambient temperature. The targets were a high degree of sound insulation and low acoustic noise (20 dB or less above 200 Hz), reduced floor vibration (7.2×10^{-4} cm/s² at 100 Hz), and temperature variability in the vicinity of the microscope (ambient temperature) of $\pm 2^\circ\text{C}/8$ h. To minimize magnetic fields, the area around the electron microscope was enclosed in permalloy to provide magnetic shielding.

Proposal and Adoption by FIRST Program

To achieve Dr. Tonomura's dream of a holography electron microscope with atomic resolution, it was first necessary to make solid progress on developing the technologies for overcoming the four main obstacles. In fact, the technologies were developed through participation in the Development of Elementary Techniques for Electron Microscope in Next Generation project, an experimental science and technology research project sponsored by the Ministry of Education, Culture, Sports, Science and Technology that ran from 2006 to 2008. Specifically, this included an ultra-precise reference voltage source, a high-voltage cable with built-in noise filter, a highly stable low-noise resistor module, and a high-resolution

crystal lattice image evaluation technique. In the case of the high-resolution crystal lattice image evaluation technique⁽²²⁾ in particular, Dr. Tonomura's team succeeded in setting a new record of 25.9 pm for the smallest crystal lattice spacing ever observed (beating the old record of 49.8 pm) by minimizing external noise (such as vibration or electromagnetic fields), which degrades electron microscope resolution, and by using the dark field method whereby imaging only includes the high-order waves scattered by the crystal lattice. Most of the technologies developed through this work are now featured in the latest electron microscopes. However this also included one factor that caused major difficulties. This is described in the "Development Story" section below.

Believing that development of the electron microscope would require a budget of around 10 billion yen, Dr. Tonomura asked everyone involved to start working toward the formulation of a proposal for a major national project. With the aim of strengthening science and technology in Japan, the government set up a groundbreaking scheme for national projects in 2009 whereby 30 leading researchers would each be given budgets of several billion yen that they would be free to use as they wished over a five year period. This was called the Funding Program for World-Leading Innovative R&D on Science and Technology (FIRST Program) and began with a process for selecting the researchers. Dr. Tonomura set out to win selection under this scheme and was rewarded for his efforts with the prospect of being granted a budget of nine billion yen. Unfortunately, this coincided with a change of government, raising concerns that the project budget might be cut to about one-third of the anticipated sum. There was no chance of Dr. Tonomura building the instrument he envisaged for three billion yen. In the end, with cooperation from various different people and by cutting back his plans to include only the microscope development, and by excluding the application research planned for the completed instrument, the work was able to get underway with a budget of five billion yen.

At the same time, work continued on setting up the development program. A major problem was the question of where to base the development. All past electron microscopes had been first assembled in the factory for basic performance testing and then broken down to be reassembled at the customer site or at a Hitachi laboratory. However, given the limited development schedule and the fact that the microscope was not intended as a product to be sold to customers,

consideration was given to the option of having all parts for the microscope purchased and assembled at Advanced Research Laboratory, Hitachi, Ltd. (as it was then known) in Hatoyama in Saitama Prefecture. An electron microscope consists of tens of thousands of parts, and the laboratory had no experience with assembling such a large piece of apparatus from its individual parts. To achieve this, dedicated staff were assigned to tasks such as purchasing and accounting and based at Hatoyama. Former staff involved in engineering work at the plant that Hitachi High-Technologies Corporation had at Naka at that time were also requested to relocate to Hatoyama. Because it was clearly impossible to do all the machining work at the laboratory, Mr. Matsui and experienced engineers from the prototyping department at Hatoyama consulted with precision engineering companies around Japan to request their assistance. Numerous engineers from companies such as Hitachi High-Technologies Corporation and Hitachi High-Tech Fielding Corporation, including people with experience from the development of the ultra-high-voltage electron microscope, were requested to relocate to Hatoyama via transfers or secondments to work on design, development, and assembly. Naturally, the project also included developers from Hitachi's Central Research Laboratory, Hitachi Research Laboratory, and Production Engineering Research Laboratory (as they were then known), with the team amounting to more than 30 people at its peak.

Project Beset by Series of Unanticipated Events

The project formally commenced in March 2010. They started by setting to work on the overall concept and on the design and fabrication of bulky components that would take a long time to manufacture. Simultaneously, preparations also commenced for the construction of the building that would house the electron microscope. Once these initial birth pangs had been overcome, the project appeared set to proceed smoothly. Unfortunately, they were beset by an unanticipated event. Isao Matsui, who was one of the originators behind the project and the engineer who had made Japan the home of the ultra-high-voltage electron microscope was found to have a scirrhus carcinoma of the stomach. He died in December 2010, less than two months after the diagnosis. This was a major setback for the development. The surviving engineers were overwhelmed by this development, and the most

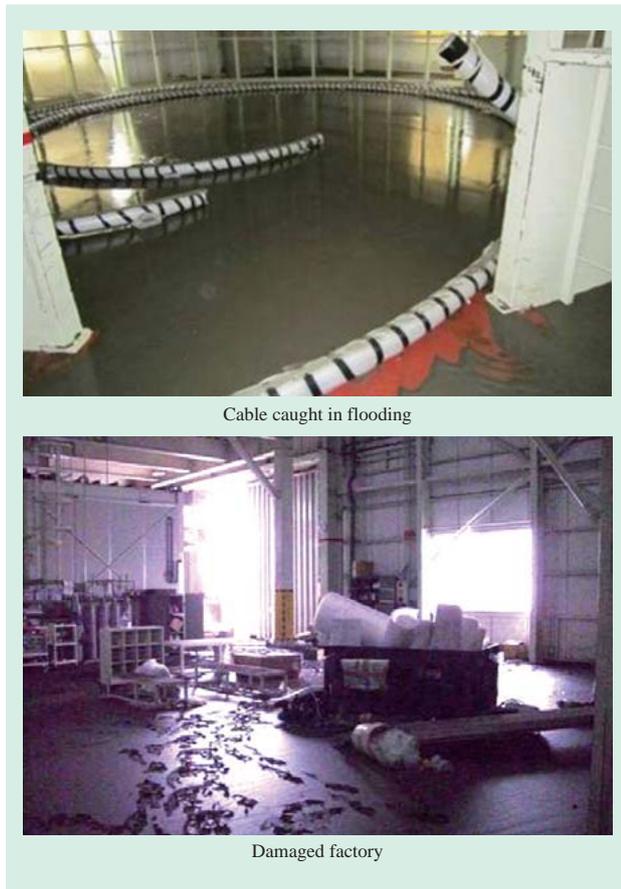


Fig. 6—High-voltage Cable Struck by Tsunami Just Short of Completion and State of Factory after Tsunami. The high-voltage cable being manufactured at a factory located on the shoreline near Hitachi City port was rendered useless by the large tsunami. The ground floor of the factory building was inundated and was not able to return to operation immediately after the water receded.

shocked of all must have been Dr. Tonomura who had worked in tandem with Mr. Matsui to promote the project. Nevertheless, by spurring on the project team without betraying any outward signs of his distress, Dr. Tonomura and the remaining team members pressed on with the development despite the somber mood. The construction of a purpose-built building to house the electron microscope proceeded smoothly, and work started on the design and manufacturing arrangements for other bulky components, including the high-voltage cable, the “vessel on vibration isolators” on which the electron microscope was to be mounted to isolate it from external vibrations, and the high-voltage tank that would house the high-voltage components, under the auspices of the designers who had taken over from Mr. Matsui. On the other hand, the design and manufacturing arrangements for the many components of the electron microscope itself were progressing only slowly.

In March 2011, an engineer working on building the mechanical components raised the alarm, reporting that because most of the manufacturing drawings had yet to be supplied, if nothing was done, it would not be possible to arrange for the work to be done by the engineering subcontractors and still get it completed on time. Then, on March 11, just as a meeting was getting underway to overcome this problem, the team experienced a shaking unlike anything they had been through before. It was the Great East Japan Earthquake. The microscope building was in the middle of pouring concrete for the foundations when the earthquake struck. After confirming that nobody was injured and that there was no damage to existing buildings, the team rushed off to the construction site only to find that work had already resumed with no signs of anything untoward. Impressed by this dedicated professionalism, the team was instilled with a commitment to complete the electron microscope this building was intended to house, no matter what it took. While there was no notable damage in the Hatoyama region, component-producing factories in Miyagi and Ibaraki were in a bad state. The high-voltage cable that was close to completion thanks to use of technologies developed in the 2006–2008 project was inundated by the tsunami and no longer in a usable condition (see Fig. 6). Rebuilding the special 12.5-cm-diameter high-voltage cable, which topped the list of expensive components with long lead times, was tantamount to restarting the budget and schedule from the beginning. Miraculously, the backup cable had escaped the flooding, and in the end it was completed only about three months late thanks to the efforts of Hitachi Cable, Ltd. (as it was then known). Another narrow escape was had by the power supply manufacturer that was building components such as the ultra-stable high-voltage power supply and lens power supply, both seen as critical components of the microscope. The headquarters factory of this company was located close to Sendai Airport. However, although the scenes of the airport being inundated by the tsunami were repeated time and again on television over the following days, the images stopped just short of showing the factory site, and because communications were down, there was no way of confirming whether the company was all right. A few days later the news came through that the tsunami had fortuitously stopped just short of the factory and the completed power supplies were unharmed. Despite the shortage of gasoline and other daily necessities, they still managed to deliver the power supplies to



Fig. 7—Purpose-built Electron Microscope Building Providing Stable, Low-noise Environment.

The building provides sound insulation and low acoustic noise (20 dB or less above 200 Hz), reduced floor vibration ($7.2 \times 10^{-4} \text{ cm/s}^2$ at 100 Hz), and temperature variability in the vicinity of the microscope (ambient temperature) of $\pm 0.2^\circ\text{C}/8 \text{ h}$.

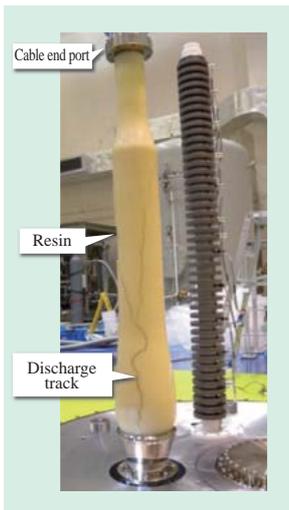


Fig. 8—Electrical Discharge Occurred at Tip of High-voltage Cable.

The end of the cable is at a high negative voltage while its base is grounded. When testing was performed by applying a high voltage to one end of the cable, a discharge occurred before reaching the rated voltage of 1.2 MV, causing a discharge track on the surface of the resin.

Hatoyama all but on time. The team members were so grateful they almost cried. When returning to Miyagi after the delivery, the team provided them with some supplies to take back for the people caught in the disaster. Although Hatoyama endured planned power outages that disrupted proper work for a time after the earthquake, the project team still came in to work nearly every day and continued getting on with what development they could. Nobody believed that anything worse could be yet to come.

It was a great shock then when it was revealed on March 16 that Dr. Tonomura had been diagnosed with pancreatic cancer. Bravely, Dr. Tonomura immediately passed on the news to the project team and pledged to return after his treatment. Trusting in these words, the

team continued with the development. The delayed design of the electron microscope unit was able to proceed thanks to the efforts of everyone involved, rigorous progress management, and the reinforcements that made up for the lack of staff.

The following section tells the story of the development in chronological order, up to its completion.

Development Story

—Overcoming Numerous Obstacles— High-voltage Cable

The microscope building was completed on schedule in late August 2011 (see Fig. 7). The building succeeded in providing the required low-noise environment, with its floor vibration, acoustic noise, and temperature variation performance all coming in within specification. The temperature variation of $\pm 0.2^\circ\text{C}/8 \text{ h}$ exceeded the target by an order of magnitude⁽²³⁾. From then on, the major components were delivered one after another, to the point where the apparatus looked largely complete to an outside observer by the 2012 New Year. After the high-voltage cable that had been rebuilt following the tsunami damage was delivered and installed about three months later, live high-voltage testing at 1.2 MV was conducted on January 31 and February 7, 2012. Unfortunately, a major problem was encountered in the first stage of performance testing that made it impossible to apply the full voltage due to electrical discharges occurring at 900 kV and 600 kV (see Fig. 8). Because the cable had already passed 1.3-MV testing prior to delivery from the factory, these two discharges occurring at such low voltages were unexpected and very distressing. While there was a strong suspicion that the discharges were the result of foreign material, the apparatus had been assembled with great care and attempting any more rigorous measures for excluding foreign material would have been impractical. Even if some foreign material did still remain due to the current work environment and assembly practices, the voltage still needed to be applied. A joint team of high-voltage experts from within Hitachi and the cable manufacturer (Hitachi Cable) was quickly formed to work on the problem, and they embarked on a program of simulation and testing. An electric field simulation that was conducted to assess the influence of foreign material found that, unless metal fragments too large to miss (around 10 mm) were present, then the rated voltage should still have been possible. Another suspected cause was salt contamination in the

insulators resulting from their being manufactured in a plant that had been restored after the tsunami damage. A sample component deliberately sprayed with saltwater was made and its insulation performance was tested only to find that no change whatsoever could be detected in the characteristics for the sort of salt levels found by the sea. As a result of ongoing investigations, and with advice from senior laboratory personnel, a dynamic simulation that considered changes over time found that the electric field at certain locations where it was expected to be close to zero was at times several tens of times higher. On a timeframe of several minutes to several hours after the voltage was turned on, the electric field at certain locations would increase temporarily. This phenomenon had been overlooked by the static simulations used for the design. Nevertheless the calculated electric field strength still did not reach the level that would trigger a discharge. Furthermore, this was in agreement with the fact that numerous live voltage tests had been conducted at the cable factory without discharges occurring. This led them to think that, while the increase in electric field strength was not on its own causing the discharges, they were likely being caused by a combination of other factors. It was concluded that small pieces of foreign material were one such factor, and the presence of residual air in component connectors was likely another. It is impossible to eliminate small pieces of foreign material and residual air entirely. However, because preventing the increase in the electric field at the problematic locations would be enough to prevent the discharges, it was decided to install electrodes for this purpose. By the time the modifications were completed and the voltage testing passed, it was already past July 2012. There were no further instances of discharges caused by the cable.

Death of Dr. Tonomura

After undergoing surgery in April 2011, Dr. Tonomura went into remission and was well enough to return to the laboratory six months later. A symposium to be attended by notable physicists and microscopy researchers from around the world was planned for May 2012 and everyone hoped that he would be able to attend as the event's host. Instead, he appeared before the project team for the final time in April 2012, passing away on May 2, 2012, just before the symposium was to start. This was at a time when the development team was focused on ways of dealing with the high-voltage cable discharge problem, and the news came through while it was at Hitachi Cable attending a meeting on the subject. Although rocked

by this bad news, it was not unexpected because everyone involved in the development had, at Dr. Tonomura's insistence, been made aware of his condition, and so the work continued. The symposium, too, went ahead as planned, and included the playing of a video message recorded by Dr. Tonomura before his death. This loss of its principal investigator left the project with the problem of deciding who should take his place, putting the possibility of continuing the project in doubt. Accordingly, Nobuyuki Osakabe, who had racked up numerous notable results during his time working as a researcher in Dr. Tonomura's group and was at that time general manager of Central Research Laboratory at Hitachi, Ltd., appeared before the FIRST committee in the role of acting lead researcher to explain the situation and convinced them that the microscope would still be completed on schedule. As a result, the project was able to continue.

High-voltage Resistor Module

No sooner had the problem with the high-voltage cable been dealt with than a new problem emerged. This was the presence of noise indicating that small discharges were taking place, the problem being discovered by voltage stability measurements made after it became possible to operate with a voltage of 1.2 MV in late July 2012. To identify the cause of the noise, the large tank was opened and closed many times to enable a series of tests. An inspection of the components inside the tank found anomalous bulges on some of the nearly 100 resistor modules that were used to maintain stability at high voltage and to measure the voltage and provide feedback to the high-voltage control system. As noted in the section, "Four Major Obstacles to Overcome," development of these resistor modules to improve stability at high voltage had been completed prior to the commencement of the project. The module specifications stipulated high performance, including a temperature coefficient of resistance of ± 2 ppm/ $^{\circ}$ C and current noise of less than 0.3 ppm/min. The culprit was the silicone rubber molding material that had been selected on the assumption it would be suitable for these modules. Silicone rubber is porous, meaning it is easy for gas to pass through it. Because all of these components are held in SF₆ gas at high pressure (4 atmospheres), the gas permeates the silicone rubber, forming voids and cracks. The noise resulted because these voids cause small discharges (partial discharges) to occur. Although an attempt was made to fabricate alternative parts that could prevent void formation while still continuing to use the silicone rubber, this produced no signs of

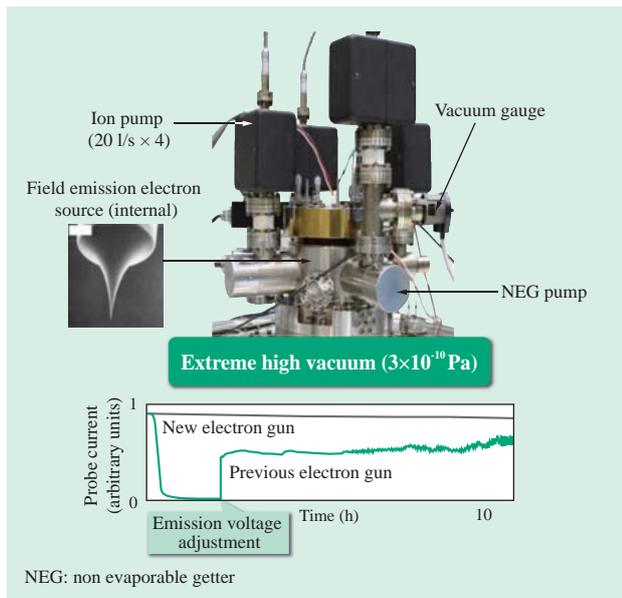


Fig. 9—Highly Stable High-brightness Electron Gun. With previous electron guns, because the electron current drops off quickly after electron emission is initiated, it is necessary to adjust parameters such as the emission voltage and wait for stability to be achieved before it can be used. In contrast, by maintaining extreme high vacuum, the new electron gun achieves stability and is ready for use immediately after initiating emission of the electron beam.

improvement. When an investigation was conducted in parallel with this to track down information about resin molds, which included web searches rather than restricting itself only to in-house sources, it returned a search result for a doctoral thesis from The University of Tokyo. In a case of the answer being close at hand all along, the author is a researcher at Mechanical Engineering Research Laboratory, Hitachi, Ltd. (as it was then known). Based on the researcher's advice, the team gave up on silicone rubber and went back to using tried and tested epoxy resin. However, resistors embedded in epoxy resin are subject to stresses as it hardens, with the risk that this will degrade their temperature coefficient of resistance. Nevertheless, the target performance was achieved by making changes to the coating material and shape of the resistors to minimize these stresses. It took until August 2013 before the team was confident that this would resolve the problem and eliminate the noise that was causing difficulties for high-voltage stability testing. However, problems continued, with a major discharge occurring when operating at 1.2 MV after the new resistors were installed. The source of the discharge was the newly manufactured resistor modules, caused by the presence of abnormal protrusions or metal fragments due to

soldering defects in the modules. There was also a problem with the testing method used to determine how high a voltage each module could withstand. A full set of modules believed to be compliant finally became available only after improvements were made to the manufacturing process in discussion with the supplier, and changes to the orientation of the electric field were applied during testing.

Highly Stable High-brightness Electron Gun

Fortunately, the development of the highly stable high-brightness electron gun described in the section, "Four Major Obstacles to Overcome," proceeded without any dramatic incidents, achieving the target of an extreme high vacuum of 3×10^{-10} Pa in January 2013, roughly on schedule. Testing confirmed that a stable electron beam could be generated for 15 or more hours at a time without needing recalibration^{(24), (25)} (see Fig. 9).

Contamination of Acceleration Tube

In the spring of 2013, at the same time as the team was going back to using epoxy as the mold material for rebuilding the resistor modules, progress was being made on getting the acceleration tube working. The acceleration tube is a 1.8-m-high cylindrical component made up of 44 layers of electrodes and insulators. There is a high potential difference across each layer (approximately 30 kV), with the topmost electrode having a potential of -1.2 MV and the bottommost electrode a potential of 0 V. The electron beam is accelerated as it travels from the top to the bottom of the tube, the interior of which is maintained at ultra high vacuum in the 10^{-9} -Pa range. Although it has been estimated that the acceleration tube could withstand potential differences across each layer as high as around 80 kV under ideal conditions, at the time they were delivered from the supplier, stability could only be maintained up to 20 kV due to factors such as residual gases, protrusions, or fragments of foreign material in the insulators and in the joints between the electrodes and insulators. Accordingly, the acceleration tube (with its interior kept in a state of ultra high vacuum) was first placed inside a large chamber equipped with a heater and baked to drive out any gases, then voltages were applied across each layer to deliberately induce a small discharge and thereby expel any foreign material and smooth out any protrusions. This improved the performance of the acceleration tube to the level needed in practice, which was to be able to operate stably for long periods of time with voltages of 30 kV or more across each layer. An inspection of the acceleration tube after the final

baking in May 2013 found discoloration (browning) on the exterior of the insulators. While the reduced ability to withstand high voltages was not fatal, some layers had degraded somewhat or were subject to an increasing number of micro discharges, causing noise. An analysis of the discolored material found it to contain metallic copper and copper chloride. Eliminating this copper entirely would have required shaving off a layer of skin from the surface of the acceleration tube insulators, washing them again, and repeating the baking process, a job that would have taken months to complete for such a large component. With only 10 months of the original development schedule remaining, this would have removed any hope of achieving the original objective of world-leading resolution by March 2014. Although there was a chance that performing repeated baking cycles without first removing the contamination would be enough to achieve the desired voltage, when the implications of failure were considered, it was decided instead to go back to fundamentals and look at causes and countermeasures. The problem-solving work, too, was aided by knowledge from Hitachi laboratories and the capabilities of the analysis team. The jobs of analyzing and identifying materials, identifying which material was causing the problem, and determining the mechanism of copper adhesion and how to stop it were completed in less than two months. The reprocessing and cleaning of the acceleration tube were completed around the same time, so thanks to the reapplication of a voltage to the tube and the success of baking at the increased voltage it could withstand, it had become possible to operate stably at a voltage of 1.2 MV by the end of December 2014. Two years had elapsed since the live testing of the high-voltage cable. The FIRST project ran up to March 2014, and it was looking like achieving the target resolution by that date would be impossible no matter what they tried. However, by making the case that, although the date for achieving the target resolution had been put back to the 2014 fiscal year, it would definitely be achieved, they succeeded in convincing those involved. They set out to produce 1.2-MV electron microscope images before the end of the fiscal year, and achieved this goal on March 12.

Taking on Challenge of Achieving World-leading Resolution

Operation of Aberration Corrector

Once the basic set of functions for the electron microscope had been demonstrated, the next challenge

was to achieve world-leading resolution. If all worked to its design specifications, the resolution should be in the 40-pm range. Although factors such as the stability at high voltage, installation conditions, electron beam stability, and the operation of the aberration corrector had been checked individually, it was not until system testing was performed that they would find out how well these would function when working all together. The aberration corrector is one of the key components of the microscope but its performance cannot be determined until the electron microscope itself is operating at close to its target performance. To achieve this, the aberration corrector was removed temporarily and the performance determined for the electron microscope on its own. It took until July 2014 before the aberration corrector was put back and its performance tested.

Achievement of World-leading Resolution

Although resolution of 60 pm had already been achieved by August, subsequent progress was slow, improving to no better than 50 pm by October. Around this time it was announced at the International Microscopy Congress held in Prague in September that a team from The University of Tokyo and another company had reduced the world record for electron microscope resolution from 47 pm to 45 pm⁽²⁶⁾. This increased the pressure on the team to achieve world-leading resolution. Another piece of news was that a FIRST ex-post assessment hearing was scheduled for November 26. It was vital that world-leading resolution be achieved before this date. The inability to improve resolution beyond 50 pm was believed to be due to a combination of factors: the static performance of the stage, external electrical noise, and damage to the specimen caused by the electron beam. In other words, it was believed that while the microscope had the necessary potential for performance, this had not yet been realized. Accordingly, the team adopted a strategy of imaging a large number of locations in as short an exposure time as possible. The specimens being used consisted of gallium nitride (GaN) crystals that had been formed into a wedge shape by partitioning in order to image the atoms at its peak (a thickness of several nanometers). However, exposing the specimen to the electron beam for too long caused its structure to be disrupted by the beam's energy. Accordingly, they adopted a practice of first focusing the electron beam at a separate location and then quickly redirecting it to the location to be observed in order to image a large number of sites, each several atoms thick, in the style of random sampling. This

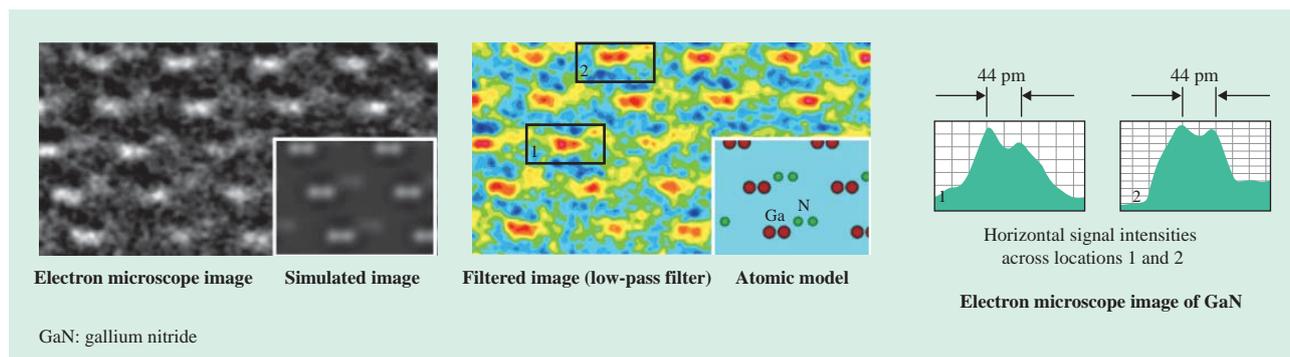


Fig. 10—Imaging of Ga Atom Spacing in GaN.

To determine the actual resolution of the electron microscope, it was used on monocrystalline GaN to capture an image of the 44-pm spacing between Ga atoms.



Fig. 11—Atomic-resolution Holography Electron Microscope. The photographs show the three large high-voltage tanks (top-left), the electron microscope operation console (bottom-left), the electron microscope itself (lens, specimen stage, detector, etc.) (center), and the electron microscope viewed from the front (right).

succeeded in imaging the 44-pm spacing between Ga atoms (see Fig. 10). By this time it was already November 14, but it was in time to report a world-leading resolution of 44 pm at the ex-post assessment hearing. Next, the team started preparing to submit a paper to a scientific journal in order to announce their results publically as soon as possible. However, when considering the resolution of a transmission electron microscope in an academic context, rather than just being able to see the gaps between atoms, what is required are images that clearly indicate the lattice spacing of the crystal. A subsequent experiment performed using monocrystalline tungsten improved on the 44-pm record by imaging a lattice spacing of 43 pm with good repeatability. A paper including this data was submitted to a journal, *Applied Physics Letters*, in early January 2015, and received an unusually prompt response confirming its acceptance by the end of the month⁽²⁷⁾. With the news also appearing in newspapers on February 18, the date of publication of the paper,

the instrument was finally recognized as the electron microscope with the world's highest resolution. Fig. 11 shows the main components for the completed atomic-resolution holography electron microscope.

NEW REALMS OPENED UP BY ATOMIC-RESOLUTION HOLOGRAPHY ELECTRON MICROSCOPE

While this article has focused on resolution, which is the simplest expression of microscope performance, in fact the real value of the instrument comes not merely from its world-leading resolution, but also from its ability to make quantitative measurements of electromagnetic fields at atomic resolution. Materials are made up of atoms, and their properties are determined by the types of these atoms and how they are arranged, and by the fields they produce. The key to the successful development of advanced functional materials that provide new functions or outstanding performance lies in determining how atoms are arranged and the associated fields. Because it is particularly common for the electromagnetic fields inside materials be a major determinant of their characteristics, it is important to measure electromagnetic fields at the atomic level. Examples include the electrode materials used in rechargeable batteries, magnets and superconductors, and thermoelectric conversion materials (see Fig. 12). Developments such as improvements to the performance of these materials or the commercialization of new materials will transform society by dramatically improving the performance of the many systems that use them.

One example is the high-performance magnets that play an important role in making hybrid-electric vehicles and electric vehicles viable. There is a need to find new magnets that can outperform the

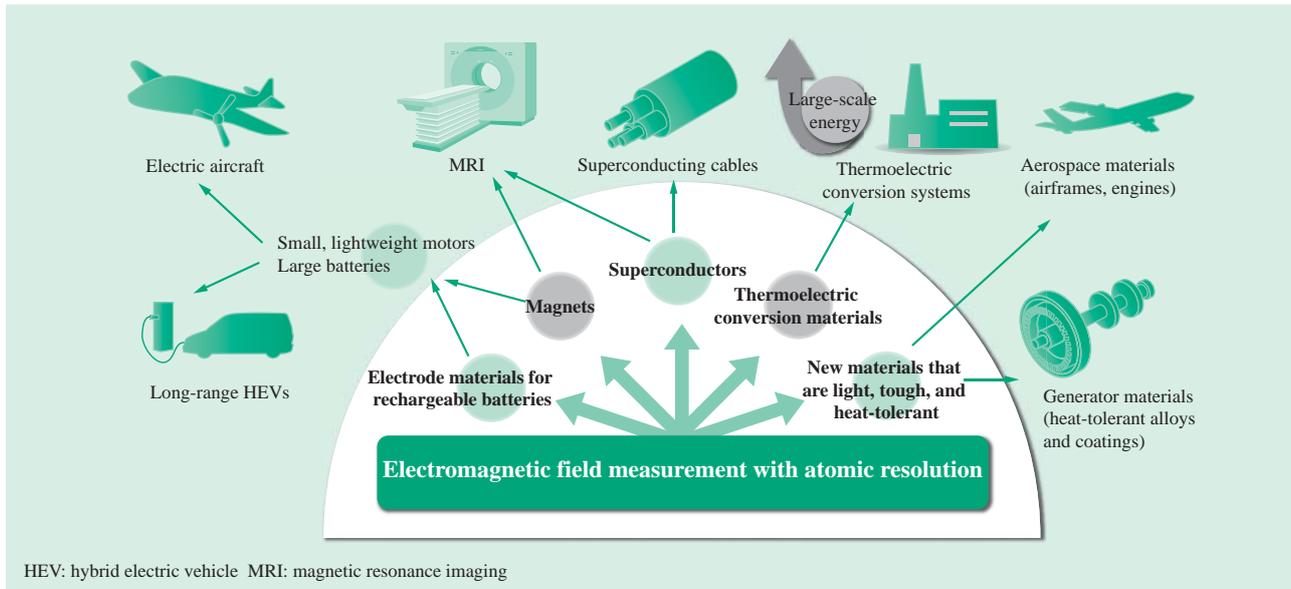


Fig. 12—Possibilities Opened up by Electromagnetic Field Measurement with Atomic Resolution.

The performance of new functional materials that underpin energy-saving and resource-saving innovations, such as magnets, superconductors, and thermoelectric conversion materials, is closely related to the behavior of electromagnetic fields in microscopic regions inside these materials. Through the elucidation of these mechanisms, electromagnetic field measurement with atomic resolution will lead to the development of groundbreaking new materials.

neodymium magnets used currently. Along with improving performance at room temperature, there is a need for materials that can be used in harsh conditions with high temperatures or strong magnetic fields. To develop magnet materials like these, it is important to determine the arrangement of the atoms from which they are made and the resulting magnetic properties so as to gain insights into which materials to use, their composition, and manufacturing techniques. Viewing magnetic fields at the atomic level has become essential to achieving this. As described above, the atomic-resolution holography electron microscope will drive the development of new materials, not only for resolving problems such as energy and the environment, but also for making life more comfortable and convenient. It is also anticipated that the development of these advanced and groundbreaking materials will facilitate progress in fundamental science. Hitachi intends to undertake research into these applications through open innovation in conjunction with leading research institutions in Japan and elsewhere. For example, Hitachi intends to work on groundbreaking material developments and the elucidation of their physics by working with research institutions in Japan and elsewhere, such as RIKEN, with which Hitachi is already working on research into techniques for using the holography electron microscope.

CONCLUSIONS

The atomic-resolution holography electron microscope is now complete. Tragically, both Dr. Tonomura, who came up with the concept for the microscope, and Isao Matsui, who, as an engineer, turned the concept into reality, were felled by illness before the microscope was finished. As described in this article, the project also faced numerous technical difficulties that brought its very completion into doubt any number of times. The depth of Hitachi's research and development and "monozukuri" manufacturing know-how; the extent of its practical capabilities; and its tradition of unstinting collaboration were all demonstrated by the fact that these difficulties could be overcome to complete the microscope. It is also thanks to the support of academics at numerous universities and other research institutions, the staff of supporting institutions, and the willingness of the staff of companies from outside the Hitachi Group to work alongside Hitachi to confront difficult manufacturing challenges. The author would like to express his heartfelt gratitude for this assistance.

This research was supported by the Japan Society for the Promotion of Science through the FIRST Program designed by the Council for Science, Technology and Innovation.

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ABOUT THE AUTHOR



Hiroyuki Shinada, Ph.D.

Center for Exploratory Research, Research & Development Group, Hitachi, Ltd. He is currently engaged in the development of electron microscopes and the study of their applications. Dr. Shinada is a member of the Japanese Society of Microscopy (JSM), the Japan Society of Applied Physics (JSAP), and the Society of Instrument and Control Engineers (SICE).