

Detailed Description of Scientific Achievements of Dr. Akira Tonomura

1. Overview

Dr. A. Tonomura developed new methods of electron microscopy, electron holography [1] and “coherent beam” Lorentz microscopy [2], which make use of the wave nature of electrons. These methods may generically be described as Electron Phase Microscopy. He used his methods to clarify the fundamental questions of quantum mechanics, demonstrating the Aharonov-Bohm effect decisively [3] and bringing to light how the wave-particle duality of electrons is realized in Nature [4]. Recently he has been applying these methods extensively to make visible the microscopic structure of matter that has been so far inaccessible, including the space-time behavior of the quanta (so-called vortices) of magnetic flux in both conventional and high-temperature superconductors [2].

1.1 Making use of Phases

The methods developed by Tonomura make use of coherent electron waves from a pointed field-emission (cold emission) sources and their quantum mechanical phase shifts. The holography principle compares the phases of two waves along the two paths, one path passing inside the material and the other outside. Coherent Lorentz microscopy compares the phases of two waves along the paths embracing a vortex in the material.

The basic idea of the electron holography was put forward by D. Gabor [5] in 1949. Also, the first practical use of the pointed field-emission electron source was made by A.V. Crewe [6] in 1968 for his scanning electron microscope. Moreover, (incoherent beam) Lorentz microscopy for the observation of magnetic domain structure was known.

However, without Tonomura’s efforts over four decades, electron phase microscopy would not have been born, grown, and developed to its current state, and we would not be able to see the microscopic magnetic structure inside materials quantitatively and directly. In order for suggestions and ideas to materialize, numerous difficult obstacles must be overcome in the laboratories.

To realize the electron holography, coherence of the electron waves is a must and hence the radius of the tip of the pointed electron source must be less than $0.1\mu\text{m}$. He needed a bright beam of electrons to uniformly and coherently illuminate the whole

specimen region to be observed [7]. These are just a few examples of a large number of problems that Tonomura had to solve. Indeed, it took more than 10 years of focused effort for Tonomura to finally realize his first practical electron holography microscope in 1979 [8]. Since then, Tonomura has been working steadily and relentlessly to improve his methods and apparatuses to make them useful in materials research.

1.2 Particle-wave Duality

Using his coherent electron beam, with a very weak intensity for this experiment, Tonomura demonstrated in 1987 how, in the two-slit interference experiment (using actually electron biprism in place of the two slits), the interference pattern emerges from electrons detected one by one on the image plane equipped with a position-sensitive detector [4]. In the beginning when few electrons had arrived, their landing locations looked random. However, when the number of electrons landing on the image plane increases, a remarkable effect is observed step-by-step: there slowly appears an interference pattern, as if the electrons knew from the beginning the places where they should land more frequently and the places to avoid.

1.3 Development of Electron Holography

Tonomura opened a new era of the electron microscopy by demonstrating in 1968 that the electron holography is indeed possible [9]. In 1979, he realized a field emission electron gun of extremely high coherence, a decisive step which significantly improved the image resolution of reconstructed image, fulfilling the dream of D. Gabor, who gave the principle in 1949 for the purpose of overcoming the resolution limit inherent to electron microscopy. Moreover, the electron phases were found to be very sensitive to magnetic fields, enabling Tonomura to use his coherent beam to look into the microscopic magnetic structure of materials [11][12][13].

1.4 Definitive Confirmation of the Aharonov-Bohm Effect

Tonomura's coherent beam solved a question fundamental to quantum mechanics. Aharonov and Bohm [17] pointed out in 1959 that an electron wave function should be affected by a magnetic flux even if the flux was confined in a region inaccessible to the electron. This effect stood against the traditional view that electron is only affected by the field at the point where the electron is located, and thus their paper generated long

lasting disputes [18]. There were experiments supporting the prediction of Aharonov and Bohm, but skeptics were concerned about leakage fields.

Tonomura's experiment in 1986 [3, 19] settled the issue by making use of a ring magnet covered with a superconducting film (to guarantee no leakage field by its Meissner effect) and with a copper film (to keep electron waves from penetrating into magnet). The idea was ingenious and the fabrication techniques unrivaled. The ring must be tiny, with an inner radius of $2\mu\text{m}$ and an outer one of $4\mu\text{m}$.

Tonomura and collaborators established that there was indeed a phase difference between the two waves, one passing inside the hole of the ring and the other outside the ring, under the condition of no flux leakage. The phase difference between the two waves, not touching but only embracing the magnetic field confined within the ring magnet, is a definitive confirmation of the Aharonov-Bohm effect.

This experiment has changed our concept of the electromagnetism. The fundamental building block is now the equivalence class of potentials modulo gauge transformation, and not the electric and magnetic fields themselves. The theory of gauge fields, the basis to understand elementary particles, was developed in line with this view.

1.5 Imaging the Space-time Behavior of Magnetic Vortices in Superconductors

Tonomura opened new windows to look into the electrodynamics of a superconductor by utilizing the coherence of his electron beam. In 1989, he used the electron holography to observe a single quantum of magnetic flux (vortex) sticking out from a superconducting lead film [20]. In 1992, he succeeded in observing the vortices, and their dynamics, in a high-temperature superconductor by adopting “coherent beam” Lorentz microscopy [22, 23], which makes use of the Aharonov-Bohm effect. Vortex motion is a key issue also for the technology of high- temperature superconductors, because it can destroy superconductivity.

The use of transmission electron microscopy enabled Tonomura to look inside the magnetic materials, as opposed to the Bitter decoration technique where one can only see the “outcrop” of the lines of force on the surface. However, the low penetration power of the incident electron beam initially used only allowed observing inside a film thinner than the diameter of a vortex.

In 2000, Tonomura developed a microscope [28] with 1 MeV electron beam having the penetration power more than twice as large as the previous one, with which he is now using to bring out features of vortices peculiar to the high-temperature layered superconductors.

2. Detailed Description of Scientific Achievements

The description below overlaps with the text above, but adds more details.

2.1 Development of Electron Phase Microscopy

Tonomura's new methods, the electron holography [1] and the "coherent beam" Lorentz microscopy [2], may generically be called "Electron Phase Microscopy." He used his methods to clarify fundamental questions of quantum mechanics, decisively demonstrating the Aharonov-Bohm effect [3] and bringing to light how the wave-particle duality of electrons is realized in Nature [4]. He also has been applying them extensively to make visible the previously inaccessible microscopic magnetic structure of matter, in particular the space-time behavior of quanta (so-called vortices) of magnetic flux in low- and high-temperature superconductors [2].

The methods make use of coherent electron waves from a pointed field-emission (cold emission) source and their quantum mechanical phase shifts by applying either the holography principle or "coherent-beam" Lorentz microscopy. More precisely, Tonomura's electrons are of such high-energy that eikonal approximation is valid and hence the phase can be defined for each one of their paths as the line integral along the path of electromagnetic potentials. Then, the difference between the phase shifts of two paths, originating from a point source and meeting at a common point on the image plane, determines their interference. Therefore, the holography principle can measure the line integral along a path of electrons of vector and scalar potentials inside a material by taking the other path lying outside (where there are no potentials), and the "coherent-beam" Lorentz microscopy compares the line integrals along two paths embracing a vortex. This is the Lorentz microscopy making use of the Aharonov-Bohm effect.

To realize the electron holography, the coherence of the electron waves is a must

and therefore monochromaticity of the beam and also a point source are required. Monochromaticity is realized by using cold emission (field emission), and the point source is realized by ingeniously making a tip of radius less than $0.1\mu\text{m}$. Furthermore, Tonomura had to keep the tip free from the slightest vibration and also the beam free from the slightest disturbances due to stray magnetic fields. The acceleration voltage of Crewe's electron gun, for his scanning microscope, was 3×10^4 V, while Tonomura needed, at least, 10^5 volts. In addition, he also needed a much larger current to uniformly and coherently illuminate the whole specimen region to be observed [7]. These are just a few examples of a large number of challenging problems that Tonomura had to solve. Indeed, it took more than ten years of effort for Tonomura to realize his first practical electron holography microscope in 1979 [8]. Since then, Tonomura has been working steadily to improve his methods and apparatuses to make them very useful in studying materials and physical phenomena. By increasing the beam brightness, for example, the precision in the phase measurement has been improved from $\lambda/4$ to $\lambda/100$.

With his coherent beam, in addition, Tonomura developed Lorentz “phase” microscopy, which allows one to see the space-time behavior of magnetic vortices inside superconductors, as if these were dancing in front of the observer. The acceleration voltage of the electron has reached 1 MV (million volts) to increase the penetrating power of the beam to such an extent that the inside of the high-temperature superconductor film of thickness of physical interest can be studied.

Thus, by relentlessly focusing, for decades, on solving numerous very challenging problems to realize the electron phase microscopy, he has secured his own field of work in the world: so far nobody else can make visible the spatio-temporal dynamics of vortices in high-temperature superconductors.

Below we shall briefly describe some details of a few selected results of his research. Additional details are presented in the published papers by Tonomura.

2.2 Particle-wave Duality

Tonomura demonstrated in 1987 how the two-slit interference pattern emerges from electrons detected one by one, using his coherent electron beam [4]. Feynman once wrote that this experiment has in it the heart of quantum mechanics, and at the

same time that one should not try to set up this experiment because the apparatus would have to be made on an impossibly small scale [4a]. Tonomura succeeded in doing this experiment, at the heart of quantum mechanics, and that it was thought to be impossible to make. The coherence of Tonomura's electron beam made it possible to do the experiment with the two slits replaced by an electron biprism which consisted of a fine wire of diameter less than $1\mu\text{m}$ (positive potential) placed in between two electrodes (negative potential) 1 cm apart. Coherent electron waves from a field emission tip were sent to the electron biprism, and the biprism interference pattern was enlarged by the electron microscope, to be observed by a two-dimensional position-sensitive electron counting system. The electrons were counted one by one. The electron arrival rate was approximately 10^3 electrons per second, so that there is very little chance for two or more electrons to be present simultaneously between the source and the detector (hence the interference pattern cannot be ascribed to electron-electron interaction).

Tonomura saw the single-electron buildup process of the interference pattern on a TV monitor connected to the counter. In the beginning, when the number of electron arrivals was small, the landing points appeared to be distributed at random. But, as the number of arrivals increased, interference fringes gradually appeared and became quite complete when the number reached 70,000. This experiment clearly demonstrated how the particle-wave duality of the electron shows up.

2.3 Developing Electron Holography

Tonomura demonstrated in 1968 that the electron holography is indeed possible [9], opening up a new era of the electron microscopy. The decisive step was taken in 1979 when he got the idea of using a field-emission electron gun to realize extremely high coherence [8]. How high the coherence was could be demonstrated most impressively by the 3,000 interference fringes the beam was able to record on film [8]. The coherence improved the resolution of the electron holography dramatically [10], fulfilling the dream of D. Gabor in 1949 [5] to overcome the resolution limit inherent to the traditional electron microscopy. Moreover, the electron phases were found to be very sensitive to magnetic fields, enabling him to use the electron holography and his coherent-beam Lorentz microscopy to look quantitatively into the microscopic magnetic structure of materials [11][12][13].

Applications range from solid state physics to practical materials research , such as the quantitative measurement of the spatio-temporal dynamics of magnetic vortices in high-temperature superconductors, magnetic tape development [14][15] , and thickness measurements at the atomic scale [16]. Reference [16] reports that the optical phase-difference amplification method, as applied to electron holography, achieved the detection of one atomic layer difference in thickness of molybdenite thin films. Electron phase microscopy is applied to many problems, several listed in Tonomura's list of publications (e.g., the reviews in [1]).

2.4 Definitive Confirmation of the Aharonov-Bohm Effect

Tonomura used his coherent beam to solve questions fundamental to quantum mechanics, revolutionizing our concept of electromagnetism.

Aharonov and Bohm [17] pointed out in 1959 that an electron wave function should be affected by a magnetic flux even if the flux was confined in a region inaccessible to the electron. More precisely, suppose a magnetic field is confined in a region. Then, two electron waves embracing the region without ever penetrating into it can develop a phase difference depending upon the strength of the confined field.

Since this effect was in contradiction with the traditional view that an electron is affected by the field at the very position where the electron is, their paper generated a long lasting disputes [18]. There were experiments supporting Aharonov and Bohm's prediction, but numerous critics emphasized the leakage fields that are unavoidable for any bar magnet or any solenoid of finite length.

Tonomura's experiment in 1986 [3][19] settled the issue by making use of a ring magnet covered with a superconducting film and with a copper film, the former guaranteeing no leakage field by its Meissner effect and the latter keeping electron waves from penetrating into magnet. The idea was ingenious and the fabrication techniques unrivaled. The ring must be minute with inner radius of $2\mu\text{m}$ and outer one of $4\mu\text{m}$ so that the two electron waves, one passing inside the ring and the other outside, can interfere with the reference wave passing far away from the magnet. The phase difference was detected between the two, under the condition of no flux leakage, by measuring the relative shift of interference fringes those two waves make when superposed respectively with the reference waves. They did not find any phase difference when the ring was not magnetized, but they found a phase difference when

magnetized. The phase difference they found between the two waves, not touching but only embracing the magnetic field confined within the ring magnet, is a definitive confirmation of the reality of the Aharonov-Bohm effect. As a byproduct, it was directly verified, by quantitative measurement, that the magnetic flux confined by a superconductor was quantized.

His definitive confirmation of the AB effect revolutionized our concept of the electromagnetic field. The equivalence class of potentials modulo a “gauge” transformation, and not the electric and magnetic fields themselves is now regarded to be the fundamental building block. The theory of elementary particles has been developing based on this idea of “gauge” fields.

2.5 Imaging Space-time Behavior of Magnetic Vortices in Superconductors

By using the phase information of electron waves passing through superconductors, Tonomura opened new windows to look into its electrodynamics, as opposed to the Bitter decoration technique with which one can see only the fixed and static “outcrop” of magnetic lines of force on the surface.

2.5.1 Until the Year 2000

In 1989, he used the electron holography to observe a single quantum of magnetic flux (vortex) sticking out from a superconducting lead film [20], and accomplished in 1991 observation of the dynamics of the vortices [21]. In 1992, Tonomura succeeded in observing the vortices in a superconductor by adopting a new “coherent beam” Lorentz microscopy, simultaneously revealing their static lattice patterns and their dynamical behavior in real time [22, 23]. The dynamics of the vortices is an important issue also in the application of the high-temperature superconductors, because vortex motion can be induced by a current, generating heat and eventually destroying superconductivity. Indeed, vortices can move very easily in some of the high-temperature superconductors.

In the 1993 article “Heroic Holograms” in Nature [24], D.J. Bishop wrote, “They [Tonomura et al.] are the first to generate real-space, real-time images of a melting magnetic flux-line lattice in a type II superconductor. It is an experimental tour de force.” Many new discoveries followed such as vortex lattices fitting an artificial regular lattice of defects, at a series of matching strengths of applied magnetic fields [25], and unconventional vortex dynamics, migration and hopping [26] peculiar to

high-temperature superconductors, i.e., completely different types of migration and hopping depending on the temperatures and magnetic fields. The electron holography was also used to look at the magnetic field structure of a single vortex [27].

2.5.2 After the Year 2000

The low penetration power of the electron beam so far used allowed only observing inside of a film thinner than the diameter of the vortex, which is insufficient to look into the layered structure of the high-temperature superconductors. In 2000, Tonomura developed a remarkable microscope [28] with 1 MV electron beam, having more than twice the penetration power of that of the 300 kV one he previously used. He had to overcome many difficulties. First of all, the virtual source size of his field-emission (cold-emission) electron beam has to be so minute as to have a diameter less than $5 \times 10^{-3} \mu\text{m}$ to realize the coherence of electrons he needed, and correspondingly the huge structure of the 1 MV machine has to be kept vibration-free, so that the relative displacement between the minute tip of the electron source and the image plane be smaller than $10^{-3} \mu\text{m}$. This is extremely difficult to achieve. At the same time, the stray electromagnetic fields in the microscope had to be carefully removed. Eventually, he achieved the highest coherence and also the highest brightness ever attained, increasing the maximum number of interference fringes increased up to 11,000 [29]. Moreover, a record of the lattice resolution limit of $0.497 \times 10^{-10} \text{ m}$ was obtained by improving the monochromaticity of the electron beam [30].

With this powerful new instrument, Tonomura brought out features of vortices inside the high-temperature superconductors.

In a cleaved film of a high-temperature superconductor $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi-2212) with tilted columnar defects sparsely made by irradiation with Au^{15+} , and under an applied magnetic field, he discovered two kinds of vortices, one trapped along the tilted columnar defects and the other perpendicular to the film [31]. When the applied magnetic field was tilted, vortices lying along the columnar defects remained trapped. Even after the applied field was removed, they remained there while those perpendicular to the film left. These findings vividly showed that the trapping of the vortices by the columnar defects can be strong and effective. When he lowered the temperature of the superconductor to reach 12 K, he found to his surprise that the vortices lying along the tilted columnar defects “stood up” perpendicularly to the film

[32]. This would show that there were numerous minor defects which became effective at the low temperature. He studied further the effects of these minor defects. Theoretical analyses of the Lorentz microscopy images of vortices were made. To obtain satisfactory agreement with experiments, it was found important to take the anisotropy of the material into account [33, 34].

Without the defects, the vortices form a triangular lattice when the magnetic field is applied perpendicularly to the film. But, if the field is tilted, then, in the case of Bi-2212, linear chains of vortices run through the triangular lattice.

Tomomura discovered that, below a temperature T_d , which depends on the applied field, the chains appeared to be cut here and there with some of their vortices disappearing. He interpreted this phenomenon not as vanishing vortices, but by considering longitudinal vibrations that happened for the vortices located incommensurate to the triangular lattice [35].

When the applied magnetic field is tilted away from the normal to the layer plane of their layered structure, all the vortex lines in YBCO ($\text{YBaCu}_3\text{O}_{7.8}$) are tilted while those in Bi-2212 ($\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$) consist of two kinds, one lying parallel to the layer plane (the Josephson vortices) and the other perpendicularly to the layer plane (the pancake vortices). Only those perpendicular vortex lines that cross the Josephson vortices line up closely thus forming chains [36]. This discovery could be made because Tomomura's beam could look inside the superconductor film relatively thick. Any other observation methods can detect only the outcrop of vortices on the surface, hence unable to determine vortex tilting inside superconductors.

In 2005, Dr. Tomomura's group achieved [37] the first direct observation of vortex-motion-control in superconductors (see, e.g., [38] and references therein). Using Lorentz microscopy to directly image vortices, he investigated vortex motion control and rectification in a niobium superconductor [37]. He directly observed a net motion of vortices along microfabricated channels with a spatially asymmetric potential, even though the vortices were driven by an oscillatory field. By observing the individual motion of vortices, he clarified elementary processes involved in this rectification. To further demonstrate the ability to control the motion of vortices, he created a tiny vortex "racetrack" to monitor the motion of vortices in a closed circuit channel.

These are just a few of the results of Tonomura's development of the electron phase microscopy. Many other applications, not included here for brevity, are described in the references below (a few examples are, e.g., in [39-43]). If history is any guide to future developments, microscopy will continue to grow and blossom in the future. Further developments of the electron phase microscopy, with this high energy and bright electron beam, are necessary in order to push microscopy into a new regime, where it can observe the spatio-temporal dynamics of phenomena never seen before, including the visualization of quantum phenomena in three dimensions.

(Text and references provided by Dr. Yoshimasa A. Ono, RIKEN)

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