# "The Picture is My Life"

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This article is dedicated to the memory of the late Dr. Akira Tonomura, with whom I worked at Hitachi Advanced Research Laboratory (HARL) from 1985 to 1990. I report my research experience at HARL and at the University of Tokyo after 1990 together with intriguing experimental photographs from my favorite collection taken by myself and my colleagues. These photographs include electron holography micrographs, scanning tunneling micrographs, electron diffraction patterns, photoemission band dispersion, and images of four-tip scanning tunneling microscopes. I conclude with a discussion on my 'beyond seeing' study.

# 1. Introduction

After my graduation from Graduate School of Science, The University of Tokyo with MS, I spent five years as a researcher in Tonomura group at Hitachi Advanced Research Laboratory (HARL). When I joined his group, they were at the final stage of verification experiments of the Aharonov-Bohm effect using a ring magnet covered with a toroidal superconductor. I participated in the sample preparation and optical reconstruction processes. In addition, I engaged in the development of a new computer-controlled optical reconstruction interferometer to enhance the sensitivity in phase measurement of electron waves, which was later used for detecting very weak magnetic fields, such as leakage fields from magnetic recording media and quantized magnetic fluxes penetrating superconductors. Research results from these topics constituted my Ph.D. thesis in 1991.

When we received the galley proof of our Phys. Rev. Lett. (PRL) paper<sup>1</sup> reporting the first observation of quantized magnetic flux lines penetrating a superconducting Pb film, we found out that the holographic micrograph of the flux was in a half-column size. The contents of the micrograph were simple so that I thought the half-column size was acceptable. However, Tonomura-san was not satisfied and wrote to the PRL editor asking for an enlargement to the double-column size with a comment "Since the picture is my

life, please show the micrograph as large as possible in the printed paper." I was strongly impressed by his words at that time that really symbolized his attitude toward research (See Fig. 1 in Ref. 1 for the double-column figure).

After moving to The University of Tokyo as an assistant professor at Department of Physics, I had to give up holography the electron research because I could not purchase an expensive electron microscope. Instead, I started research on surface physics using simple electron diffraction and later extended to the study of electronic transport on crystal surfaces. Just after publishing my first PRL paper on the surface transport<sup>2</sup>, someone remarked that it was the least expensive PRL paper but interesting one. Later I scanning purchased а tunneling microscope (STM) and a photoemission spectroscopy instrument that enabled me to explore the surface physics from different aspects. After establishing my own research group, I started to develop a world-first four-tip STM.<sup>3</sup> Tonomurasan's passion for developing new equipment has been carried on to my research.



Fig. 1. Digital magnetic recording on a CoCr film in the perpendicular magnetization mode, observed by electron holography with phase amplification (Ref.4).

#### 2. Electron holography results

Figure 1 shows reconstructed holographic interference micrographs<sup>4</sup> with its sample structure, showing leakage fields from a CoCr thin film for a digital magnetic recording medium. The bit width was ca. 100 nm and the magnetizations were alternately up or down along the perpendicular direction. The phase sensitivity is enhanced with the new interferometer I developed: single interference fringes in units of h/e (= 4×10<sup>-15</sup> Wb) are as follows: 1/2 in Fig. 1b, 1/10 in Fig. 1c, 1/30 in Fig. 1d, 1/60 in Fig. 1e, and 1/100

in Fig. 1f. Here h is Planck's constant and e is the elementary charge. The measurement resolution of the phase of electron wave is as small as  $2\pi/100$ , which is roughly an order of magnitude improvement compared with the previous measurements.

Figure 2 shows holographic interference micrographs of quantized magnetic fluxes penetrating superconducting Pb films.<sup>5</sup> A single interference fringe corresponds to a single flux quantum h/2e. The magnetic flux structures vary depending on the film thickness d and the applied magnetic field H. For thin films, individual fluxes are made of single quanta, while for thicker films, every flux is made of multi-quanta. While the quantized magnetic fluxes at the surface of superconductors are described only by schematic illustrations in many of the text books of solid-state physics, they should be replaced by the present pictures.



Fig. 2. Quantized magnetic fluxes penetrating superconducting Pb thin films with different thickness *d*, observed by electron holography under an applied transverse magnetic field *H*. The field is increased from the top panel to the bottom panel at each thickness. The dark area in the lower part in each micrograph is the Pb film and the upper part is vacuum (Ref. 5).

# 3. Scanning tunneling microscopy experiment

Figure 3 shows an STM image of a Si crystal surface partially covered with a monolayer of Ag film.<sup>6</sup> The Ag-covered area is denoted by  $\sqrt{3} \times \sqrt{3}$ , while the uncovered area is indicated by 7×7. Fine and regular arrays of protrusions correspond to atomic lattices with different periodicities in two areas. There are meandering atomic steps running from left to right in the center (indicated by (A)) and straight domain boundaries (indicated by (B)). In addition to the regular lattice pattern, we find wavy patterns with about 3 nm wavelength near the peripheries of the  $\sqrt{3} \times \sqrt{3}$  area, which are just like ripples on water surface. These wavy patterns are electron standing waves. Electrons in the monolayer-Ag film freely moves around along the surface and they are reflected by defects such as steps and domain boundaries. The incident wave and reflected wave interfere with each other to form the standing wave. The STM enables imaging not only geometric protrusions at an atomic scale, but also distributions of electron density. Although I had learned the wave nature of electron through electron holography study in the Tonomura group, I was really impressed by this STM picture of electron standing waves on the crystal surface. Electrons confined in narrow domains make concentric standing waves, revealing quantum-well states. This is nothing but a visualization of quantum mechanics.



### 4. Electron diffraction experiment

Fig. 3. STM image of electron standing waves in a monatomic-layer Ag on a Si crystal surface (Ref. 6).

The wave nature of electron was

historically verified as a form of electron diffraction from crystals by Davisson and Germer, Thomson, and Kikuchi. Electron diffraction is now widely used to investigate atomic structures on crystal surfaces. Figure 4 shows reflection-high-energy electron diffraction (RHEED) patterns taken from a single sample, a monatomic-layer In adsorbed on a Si surface, at room temperature (RT) (left figure) and at 100 K (right figure).<sup>7</sup> Diffraction spots in the RT pattern indicate a regular atomic arrangement of a so-called  $4 \times 1$  surface superstructure. This surface structure is now explained by massive arrays of In atomic chains aligned along a particular crystal orientation of the Si substrate, resulting in a quasi-one-dimensional metallic surface. By cooling the sample down to 100 K, several additional spots (indicated by black arrowheads) and streaks (indicated by arrow) appear, showing a surface superstructure of so-called  $8 \times 2'$ . This is a temperature-induced surface phase transition we have found; the nature is believed to have a Peierls-like meta-



Fig. 4. Electron diffraction patterns taken from a monatomic-layer In adsorbed on a Si surface, at room temperature (left) and at 100 K (right), showing a phase transition (Ref. 7).

insulator transition accompanied by a charge-density wave (CDW) formation and lattice distorsion.<sup>7</sup> Its mechanism, however, is not clarified yet.

# 5. Photoemission spectroscopy experiment

While electron microscopy, electron holography, and STM enable real-space imaging, electron diffraction and photoemission spectroscopy enable reciprocal-space imaging.

From Einstein's theory of photoelectric effect, electrons in crystals and on the surfaces are emitted outside by absorbing the energy of irradiated photons. This phenomenon is now widely used as photoemission spectroscopy for analyzing electronic states in/on crystals. We can estimate the initial-state energy levels and momenta of electrons in/on crystals by measuring the kinetic energy and emission angle of the emitted photoelectrons. Then, we can deduce the energy-wavenumber relation (band dispersion), since the wavenumber  $\mathbf{k}$  is defined as momentum  $\mathbf{p}$  divided by  $\hbar$ .

Figures 5(a)-(c) show the band dispersion of Bi thin films of different thicknesses, measured by angle-resolved photoemission spectroscopy (ARPES).<sup>8</sup> The vertical axis expresses the binding energy E with respect to the Fermi energy ( $E_F$ ). The horizontal axis expresses the component of the wavenumber parallel to the surface. We notice that several bands are dispersed, and that with the increase in film thickness, the number of bands increases and the energy separation between the bands becomes smaller. As schematically shown in Fig. 5(d), the electrons are confined in the direction perpendicular to the film surface, which results in formation of quantum-well states and discrete energy levels. The energy separation between the quantized energy levels becomes smaller as the width of quantum well (thickness of the film) becomes larger. On one hand, the electrons in the film can move around freely along the surface, resulting in dispersive bands in the surface-parallel direction. The bands observed in Figs. 5(a)-(c) are



Fig. 5. Bismuth single-crystalline ultrathin films grown on Si, observed by angle-resolved photoemission spectroscopy. (a)-(c) Band dispersion of the films of different thickness, 28 ML (monolayer), 60 ML, and 80 ML thick, respectively. (d) Schematics showing quantum-well states and sub-bands in a thin film (Ref. 8).

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sub-bands of the quantum-well states formed inside the film.

However, the bands crossing  $E_F$  do not change with the film thickness changes, indicating that these metallic states do not come from the inside of the film, but come from the surface of the film. This means that the Bi crystal has a metallic surface though the inside is a semi-metal. Since the metallic surface states are found to have a high electrical conductivity<sup>9</sup> and a spin-split characteristic,<sup>10</sup> existence of the surface state might lead to a possibility of spin-polarized current flowing at the non-magnetic surface.

## 6. Four-tip STM experiment

A single-tip STM enables atomic-scale imaging of structures and electronic density. A multi-tip STM enables us to measure electrical conductivity at a nanometer scale by utilizing these tips as electrodes. Figure 6 shows scanning electron micrographs of tips of the developed four-tip STM.<sup>3, 11</sup> These tips are mechanically and electrically independent of each other and can be controlled by a single PC. Two of the four tips are used for source and drain electrodes and the other two are used for voltage measurement. The tip separation and arrangement can be changed arbitrarily under scanning electron microscope observation. With carbon nanotube tips, the tip separation can be reduced to as small as 20 nm<sup>11</sup> (See the right figure of Fig. 6). This instrument is now commercially available from Unisoku Co. and used world-wide to measure electronic transports on surfaces of nano-materials such as nanowires, graphene, and molecule assemblies.

# 7. Beyond Seeing - Electron/Spin Transports in Relation to Structures-

Microscopy, diffraction, and spectroscopy analyses provide fundamental information on structures and electronic states so that it becomes possible to interpret various kinds of physical and chemical properties. My strategy in research is to combine electronic and spin transport studies with microscopy/diffraction/spectroscopy studies. Before my first



Fig. 6. Scanning electron microscope images of the four tips in the developed four-tip scanning tunneling microscope. Left figure shows W tips (Ref. 3) and right figure shows carbon nanotube tips (Ref. 14).

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paper on this subject<sup>2</sup> in 1992, no one considered electrical conduction at surfaces in relation to surface structures. Twenty years later, no one now doubts the importance of surface-state transport, i.e., electrical conduction only at the topmost atomic layers.

Figure 7 shows resistance of a Si crystal measured as a function of temperature.<sup>12</sup> The surface is covered by (sub)monolayer-indium, and different surface superstructures are created depending on the In coverage;  $\sqrt{3} \times \sqrt{3}$  (1/3 ML),  $\sqrt{31} \times \sqrt{31}$  (2/3 ML),  $4 \times 1$  (1 ML), and  $\sqrt{7} \times \sqrt{3}$  (2 ML). It should be noted that the resistance of a Si crystal having a macroscopic thickness (0.5 mm) changes many orders of magnitude just by adsorption of monoatomic-layer In on the surface. These findings can be obtained only through the surface-sensitive measurements using the developed microscopic four-point probes as shown in Fig. 6. The temperature dependence of resistance is also quite different depending on the surface structures. These properties can be explained from the surface-state bands revealed by photoemission spectroscopy. The  $\sqrt{7} \times \sqrt{3}$  surface superstructure shows superconducting transition<sup>13</sup> at 2.8 K (See the right figure in Fig. 7). This is the first example of surface-state superconductivity.

# 8. Concluding Remarks

Electron holography visualizes magnetic field in a form of distribution of magnetic lines of force. Scanning tunneling microscope visualizes local electronic density of states. Photoemission spectroscopy visualizes energy levels and Fermi surfaces. As indicated in these examples, epoch-making experimental methods and instruments visualize what



Fig. 7. Temperature-dependent electrical resistance of different surface superstructures formed with (sub)monolayer In adsorption on a Si crystal (Refs. 12, 13). The inset in (b) is a RHEED pattern showing Si(111)- $\sqrt{7} \times \sqrt{3}$  -In surface superstructure.

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used to be invisibles just as if they were real objects. This is a real pleasure of science. Tonomura-san's words 'The picture is my life.' shows the heart of science. Four-tip STM, my life work, can be used for visualizing electronic transport in many ways. I will continue to struggle to obtain pictures that visualize hitherto invisible quantities.

I dedicate the pictures shown in this article to Tonomura-san in token of gratitude for his warm encouragement throughout my research life.

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