Vortex Matter Research Using Electron Microscopy: Memorial to Akira Tonomura

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Abstract **- Using electron holography and "coherent beam" Lorentz microscopy, Akira Tonomura studied vortex physics in metal and high-temperature superconductors for more than 20 years. The new methodology he introduced involved coherent electron waves from cold emission (field emission) sources and their quantum mechanical phase shifts. Using 300 kV and 1 MV electron microscopes Tonomura and his collaborators studied dynamic behavior of vortices in metal superconductors, Pb and Nb, and in high-temperature superconductors, $YBa_2Cu_3O_{7-\delta}$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$. In this memorial paper for Akira Tonomura the static and dynamic vortex behavior in superconductors is reviewed based on the group's results.**

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I. INTRODUCTION

This paper is a memorial publication to honor the memory of Akira Tonomura* by describing the achievements of his group in the field of superconductivity research, in particular in the observation of vortices (magnetic fluxes) in metal and high-temperature superconductors using electron microscopy. Tonomura developed new methods of electron microscopy [1-5], the electron holography and "coherent beam" Lorentz microscopy, which make use of the wave nature of electrons. These methods may be generally described as "electron phase microscopy" and make use of coherent electron waves from a pointed field-emission (cold emission) source and their quantum mechanical phase shifts. 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 the two paths, originating from a point source and meeting at a common point on the image plane, determines their interference. Therefore, the holography principle permits one to measure the line integral along a path of electrons of vector and scalar potentials inside a material by taking the line integral along the other path lying outside (where there are no potentials) as the reference. The coherent beam Lorentz microscopy compares the line integral along two paths embracing magnetic domain walls or magnetic fluxes in materials.

The basic idea of the electron holography was put forward by D. Gabor [6] in 1949. Also, the first practical use of the pointed field-emission electron source was made by A. V. Crewe [7] in 1968 for his scanning electron microscope. Moreover, incoherent beam Lorentz microscopy for the observation of magnetic domain structures 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 it would have been very difficult to see the microscopic magnetic structure inside materials quantitatively and directly.

To realize the electron holography, 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 100 nm. Furthermore, Tonomura had to keep the tip free from the slightest vibration and also the beam free from the slightest disturbances due to stray electromagnetic fields. The acceleration voltage of Crewe's electron gun for his scanning electron microscope was 30 kV, while Tonomura needed, at least, 100

* Dr. Akira Tonomura passed away on May 2nd, 2012. In addition to his obituary published by ESNF on that day, we provide one written by Christian Colliex, Past President of IFSM (International Federation of Societies for Microscopy).

kV for his holography electron microscope. In addition, he also needed a much larger current to uniformly and coherently illuminate the whole specimen region to be observed. 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] equipped with a highly coherent beam from the field-emission electron gun. The coherence improved the resolution of the electron holography dramatically, fulfilling the dream of Gabor to overcome resolution limit inherent to the traditional electron microscopy. Moreover, electron phases were found to be very sensitive to magnetic fields, enabling Tonomura to use his coherent beam to look quantitatively into the microscopic magnetic structure of materials [9-11]. With his coherent beam, in addition, Tonomura developed coherent beam Lorentz microscopy, which allows one to see dynamic behavior of magnetic vortices inside superconductors. He used these methods to clarify fundamental questions of quantum mechanics, demonstrating the Aharonov-Bohm effect decisively [12,13] and bringing to light how the wave-particle duality of electrons is realized in Nature [14]. Recently he has applied these methods extensively to make visible the microscopic structure of matter that has been so far inaccessible [4], including the space-time behavior of the quanta (so-called vortices) of magnetic flux in metal and high-temperature superconductors [15-17], The methods have been also used in magnetic tape and head developments [18-20], and thickness measurements at the atomic scale [21].

II. IMAGING THE 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. The use of transmission electron microscopy enabled Tonomura to look inside the magnetic materials, as opposed to the Bitter decoration technique, where one can see only the fixed and static “outcrop” of magnetic lines of force on the surface.

Figure 1(a) shows the optical system of electron holography for vortex observation [2]. Though not shown in Figure 1, low-temperature specimen stages are necessary for observing vortices in superconductors, and additional magnetic-field application systems [5, 22] are indispensable for observing both the magnetization process of ferromagnetic samples and the dynamics of vortices in superconducting samples. For such purposes, a low-temperature stage with a magnetic field application system was developed. This stage can hold a specimen at an arbitrary temperature between 5 and 100K for three hours. A magnetic field up to 50 mT can be applied to the sample in any arbitrary direction. Electric current can also be applied to the sample. For vortex observation, it is necessary to have high-temperature superconducting samples with a large uniform thin flat area, for example, samples with the area of 100x100 μm^2 and thickness of 200 nm for $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) observation [23] or with the area of 30x30 μm^2 and thickness of 400 nm for $YBa_2Cu_3O_{7-\delta}$ (Y-123) observation. Superconducting samples cooled down below the transition temperature T_c are located in the half space. The positively charged filament electrode of the electron bi-prism attracts the incident electron beams of the object wave that penetrates the tilted superconducting thin film and the reference wave which passes through the vacuum region. Both waves overlap under the bi-prism to create interference fringes. These interference fringes are recorded as the electron hologram under the in-focus condition for the specimen. After a reconstruction procedure, for example by a Fourier transform algorithm, magnetic lines of force are visualized as the electron phase distribution.

Figure 1(b) shows the optical system of Lorentz microscopy for vortex observation, in particular for observation of vortex dynamics [2]. Since vortices are complete phase objects for the electron beam, no contrast is produced in in-focus images. However, electron beam that passes through the magnetic field at the vortices inside the tilted superconducting thin film is deflected slightly and the other electron beam which penetrates the magnetic-field-free region (the Meissner region of the type-II superconductor where no vortices exist) is not deflected at all. When these two electron beams are overlapped and interfere at an out-of-focus plane, then the phase modulation by the vortices is visualized as intensity variation depending on the vortex location. Each single vortex can be visualized as a globules-like black and white contrast feature. That contrast feature depends on the polarity of the vortices and the defocusing direction (over-focus or under-focus), and the intensity of the contrast depends on the defocusing distance. We note that the size of a black and white contrast feature is not equal to that of an actual vortex, because the quantification of Lorentz images requires indirect model-based approaches for interpreting image contrast. This experimental setup of out-of-focus imaging method is similar to the domain structure observation of the ordinary ferromagnetic thin-film materials. For vortex observation, however, coherent electron beams with parallel illumination are required, because they have a very small spatial size of about 100 nm in diameter and the deflection angle of the traversing electron beam is very small (~ 10^{-6} rad). Therefore, the defocus distance for vortex observation is approximately 100 mm, while that for magnetic domain structure observation is approximately 100 μm.

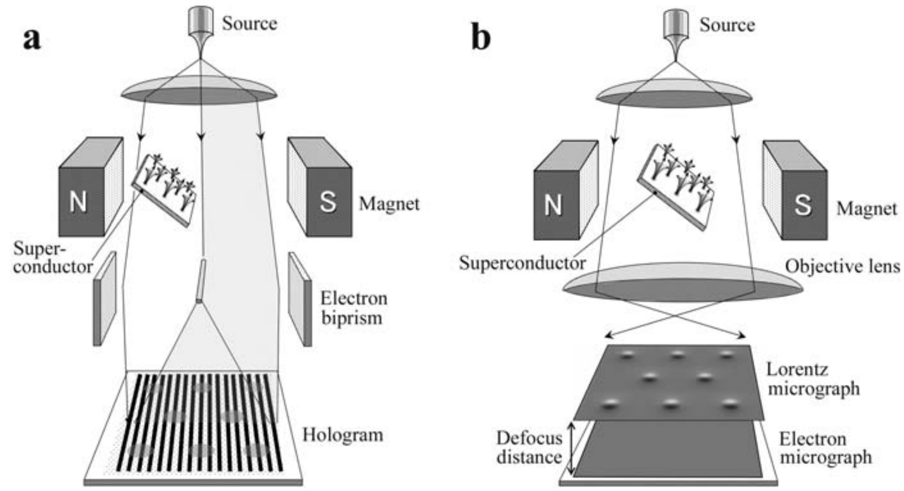

Fig. 1. Optical systems for vortex observation: (a) electron holography;
(b) Lorentz microscopy. The magnetic field of the magnet induces the vortices in the superconductor.

Unfortunately the low penetration power of the incident electron beam in the 300-kV electron microscope initially used in these experiments allowed one to observe vortices only inside films thinner than the vortex radius (the penetration depth). This is insufficient to look into layered structures of high-temperature superconductors.

In 2000, Tonomura and his collaborators developed a microscope with 1-MeV electron beam having a penetration power more than twice as large as the previous one [24, 25] so that the inside of the high-temperature superconductor thin films with thickness of physical interest can be studied. Since then they have been using this unique microscope to bring out features of vortices peculiar to the high-temperature layered superconductor crystal structure.

III. VORTEX OBSERVATIONS UNTIL THE YEAR 2000

In 1989, a single quantum of magnetic flux (vortex) in a superconducting lead (Pb) film was for the first time observed by using electron holography [26]. Figure 2 shows interference micrographs of magnetic lines of force for Pb films with 0.2-μm thickness (Fig.2a) and with 1.0-μm thickness (Fig. 2b).

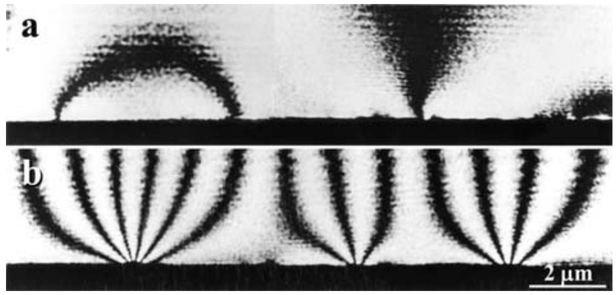

Fig. 2. Interference micrographs of magnetic fluxes penetrating superconducting Pb films. Film thickness: (a) 0.2 μm; (b) 1.0 μm.

The right pattern in Fig. 2(a) shows a magnetic line from a single quantized vortex with a flux of $h/2e$ corresponding to the phase shift of π, while the left side pattern shows a pair of vortices oriented in opposite directions and connected by lines of magnetic field. The vortex pair creation is probably due to the Kosterlitz-Thouless transition [27]: during the cooling process, an antiparallel vortex pair appeared and disappeared repeatedly due to thermal excitation; it was eventually pinned at some imperfections and frozen. The behavior of magnetic flux in the thicker film of Fig. 2(b) is completely different: magnetic fluxes penetrate the superconductor in bundles and no vortex pairs were observed. This shows that a superconducting Pb film with less than 0.5 μm thickness behaves like a type-II superconductor and magnetic flux penetrates the superconductor in the form of individual vortices. In 1991, the first observation of vortex dynamics were reported [28].

In 1992, vortices in a Nb superconducting thin film were observed [29] by adopting a new "coherent beam" Lorentz microscopy, simultaneously revealing their static lattice patterns and their dynamical behavior in real time. Figure 3 shows the result: the specimen was field-cooled at 10 mT down to 4.5 K. Tiny globules-like features of black and white contrast indicate single vortices. When the applied magnetic field was changed, vortices began to move. However, those trapped at strong pinning centers in the film did not move easily.

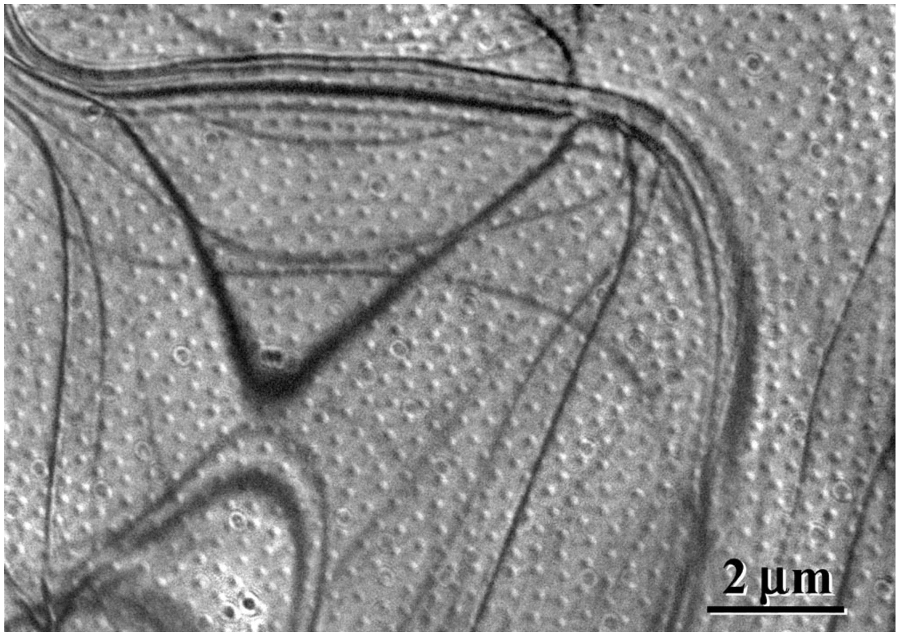

Fig. 3. Lorentz micrograph of a Nb thin film. B = 10 mT, T = 4.5 K.

The dynamics of the vortices is an important issue also in the application of high-temperature superconductors, because vortex motion can be induced by a current, thus generating heat and eventually destroying superconductivity. Indeed, vortices can move very easily in some of the high-temperature superconductors. In 1993, using the same technique, vortices and their dynamics were observed in a high-temperature superconducting thin films of $Bi_2Sr_{1.8}CaCu_2O_\chi$ [23].

In the 1993 article "Heroic Holograms" published in *Nature* [30], D.J. Bishop wrote the following:

> "They [Tonomura and his collaborators] 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 based on Lorentz microscopy observation. In 1996, dynamic interactions of vortices with artificial pinning centers in a Nb thin film were reported [31], resulting in vortex lattice movements in the form of vortex "rivers" when the temperature increased. This is illustrated by video frames of Figure 4.

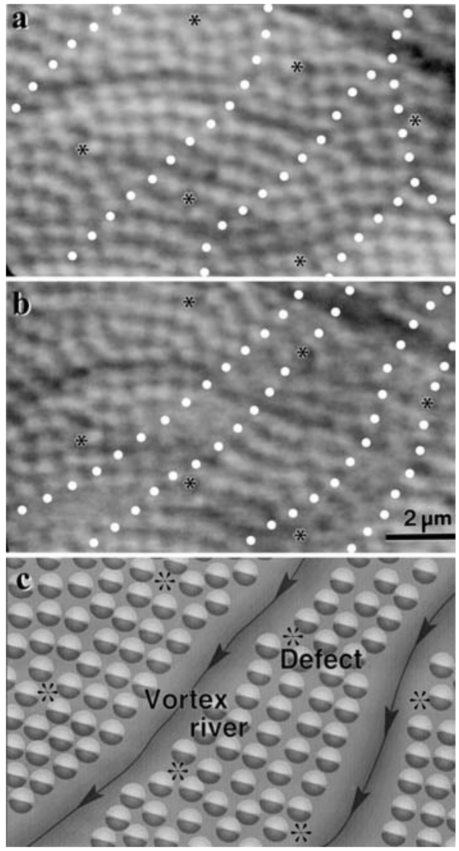

Fig. 4. Video frames and an illustration of vortex lattice movement. (a) t = 0.0 s; (b) t = 0.43 s; (c) illustration of the vortex "rivers". Asterisks indicate defects. White dots in (a, b) show the domain boundaries.

When strong pinning centers exist, vortices form lattice grains with boundaries, because some vortices were strongly fixed by the pinning centers. When the temperature was raised, the lattice resisted moving for a while, and then suddenly some domain boundaries began moving like avalanches (compare video frames "a" and "b" in Figure 4), breaking down into pieces and flowing like "rivers" along the domain boundaries located near the defects. At higher temperatures, the river became wider until the vortex lattice flowed as a whole while keeping its lattice form. These observations enabled them to visually monitor the transition from elastic to plastic flow of vortices [31, 32].

Also in 1996, matching effects were visualized where vortex lattices fitted an artificial regular lattice of defects at a series of matching strengths of applied magnetic fields [33]. Dynamic behavior of vortices was observed when the external magnetic field was increased.

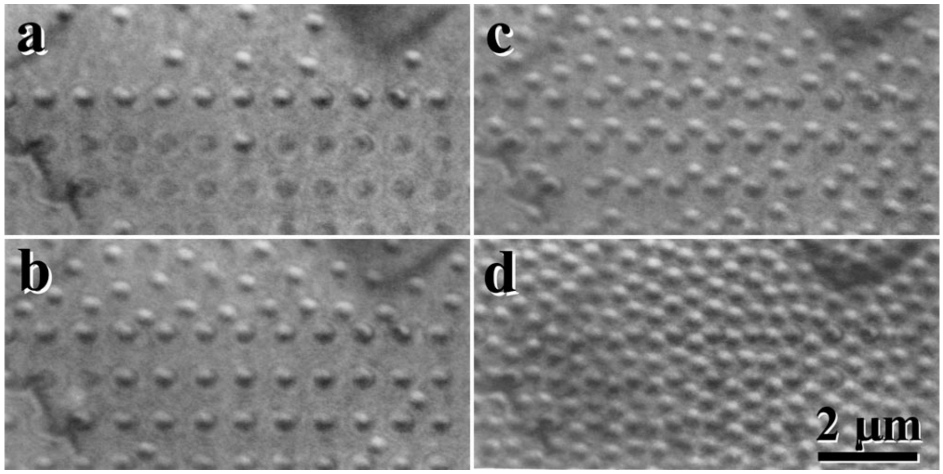

Fig. 5. Lorentz micrographs of vortex dynamics with the increase in magnetic field at 4.5 K. (a) 20 μT; (b) 50 μT ; (c) 0.12 mT; (d) 0.25 mT.

As shown in Figure 5, at first vortices occupied the front row of defects and subsequent vortices were stopped by this barrier. With increasing field, the accumulated vortices suddenly broke through this barrier and jumped to defects located far away. After the distant defects were occupied, vortices began to jump to the nearer defects.

In 1997, the vortex-antivortex pair annihilation in Nb thin film was found when the applied magnetic field was reversed [34]. When the magnetic field was switched off, about 10% of the vortices were trapped at pinning centers inside the Nb film. With the application of the magnetic field in the opposite direction, vortices with the opposite direction, *i.e.*, anti-vortices, hopped into the film. When these anti-vortices collided with the trapped vortices, they annihilated as shown in Figure 6.

In 1999, Tonomura and his collaborators [35] discussed unconventional vortex dynamics, migration and hopping 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 in the 2D vortex lattice [36]. The obtained results have been interpreted by comparing them with calculations based on a theoretical model accounting for the finite core size of the vortices. The calculated phase difference across a singly quantized vortex agreed with the measured value of 0.55 π, corresponding to a flux of 1/2 $h/2e$, where the preceding factor of 1/2 comes from the 45-degree tilting of the superconducting thin film.

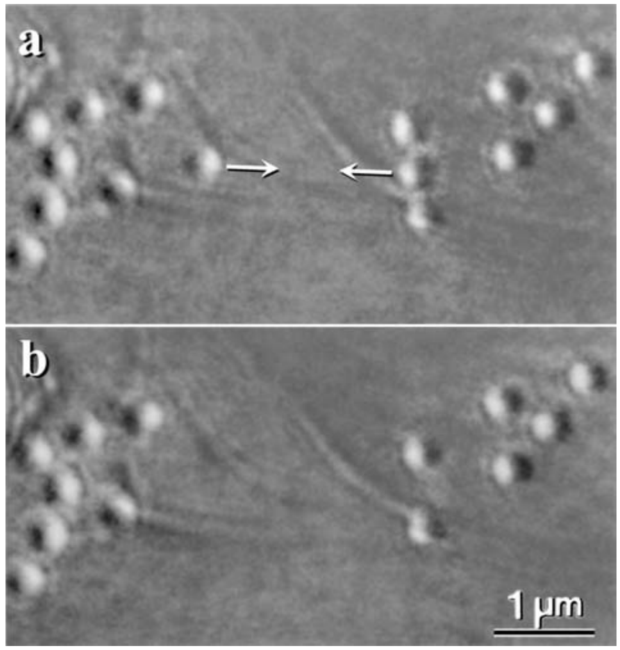

Fig. 6. Vortex-antivortex pair annihilation:
(a) before annihilation; (b) after annihilation.

IV. VORTEX OBSERVATIONS AFTER THE YEAR 2000

In 2000, Tonomura and his collaborators developed the above-mentioned unique microscope [24, 25] with 1 MV electron beams, having more than twice the penetration power of that of the previous electron microscope. They had to overcome many difficulties: The virtual source size of the field-emission (cold-emission) electron beam must be so minute as to have a diameter less than 5 nm to realize the coherence of electrons needed in the experiment, and correspondingly the huge structure of the 1 MV machine must be kept vibration-free. At the same time, the stray electromagnetic fields in the microscope had to be carefully removed. Eventually, the highest coherence and the highest brightness ever were attained.

With this powerful new instrument, Tonomura and his collaborators brought out features of vortices inside high-temperature superconductors. For example, they were successful in obtaining an improved vortex lattice micrograph of a high-temperature superconductor Bi-2212 using Lorentz microscopy. Such a micrograph is shown in Figure 7.

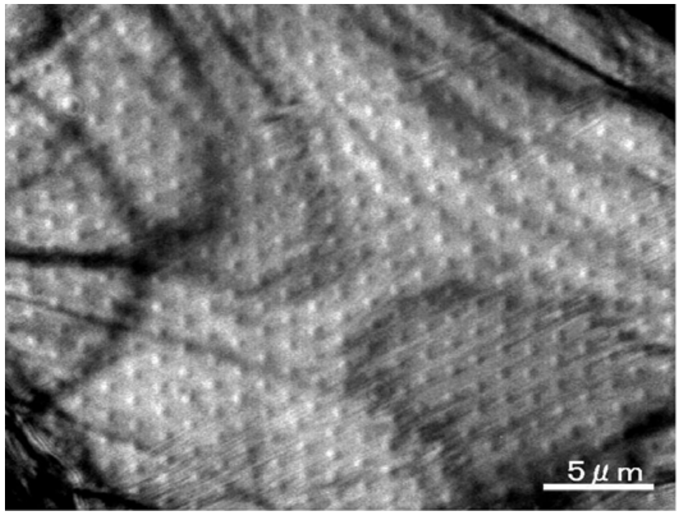

Fig. 7. Lorentz micrograph of the high-temperature superconductor $Bi_2Sr_2CaCu_2O_{8+\delta}$.

B = 1.5 mT, T = 5 K

In a cleaved Bi-2212 film with tilted and sparsely distributed columnar defects created by irradiating the sample with Au^{15+} ions at 240 MeV, Tonomura and his collaborators discovered two kinds of vortices in applied magnetic fields: one trapped along the tilted columnar defects and the other perpendicular to the film [37]. When the applied magnetic field was tilted, vortices aligned with the columnar defects remained trapped. Even after the applied field was removed they remained there while those perpendicular to the film left. These observations vividly showed that the trapping of the vortices by the columnar defects can be strong and effective.

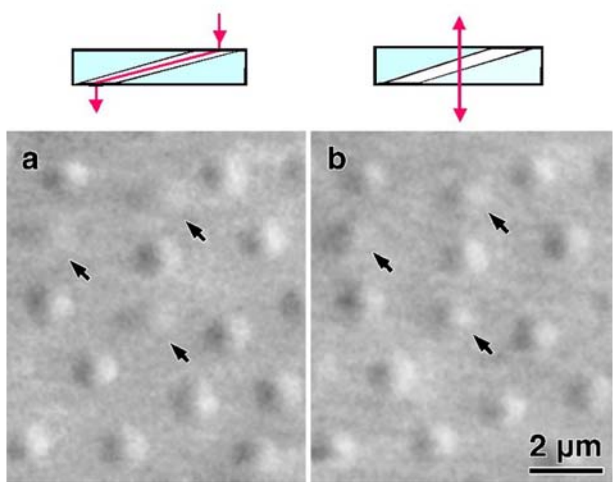

Fig. 8. Lorentz micrographs of vortex–line arrangements: (a) 35 K and (b) 10 K.

However, when the temperature was decreased from 35 K down to 12 K, the contrast of the trapped vortices changed from elongated to circular as shown in Figure 8. This was a surprising result: the vortices lying along the tilted columnar defects "stood up" perpendicularly to the film [38]. This shows that there were numerous minor atomic-size defects that became effective at lower temperatures. They studied further the effect of these minor defects. Theoretical analyses of the Lorentz microscopy images of vortices were performed [39-41]. To obtain satisfactory agreement with experiments, they found it important to take the anisotropy of the material into account.

Without defects, vortices form a triangular lattice when the magnetic field is applied perpendicularly to the film. When the magnetic field was applied obliquely to the layer plane, however, vortices were observed (at Bell Laboratories) using the Bitter decoration method, to form alternate domains of linear chains and triangular lattices in Bi-2212 [42] and linear chains in Y-123 [43]. Tonomura and his collaborators could not observe these chain structures when using the 300 kV electron microscope. However, the 1 MV electron microscope made it possible to observe, under a tilted magnetic field condition, linear chains of vortices running through the triangular lattice (chain-lattice state) in Bi-2212 [44] and the chain vortices in Y-123 [45] as shown in Figure 9.

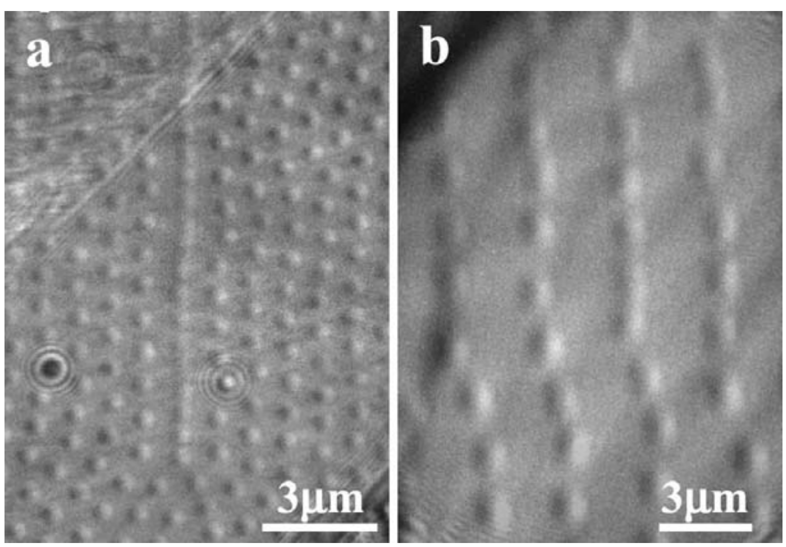

Fig. 9. Lorentz micrographs of vortices in high-temperature superconductors: (a) chain-lattice state in Bi-2212; (b) chain vortices in Y-123.

In addition, Tonomura and his collaborators discovered that below a certain temperature T_d, which depends on the applied field strength and is much lower than T_c, the chains appeared to be cut here and there with some of their vortices disappearing. They interpreted this phenomenon as characteristic of the chain-lattice state where chains of vortices become incommensurate with the potential distribution. Vortices in such chains can move longitudinally at relatively low temperature and vibrate under thermal excitation [44]. These vibrations are too fast to be detected at TV scanning rate of the detection system.

In studying these vortices inside the superconducting samples, they found that when the applied magnetic field was tilted away from the normal to the layer plane of their layered structure, all the vortex lines in Y-123 were tilted while those in Bi-2212 consisted of two kinds of vortices: one lying parallel to the layer plane (Josephson vortices) and the other perpendicular to the layer plane (pancake vortices). These findings are in accordance with the theoretical prediction of Koshelev [46]. Only those perpendicular vortex lines that cross the Josephson vortices line up closely thus forming chains [45]. Furthermore, they found unexpected results: images of chain vortices began to disappear at lower temperatures than T_c. This phenomenon was explained by incommensurate chain vortices oscillating along the chain direction owing to thermal vibrations of vortices.

These discoveries were made possible thanks to the development of 1 MV electron microscope that can observe vortex behavior inside thick high-temperature superconducting films by transmitting electron beams through them. All other observation methods can detect only the outcrop of vortices on the surfaces, hence are unable to determine vortex tilting inside superconductors.

In 2005, a direct vortex-motion control in superconductors was realized [47] (see also [48]). Using Lorentz microscopy to directly image vortices, Tonomura and his collaborators studied vortex-motion control and rectification in a Nb superconductor. They directly observed a net motion of vortices along the microfabricated channels with a spatially asymmetric potential, even though the vortices were driven by an oscillatory field. By observing the individual motion of vortices, they clarified elementary processes involved in this rectification. To further demonstrate the ability to control the motion of vortices, they created a tiny vortex “racetrack” to monitor the motion of vortices in a closed circuit channel.

In 2008, Tonomura reported the first observation of the static and dynamic behavior of interlayer vortices, or Josephson vortices, in Y-123 thin films [49].

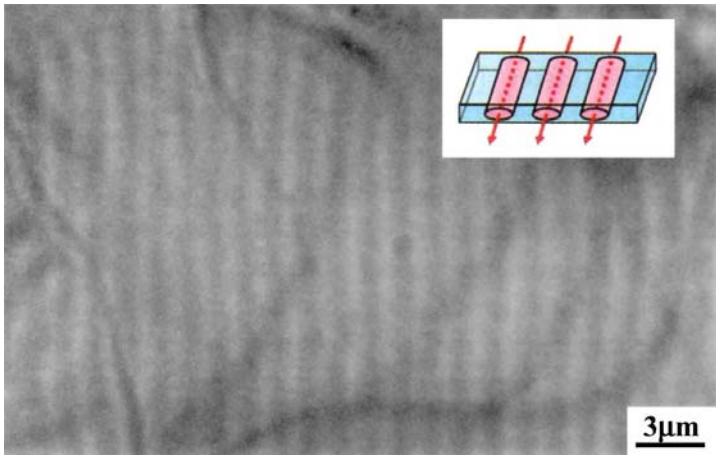

Fig. 10. Interlayer vortices (Josephson vortices) in Y-123 thin film.

Figure 10 shows Lorentz micrograph of the interlayer vortices in Y-123 ab-plane film thicker than 0.4 μm. Each line in the micrograph corresponds to an individual interlayer vortex. The enclosed magnetic flux of $h/2e$ inside a single line was confirmed by measuring a relative phase shift of π between two electron beams passing through on both sides of a single line. When the applied magnetic field parallel to the film plane was increased, the density of vortex lines increased proportionally.

V. CONCLUDING REMARKS

By developing the electron phase microscopy for more than four decades through relentlessly focusing on solving numerous very challenging problems, Tonomura and his collaborators have succeeded in observing previously unobservable microscopic objects; microscopic distribution of magnetic lines of force in h/e units in magnetic materials and vortex structures in metal superconducting thin films by electron holography, and the dynamics of quantized vortices in metal and high-temperature superconducting thin films by "coherent beam" Lorentz microscopy. Tonomura used electron waves to attain an achievement that is recorded in the history book of science. There was no end to Tonomura's challenging desire, to see worlds that nobody else has seen. In March 2010 Tonomura received a Japanese government grant of six-trillion-yen, (over 50 million US dollars) budgeted over five years, for the "FIRST Tonomura project" to develop an atomic-resolution holography electron microscope [50]. With this 1.2 MV holography electron microscope Tonomura was determined to realize his dream of reconstructing the wavefronts of electrons scattered by atoms, first proposed by Gabor in the 1940s, and to observe the atomic arrangement in three dimensions and also to elucidate quantum phenomena often appearing on microscopic scales.

To conclude this review in memory of Akira Tonomura, we quote some of his comments on R&D:

"You have to develop new equipment when you attack a new problem."

"You must have 'new ideas' and 'persistence' to attain your goal."

"'Finding the cause and 'solving riddles' is indeed the real thrill of research and development."

"We started with a strong determination that 'We'll try everything and anything to prove the Aharonov-Bohm effect.'"

"Technical difficulty is no excuse."

"Research results must be 'the first in the world' and imitations have no value."

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† In deference to the group effort overviewed in this paper, ESNF exceptionally publishes the full lists of authors' names appearing on the joint publication.

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