The Strategy for Acceptance of the Scanning Tunneling Microscope: Observations of the Si(111)7×7 Reconstructed Surface, 1959–1986

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Abstract

Tracing the history of observations of the Si(111)7×7reconstruction from 1959 to 1986, this paper will examine techniques, newly developed for surface science that emerged during that period. Several methods were introduced to probe Si(111)7×7, such as Low Energy Electron Diffraction, Ion Scattering Spectroscopy, and Scanning Tunneling Microscopy. Obtaining a consensus on the structure of the Si(111)7×7 reconstructed surface took more than twenty-five years to achieve. This paper will consider two key factors that were essential in resolving this long-standing problem: the importance of creating representations and the scientific acceptance of the Scanning Tunneling Microscope and its data.

Key words: Surface science, Si(111)7×7 reconstructed surface, Low Energy Electron Diffraction (LEED), Scanning Tunneling Microscopy (STM), credibility

1. Introduction

Introducing observational methods for investigating microscopic substances to understand their structure and properties have provided the driving force behind materials development, electronic engineering, and the medical identification of diseases, to name a few. Investigating microscopic objects has also fascinated researchers for decades, inspiring those who wish to observe the miracles of nature like the behaviors of atoms, or to see an individual atom. The efforts to expand and improve these observational methods are still vigorous and extensive.

Research on crystal structures started in the late eighteenth century. Researchers had categorized crystals by their physical appearance, but in the 1920s, X-ray diffraction methods revealed the atomic basis of crystal structures. By the 1950s, X-ray diffraction analysis was the dominant experimental technique used to determine the molecular structure of a number of crystalline compounds. On the other hand, details of the surface structure had not yet drawn the attention of researchers, since few methods were available other than scales, protractors, and optical microscopy for studying surfaces. Characterization of the

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surface at the molecular or atomic level, such as electronic states, bonds, and the arrangement of the atoms, was not yet possible.

The seeds for atomic characterization of surfaces were planted in the early twentieth century by two events: the light-photon quantum hypothesis proposed by Albert Einstein in 1905, which led to explanation of the photoelectric effect, and the confirmation of the wave character of the electron from diffraction measurements by Clinton J. Davisson and Lester H. Germer in 1927. The photoelectric effect is the fundamental basis behind Photoelectron Spectroscopy (PES) for investigating electric structures of solid surfaces. Using low energy electrons for diffraction was developed to become a new technique, Low Electron Energy Diffraction (LEED), for investigating atomic structures and arrangements of solid surfaces.[1]

Investigating surface structure became a crucial topic in solid-state electronics during the mid-twentieth century. By the 1970s, the necessity of increasing the number of components per chip accelerated the research on silicon surfaces in order to develop reliable microfabrication technology. This in turn led to the invention of instruments for investigating surface structure, and methods for simultaneously measuring the geometric and electronic structure of the surfaces. The research on silicon surfaces had become increasingly important not only to industry as a critical semiconductor material, but also as a subject of academic interest in the 1970s. Determining the surface structures, understanding the crystal growth mechanism, and investigating the properties of silicon surface have been some of the challenging academic topics for surface scientists. Among the many basic scientific questions, the reconstruction of the Si(111)7×7 surface attracted many researchers. The first report of a 7×7 unit cell, composed of forty-nine surface atoms on a Si(111) reconstructed surface, was made in 1959.[2] The structural determination of the Si(111)7×7 reconstruction continued to attract many researchers over the next twenty-five years until the Scanning Tunneling Microscope (STM) confirmed the 7×7 surface structure in 1986.

Today, the STM is one of the most powerful scientific instruments for surface science research. Going beyond its scientific applications, the STM has been attracting attention from historians and philosophers of science, who are concerned with a variety of issues including data imaging, spread and development of the instrument, and the history of nanotechnology.[3] Among the historical works on the STM, Cyrus C.M. Mody and Michael

[1] After the report by Davisson and Germer in 1927, George P. Thomson, Seishi Kikuchi, and others published their research on high energy electron diffraction (the energy of their electron beams were one thousand times higher than those used by Davisson and Germer). This instrumentation opened up a new field of physics in the 1930s. The analysis of surface structures using high energy electron diffraction appeared in the 1970s, as we will discuss in the section 1.2.

[2] R. E. Schlier and H. E. Farnsworth, "Structure and Adsorption Characteristics of Clean Surfaces of Germanium and Silicon," *Journal of Chemical Physics*, 30 (1959): 917–926.

[3] For the diffusion of the instruments, see C. C. M. Mody, "Crafting the Tools of Knowledge: The Invention, Spread, and Commercialization of Probe Microscopy, 1960–2000," Ph.D. dissertation, Cornell University, 2004. For imaging, see J. Hennig, "Changes in the Design of Scanning Tunneling Microscopic Images from 1980 to 1990," in J. Schummer and D. Baird, eds., *Nanotechnology Challenges: Implications for Philosophy, Ethics and Society* (Singapore: World Scientific Publishing, 2006): 143–163; C. C. M. Mody, "Instruments in Training: The Growth of American Probe Microscopy in the 1980s," in D. Kaiser, ed., *Pedagogy and the Practice of Science: Historical and Contemporary Perspectives* (Cambridge, Mass.: MIT Press, 2005): 185–216. For nanotechnology, see C.C.M. Mody, "How Probe Microscopists Became Nanotechnologists," in D. Baird, A. Nordmann, and

Lynch, in describing the short history of Si(111)7×7 and the STM, regard the Si(111)7×7 surface as a "test object" consisting of the material culture of laboratory science.[4]

This paper traces the history of Si(111)7×7 to reveal how researchers resolved the twentyfive year-long problem of determining the Si(111)7×7reconstructed structure. Many instruments have been developed for studying surfaces and have had a historical and dramatic impact in the specialized field of surface science. Highlighting the observations by STM, the author will attempt to demonstrate that the breakthrough in solving the problem of the Si(111)7×7reconstruction was achieved by STM measurements, and that the solution of the structural problem in turn led to the acceptance of the STM in the scientific community.

2. Studies of the Si(111)7×7 Reconstructed Surface before STM

2.1. The Use of LEED to Observe Crystal Surfaces

The first discovery and observation of the 7×7 structure of Si(111) reconstructed surface was accomplished by LEED. LEED is a method for observing the surface structure of crystalline materials using incident electrons in the energy range of 20 to 300 eV. The low energy electrons are scattered by the first few layers of surface atoms. As noted above, the first experiment using low energy electron diffraction was the Davisson–Germer experiment that confirmed the wave character of electrons in 1927. They directed an electron beam against a target consisting of a single crystal of nickel and measured the intensity of scattered electrons with Faraday cup, which was used for collecting and measuring the current of the scattered electrons in various directions from the target.[5]

Although this was a significant experimental achievement and stimulated surface research activity, LEED did not immediately become a popular device for surface analysis because of difficulties in its operation and in the processing of data. It was difficult to generate the ultrahigh vacuum required for the preparation of clean sample surfaces. Moreover, it took about twelve hours to record the diffraction pattern.

2.2. The Discovery of the 7×7 Arrangement of the Si(111) Surface

Although Davisson and Germer did not continue to pursue research using LEED, some researchers continued LEED. Among them, Harry E. Farnsworth and his members developed a LEED similar in principle to the apparatus of Davisson and Germer with a

J. Schummer, eds., *Discovering the Nanoscale* (Amsterdam: IOS Press, 2004): 119–133; A. Hessenbruch, "Nanotechnology and the Negotiation of Novelty," in ibid.: 135–144; D. Baird and A. Shew, "Probing the History of Scanning Tunneling Microscopy," in ibid.: 145–156. For instruments, see G. Granek and G. Hon, "Searching for Asses, Finding a Kingdom: The Story of the Invention of the Scanning Tunneling Microscope (STM)," *Annals of Science*, 65 (2007):101–125; D. Rothbart, *Philosophical Instruments: Minds and Tools at Work* (Champaign, Ill.: University of Illinois Press, 2007), Chapter 6: "Atoms: Easier than Ever Before."

[4] C. C. M. Mody and M. Lynch, "The Test Objects and Other Epistemic Things: A History of Nanoscale Object," *British Journal for the History of Science*, 43 (2010): 423–458.

[5] C. Davisson and L.H. Germer, "The Scattering of Electrons by a Single Crystal of Nickel," *Nature*, 119 (1927): 558–560.

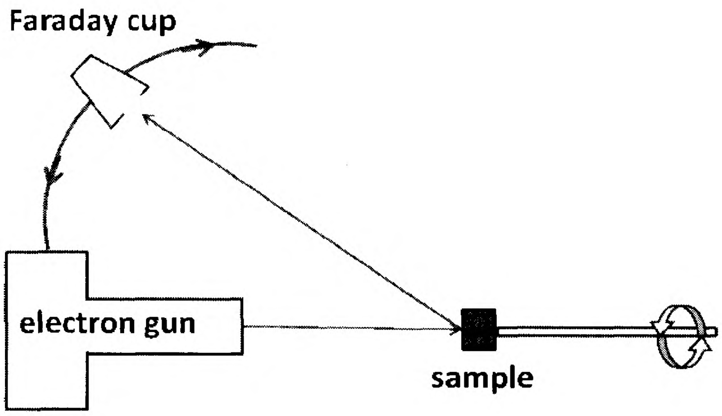

Figure 1. A diagrammatic sketch of the Davisson–Germer experiment (the author's diagram). The crystal was rotated manually around the axis of the incident beam in order to bring each azimuth plane of the crystal into coincidence with the plane containing the Faraday cup.

Faraday cup detector (this type of LEED is often called "Farnsworth type").[6] And they became the first to discover the Si(111)7×7 reconstructed surface in 1959.[7]

Before we go further, let us review the terms associated with surface structure. In the term Si(111)7×7, a set of three numbers or 'Miller indices', (111) in this case, defines a plane of atoms in an atomic crystal. Crystal axes of the unit cell allow a crystal plane to be characterized by three integers in terms of the intercepts made by the plane with these axes, a process known as "indexing." A crystal plane is indexed with three numbers, h, k, and l. The Miller indices of a crystal face are obtained by calculating the intercepts of the plane with the a,b, and c axes, as a fraction of a, b, and c, then by taking the reciprocals of these numbers, and expressing the result in the form of the smallest integer numbers (see Figure 2(a)). Figure 2(b) shows some examples, including the (111) plane. The index numbers provide three-dimensional information on the crystal plane.

An ideal surface indicates that the atoms on the cleavage plane would be the same as the arrangement of atoms in the bulk crystal. However, real surface structures often deviate from ideal behavior in order to minimize surface energy. If we consider the cleaved silicon (111) plane (see Figure 2), the bonds of the surface atoms lose half of their bonding interactions compared with the bulk crystal. These missing bonds are called "dangling bonds." The high-energy state of the exposed surface needs to rearrange, a process called reconstruction, to lower the surface energy. This is accomplished by having the atoms move around on the surface and adopting a new structural arrangement, the "reconstructed surface structure." These surfaces have different properties and structures as compared with bulk or crystalline material. When the silicon crystal is initially cleaved in the vacuum, the periodic arrangement of atoms of the Si(111) surface is 2×1. When heating this cleaved

[6] Farnsworth and his colleagues improved their LEED apparatus by adding ion bombardment to clean and anneal the crystal surface.

[7] Schlier and Farnsworth, "Structure and Adsorption Characteristics," op, cit. (note 2).

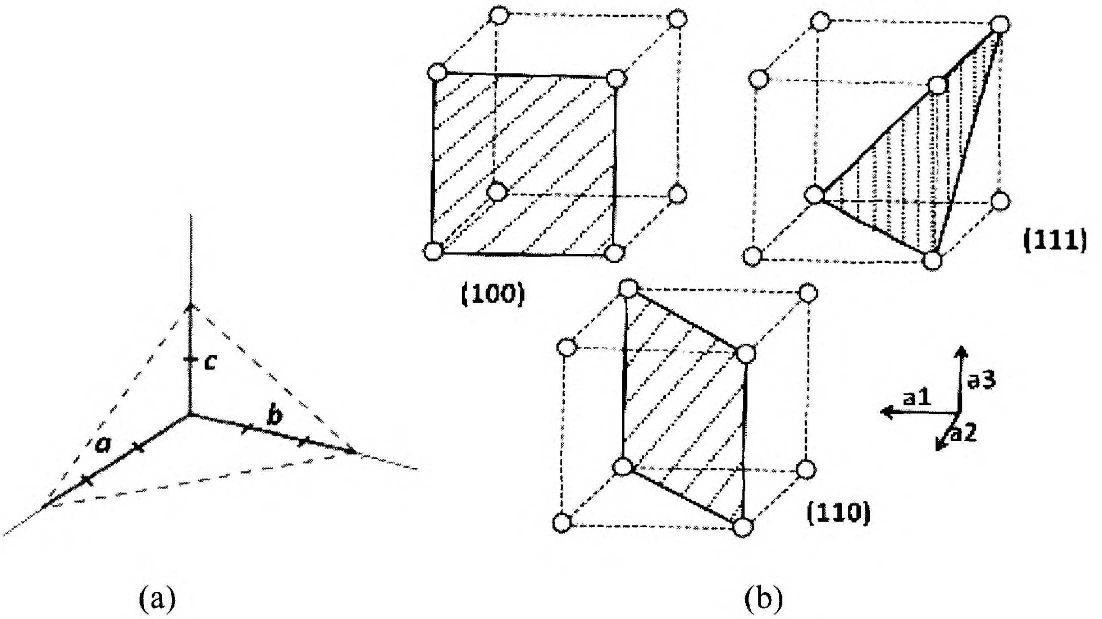

Figure 2. (a) An example of the derivation of Miller indices of a plane. This is an example of (223). (b) Low-index ideal surfaces of a cubic crystal. Vertical and horizontal markings, respectively, indicate the second and third atom layers. These circles on the edge of cubes indicate atoms of the crystals (the author's diagrams).

Si(111) surface to a temperature higher than 900°C in ultrahigh vacuum, the arrangement of the atoms turns to 7×7 (Figure 3). So, in terms of the Si(111)7×7 surface, the 7×7 represents the periodic pattern of atoms on the surface that formed from the rearrangement of the Si(111) surface. Surface reconstruction has attracted the interest of surface scientists who were eager to determine the nature of the reconstruction and examine how the properties of the surface, electronic structure, and reactivity, have changed.

Though the discovery of 7×7 structure of Si(111) reconstructed surface was a remarkable finding, most researchers were skeptical about the initial report of the reconstructed 7×7 structure of silicon by Farnsworth group, because they suspected that the change in the surface was due to contamination.[8] Subsequent to this first report, a 5×5 structure of the Si(111) reconstructed surface was reported, but this finding was later shown to be due to surface contamination and thus fueled the controversy.[9] Finally, in other experiments that followed about ten years later, the 7×7 structure was confirmed and shown not to be due to trace contamination.[10] Yet just the discovery of 7×7 arrangement did not mean that

[8] D. Haneman, "Surfaces of Silicon," *Reports on Progress in Physics*, 50 (1987): 1045–1086; S. Ino, "*Si* no (111) Hyomen no 7×7 Kozo no Mokei (Structure Models for the Si(111)7×7 Surface Structure) [in Japanese]," *Journal of the Crystallographic Society of Japan*, 23 (1981): 197–216.

[9] J. J. Lander, G. W. Gobeli, and J. Morrison, "Structural Properties of Cleaved Silicon and Germanium Surfaces," *Journal of Applied Physics*, 34 (1963): 2298–2306.

[10] For example, observation with LEED and Auger electron spectroscopy showed no trace of contamination. E. Bauer, "On the Nature of Annealed Semiconductor Surfaces," *Physical Letters A*, 26 (1968): 530–531.

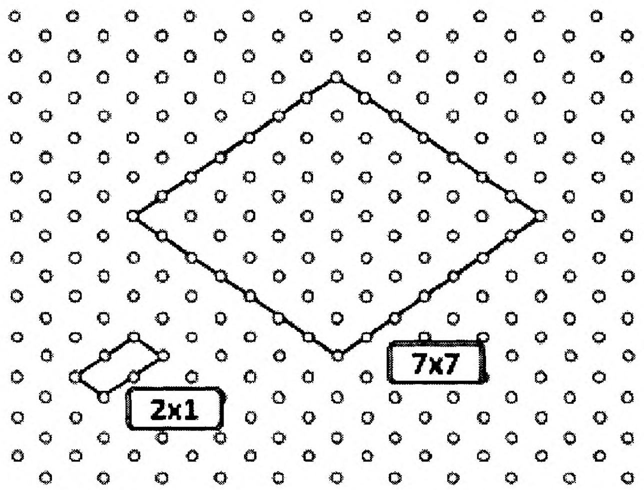

Figure 3. A cleaved (111) silicon surface creates a 2×1 periodic arrangement (left). By annealing the surface, atoms on the surface reconstruct into a 7×7 (right) arrangement. The circles represent atoms on the surface. This surface shows the highest layer of bulk, and not the top layer of the surface. The 7×7 notation means that the length of the surface structure is sevenfold of the length for the unit vector of a unit cell in the ideal surface (the author's diagram).

researchers had all the information of the arrangement of each atom on the Si(111)7×7 reconstructed surface.

2.3. Studies of the Si(111)7×7 Reconstructed Structure before the 1980s

About thirty years after his initial studies, Germer returned to using LEED, where he and his coworkers successfully introduced an innovation in capturing LEED patterns by using a fluorescent screen with accelerated diffracted electrons in 1959.[11] This diffraction pattern reflected the spacing of the atoms in reciprocal space and it showed the periodic nature of the surface structure. Furthermore, this approach dramatically reduced the data-acquisition time from twelve hours to typically thirty seconds using Polaroid film. This type of LEED with the fluorescent screen was subsequently improved.[12] The LEED apparatus with this screen type was often called "Germer type" (Figure 4).

This report of the direct observation of the surface structure came as a great shock to Farnsworth and his members.[13] Though Germer admitted the disadvantage of Germer

[11] E. J. Scheibner, L. H. Germer, and C. D. Hartman, "Apparatus for Direct Observation of Low-Energy Electron Diffraction Patterns," *Review of Scientific Instruments*, 31 (1960): 112–114. The original idea of using fluorescent screen was reported by Wilhelm Ehrenberg in 1934. W. Ehrenberg, "A New Method of Investigation the Diffraction of Slow Electrons by Crystals," *Philosophical Magazine*, 18 (1934): 878–901.

[12] The following year, they further improved the apparatus by making it smaller, eliminating the unnecessary components that could become corroded, reducing the background noise, and upgrading the electron gun. L. H. Germer and C. D. Hartman, "Improved Low Energy Electron Diffraction Apparatus," *Review of Scientific Instruments*, 31 (1960): 784–785. And after the improvement of the spherical type of fluorescent screen and grid by J. J. Lander and his colleagues this type became a standard system for LEED for surface investigation.

[13] Toshiro Yamashina worked at the Farnsworth group around 1960 and he saw that they were suffering from

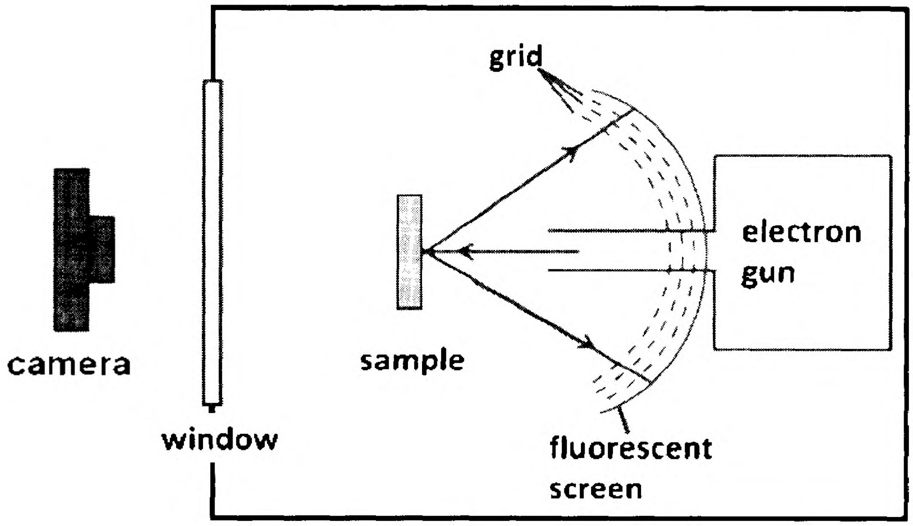

Figure 4. A diagrammatic sketch of the typical LEED with fluorescent screen (the author's diagram). The diffraction patterns are photographed by camera through the window.

type because of its inaccuracies with respect to relative intensity measurements, necessary geometrical corrections and the limited range of sampling angles, Farnsworth group had to admit their time-consuming measurements with counting each electron was also a disadvantage. But the Farnsworth group still chose to pursue their quantitative measurements. Farnsworth and his members improved their apparatus to reduce the time required for collecting the data.[14]

The Germer-type LEED, however was the apparatus that rapidly spread among surface scientists, as measurement of relative intensities became possible using a spot meter. The Germer type was used in commercial LEEDs, which the LEED more accessible to many researchers in mid 1960s.[15] The birth of LEED helped develop surface science and the use of reciprocal images of LEED patterns became a major window for surface observation.

Problems and challenges still remained. After obtaining a diffraction pattern image, researchers still needed to measure the relative intensities of each spot, and then compare these intensities with calculated intensities of each spot from several possible models.[16] These models were based on various periodic crystal structures and other conditions.

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shock. when the report by Germer group in 1960 T. Yamashina and S. Fukuda, *Hyomen Bunseki no Kiso to Oyo* (Introduction and Application for Surface Analysis) [in Japanese] (Tokyo: Tokyo Daigaku Shuppankai, 1991): p. i

[14] R. L. Park and H. E. Farnsworth, "The Structures of Clean Nickel Crustal Surfaces," *Surface Science*, 2 (1964): 527–533.

[15] E. G. McRae, "Low Energy Electron Diffraction at Bell Labs in the 1960s," in P. Goodman, ed., *Fifty Years of Electron Diffraction* (Dordrecht: D. Reidel, 1981): 197–207.

[16] Finally, in the 1970s LEED users benefited from a book, *Low Energy Electron Diffraction*. (J. B. Pendry, *Low Energy Electron Diffraction* (London: Academic Press, 1974). It included a computer program for calculating multiple scattering events, which enabled LEED users to analyze surface structure properly.

LEED users had to try to find the most probable model that agreed with the experimental data for their crystal. Despite these efforts, LEED had not provided a definitive solution for any silicon surface structures.

In the case of the 7×7, the sheer size of the unit cell posed additional complications. It was too large to investigate (as depicted in Figure 3) easily, and the prediction of a specific structure was challenging because of the number of atoms in the first four layers (ninety-eight atoms were in the top two layers). Since the LEED experiments did not provide definitive results, it required modeling of structures and theoretical analysis, where the interpretation was subject to discussion and debate.

More instruments for surface observation were developed in the 1960s, due to innovations in generating ultra-high vacuum.[17] Among the second generation instruments, Reflection High Energy Electron Diffraction (RHEED) appeared. RHEED, like LEED, was an experimental technique used to observe surface structures as a diffraction pattern (Figure 5(b)). While RHEED had some advantages over LEED, the RHEED image represented a distorted image of the reciprocal lattice and it was difficult to analyze the pattern. One of the efforts to analyze the RHEED pattern was to modify RHEED images to be like LEED images, where the periodical intensity in the patterns could be easier to identify, allowing them to be analyzed in a manner similar to LEED analysis.[18] This meant that the RHEED pattern needed to be similar to the LEED pattern since LEED pattern analysis was a standard technique for understanding surface structures.

Among other instruments, Reflection Electron Microscopy, which is a specific version of Ultrahigh Vacuum Electron Microscopy applied in reflection mode, and helium atom diffraction scattering were also used to observe the 7×7 surface in the mid-1970s.[19] A wide variety of methods and instruments for surface observation developed and combined with each other at the time.

The observations, based on these experimental methods, and some works combined with theoretical calculations, generated a number of models of the Si(111) 7×7 reconstructed structure.[20] Most notably, the vacancy model (Lander–Morrison model), which had thirteen vacancies in the first layer of silicon atoms, was the most well-known model through the 1970s.[21] Other developed models included the "milk-stool" model, which con-

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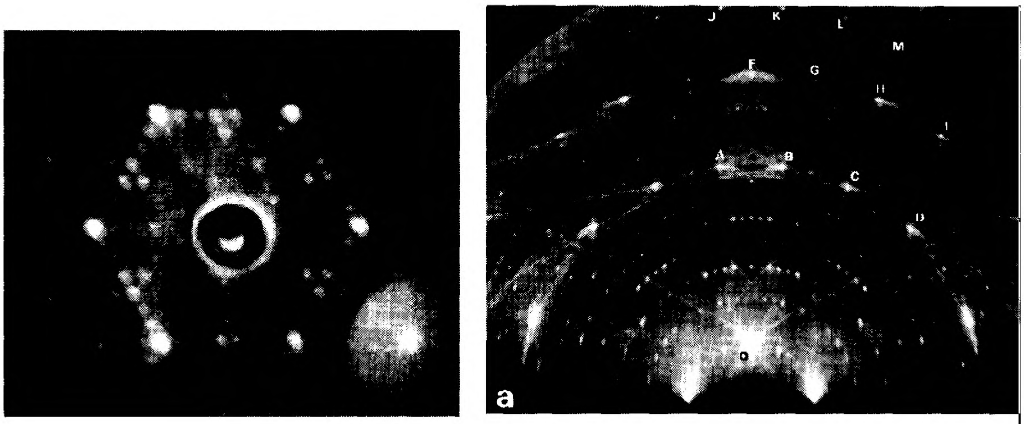
[17] One of the innovations for generating ultra high vacuum was the invention of a vacuum flange, which connects vacuum chambers.

[18] S. Ino, "Some New Techniques in Reflection High Energy Electron Diffraction (RHEED) Application to Surface Structure Studies," *Japanese Journal of Applied Physics*, 16 (1977): 891–908; S. Ino, "A New Structure Model for the Si(111) 7×7 Surface Structure," *Japanese Journal of Applied Physics*, 19 (1980): L61–L64.

[19] For helium atom diffraction scattering, M.J. Cardillo, "Nature of the Si(111)7×7 Reconstruction," *Physical Review B*, 23 (1981): 4278–4282. This helium atom diffraction scattering experiment derived a model of triangular etched regions each occupying half of the 7×7 unit cell. For REM, N. Osakabe, Y. Tanishiro, K. Yagi, and G. Honjo, "Reflection Electron Microscopy of Clean and Gold Deposited (111) Silicon Surfaces," *Surface Science*, 97 (1980): 393–408.

[20] Computer calculations were also indispensable in the analysis of surface structures. The calculations were based on several models. However, detailed computational studies on the electronic structure of the 7×7 surfaces for comparison with experiments were too difficult to carry out at the time because of the lack of computational power to perform such large calculations.

[21] J. J. Lander and J. Morrison, "Structures of Clean Surfaces of Germanium and Silicon, I," *Journal of Applied Physics*, 34 (1963): 1403–1410.



(a) (b)

Figure 5. (a) LEED pattern, (b) RHEED pattern. Reprinted with permission from S. Ino, "An Investigation of the Si(111)7×7 Surface Structure by RHEED," *Japanese Journal of Applied Physics*, 19 (1980): 1277–1290. Copyright (1980) by The Japan Society of Applied Physics.

sisted of a three-membered ring of atoms with three dangling bonds extending from the top layer of the surface atoms,[22] an adatom model (Harrison model) that involved the addition of thirteen atoms above the positions of the same atoms that would be vacant in the vacancy model,[23] a distortion model consisting of a rippling of atom layers,[24] and a hexagon model consisting of one lowered atom per unit cell surrounded by three hexagonal-shaped rings of raised atoms whose displacements diminished away from the center.[25]

Surface scientists who studied Si(111)7×7 usually presented their own 7×7 models, which were based on their specific experiments for comparison with other models. None of the models were consistent with all of the experimental data, but some of the observations by these instruments narrowed the realm of speculation.[26] In the early 1980s, the determination of the structure of the Si(111)7×7 reconstruction was a challenging problem, as if it were "a problem-solving Olympic Game."[27] Numerous review articles on silicon surface studies published prior to the invention of the STM were a clear indication of the importance placed on understanding the surface of silicon.[28]

[22] L. C. Snyder, Z. Wasserman, and J. W. Moskowitz, "Milk-stool Model for Si(111) Surface Reconstruction," *Journal of Vacuum Science and Technology*, 16 (1979): 1266–1269.

[23] Walter A. Harrison, "Surface Reconstruction on Semiconductors," *Surface Science*, 55 (1976): 1–19.

[24] J. D. Levine, P. Mark, and S. H. McFarlane , "Si(111)7×7Surface Structure," *Journal of Vacuum Science and Technology*, 14 (1977): 878–882. This model involved a depression of atoms in one half of the 7×7 unit cell and elevation of the atoms in the other half, called a "ripple" configuration.

[25] D. J. Miller and D. Haneman, "LEED Analysis and Energy Minimization Calculations for Si(111)7×7 Surface Structures, " *Journal of Vacuum Science and Technology*, 16 (1979): 1270–1285.

[26] D. Haneman, "Semiconductor Surfaces," *Advances in Physics*, 31 (1982): 165–194.

[27] Ino, "Structure Models," op.cit.(note 8).

[28] For example, D. E. Eastman, "Geometrical and Electric Structure of Si(001) and Si(111) Surfaces: A Status Report," *Journal of Vacuum Science and Technology*, 17 (1980): 492–500; Haneman, "Semiconductor Surfaces," op.cit.(note 26); Ino, "Structure Models," op.cit.(note 8); G. A. Somorjai, "Surface Science: An Old

3. The Advent of Scanning Tunneling Microscopy

3.1. The Invention of the Scanning Tunneling Microscope

The STM first appeared in the early 1980s.[29] Gerd Binnig and Heinrich Rohrer at IBM Zurich Research Laboratory, who were inventors of the STM, were about to initiate a new research study at the end of the 1970s. At that time, some IBM researchers were developing a new computer element, the Josephson element, which was expected to be capable of much higher performance than silicon elements, as a key component of a new IBM computer.[30] Unfortunately, defect holes on the surface of the Josephson element led to failures, which annoyed the element developers. Rohrer and Binnig were interested in spectroscopically mapping local surface areas of less than one hundred angstroms in diameter in order to investigate the growth and electronic properties of thin insulating films critical to the Josephson elements. Since they realized that no instrument existed for their research, they had to develop a new instrument, the STM.

The STM was a type of microscopy that used a tip to probe the sample surface. The metal tip of the STM is used to measure the topography and distribution of electronic states at the surface of the sample. When the distance between the tip and the surface of the sample is about one nanometer, current flows between the tip and the sample. It is called a tunneling current, because while there is a potential barrier impeding the flow of current, the electrons could pass through the barrier through the "tunneling effect" of quantum mechanics. As the tip is translated across the surface, the level of the tunneling current is kept constant by adjusting the vertical position of the tip. A vertical profile of the surface is produced as the tip is rastered across the surface. The resulting STM image is processed by computer and appears as a topographical map at atomic resolution (Figure 6).

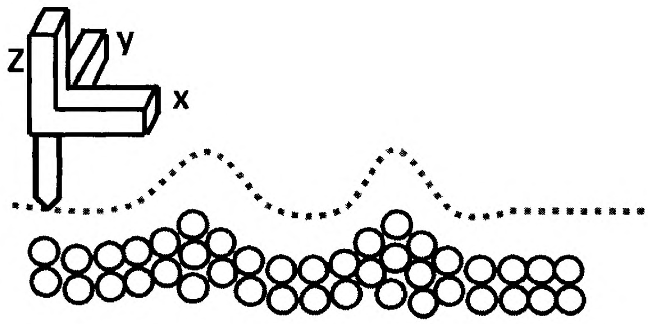


Figure 6. A diagrammatic sketch of the STM (the author's diagram).

Field Rejuvenated, demands Attention and People," *Science*, 201 (1978): 489–497.

[29] G. Binnig, H. Rohrer, Ch. Gerber, and E. Weibel, "Surface studies by Scanning Tunneling Microscopy," *Physical Review Letters*, 49 (1982): 57–61. Granek and Hon trace the process of the invention of STM at IBM in detail (Granek and Hon, "Searching for Asses, Finding a Kingdom," op.cit.(note 3)).

[30] A research project on Josephson computer technology at IBM was initiated in 1964. IBM cut the Josephson computer project in 1983.

Prior to the development of the STM, there were other microscopes that employed tips: the Field Emission Microscope, Field Ion Microscope, and Topografiner. Erwin Mueller invented the Field Emission Microscope (FEM) in 1936 and the Field Ion Microscope (FIM) in 1951. While the Field Emission Microscope did not possess atomic resolution, the Field Ion Microscope did. However, since the shape of the sample was a whisker, it was not suitable for observing broad and flat samples, and the FIM could not display every single atom on the surface of the sample. Russell D. Young, a student of Mueller, and his colleagues, invented the Topografiner in 1972. It had a tip for probing the surface of the sample, but like the FIM, it also did not possess atomic resolution.[31]

3.2. Early Reactions to the STM

IBM filed a patent application on the STM in 1979.[32] Binnig and his colleagues reported their invention at an international conference in 1981, but this did not intrigue many researchers with the instrument.[33] The inventors of STM had selected specific samples for their initial observations: Au and $CaIrSn_4$ surfaces, which were available at the IBM laboratory.[34] They succeeded in imaging atomic steps on these surfaces.[35] These results demonstrated that the STM possessed atomic resolution and were the first topographic images recorded by the STM. They represented each atom as a dot on the lateral side of the topographic image, thus visualizing the Au atomic arrangement.

The observation of atomic steps on a surface was not sufficient to trigger a fascination for the STM in the broader scientific community. In fact, many researchers were skeptical of the observations. Some thought that achieving atomic resolution would require an impossibility, a probe with a single atom on the tip. Others could not understand how the STM could image an atom with the size of about one angstrom using a probe that had a one-hundred angstrom radius of curvature. Thus, most scientists regarded it impossible to observe and image an atomic step with such a device. Since the inventors did not describe their experiment in detail and did not discuss the influence of the tip on the sample in the report, such skepticism was not unexpected.

Some theoreticians were less skeptical, and tried to prove that the STM had the capability of atomic resolution. Jerry Tersoff and Donald R. Hamann proposed a spherical model of the STM tip and mathematically demonstrated that their calculations were consistent with the experimental results for Au as obtained with the STM.[36] Theoretical research

[31] R. D. Young, J. Ward, and F. Scire, "The Topografiner: An Instrument for Measuring Surface Microtopography," *The Review of Scientific Instruments*, 43 (1972): 999–1011. The STM inventors leaned the Topografiner two year after the STM invention. G. Binnig and H. Rohrer, "Scanning Tunneling Microscopy: From Birth to Adolescence [Nobel Lecture]," *Review of Modern Physics*, 59 (1987): 615–625. This lecture was delivered on 8 December 1986, on the occasion of the presentation of the 1986 Nobel Prize in Physics.

[32] G. Binnig and H. Rohrer, "Scanning Apparatus for Surface Analysis Using Vacuum-tunnel Effect at Cryogenic Temperatures," initially filed on 20 September 1979 in Switzerland and published as CH643397. This application was also filed as the European patent EP0027517 and the US patent US4343993 using the Paris Convention Priority.

[33] Binnig and Rohrer, "Scanning Tunneling Microscopy," op.cit.(note 31)

[34] Ibid.

[35] G. Binnig, H. Rohrer, Ch. Gerber, and E. Weibel, "Surface Studies by Scanning Tunneling Microscopy," *Physical Review Letters*, 49 (1982): 57–61.

[36] J. Tersoff and D. R. Hamann, "Theory and Application for the Scanning Tunneling Microscope," *Physical*

on STM still continued, with theoreticians and experimentalists undertaking collaborative and corroborative research. The STM created not only a realm of new surface science but also of new theoretical research.

These theoretical efforts were meaningful, but not sufficient to overcome the skepticism on STM in the surface science community. While the observations of Au(110) and $CaIr_4$ (111) surfaces drew some attention from surface scientists, this work did not fall into the "hot topics" category of surface science research at the time.

Faced with the less than enthusiastic reception of the STM, the inventors of the STM strategized on a way to increase the appeal of the STM to more researchers. As Binnig and Rohrer stated, "instead of wasting further time on uninteresting step lines, we preferred to attack surface reconstructions with known periodicities and with a reasonable chance of learning and contributing something new."[37] They chose the Si(111)7×7 reconstructed surface as an ideal target with which to convince the surface science community. Regarding scientific activities, Bruno Latour points out that " ... one of the main problems to solve is to interest someone enough to be read at all; compared to this problem, that of being believed is, so to speak, a minor task."[38] Choosing the Si(111)7×7 reconstruction as their focus incorporated exactly the sort of topic that was of sufficient interest at the time to be read. Nevertheless, the inventors were not satisfied with merely having their paper read. They felt challenged, and their aim was to be believed and to have their newly invented instrument accepted.

3.3. The Si(111)7×7 reconstruction in real space and two STM images

The STM inventors successfully observed the surface of Si(111)7×7 in real space by STM in 1982.[39] They prepared two types of STM image representations for the paper. One of the images was created from the original recordings on the XY-recorder, which were acquired with the STM. They transferred the paper chart recordings onto an acrylic plate and cut along the line, tracing on the chart recordings (Figure 7). Then they pasted the cuttings together to make a "relief. [40]" They pictured the relief as a figure for the paper, as shown in Figure 8(a). The inventors visualized the observation of a surface structure of the silicon reconstruction with this non-digital approach. There are two rhombic units in the relief. And four large holes at the corners and twelve raised sites existed in each rhombic. The rhombic is the unit cell of this surface structure; twelve raised sites, shown as adatoms (adsorbed atoms), are the highest atoms on the surface and the four large holes are "corner holes." The existence of these corner holes and twelve adatoms, confirmed by STM, was decisive information related to the Si(111)7×7 surface, because the other methods which

Review Letters, 50 (1983): 1998–2001. M. Tsukada and N. Shima, "Theory of Electronic Processes of Scanning Tunneling Microscopy," *Journal of Physical Society of Japan*, 56 (1987): 2875–2885. Masaru Tsukada and Nobuaki Shima further developed the idea of the Tersoff–Hamann model with an actual tip in 1987.

[37] Binnig and Rohrer, "Scanning Tunneling Microscopy," op.cit. (note 31), p. 619.

[38] B. Latour, *Science in Action: How to Follow Scientists and Engineers through Society* (Cambridge, Mass.: Harvard University Press, 1987), p. 41.

[39] G. Binnig, H. Rohrer, Ch. Gerber, and E. Weibel, "7×7 Reconstruction on Si(111) Resolved in Real Space," *Physical Review Letters*, 50 (1983): 120–123.

[40] Binnig and Rohrer, "Scanning Tunneling Microscopy," op.cit. (note 31).

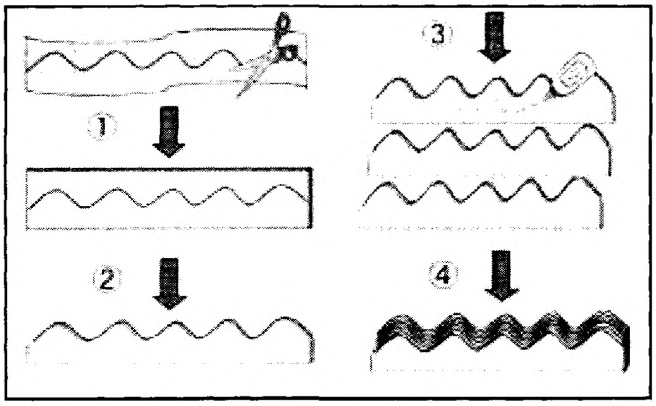


Figure 7. The STM inventors' procedure of manually crafting the image relief (the author's diagram).

produced reciprocal space images had been unable to observe the concavity and convexity of the top layer of the surface.

The previous methods for surface structure, like LEED and RHEED, provided information on surface structures from diffraction, and as such, were based on the periodicity of the surface over a wide area of the crystal. On the other hand, the STM image provided a topographic and localized view of the surface. The STM relief image for the silicon surface identified the arrangement of individual atoms.[41]

Nevertheless, the STM image of surface structure in real space are intuitive representations because of its three dimension representation. It was significant that the raw data from the STM could directly provide an image of the surface without laborious calculations or well-trained eyes. Consequently, most surface scientists, who used a variety of other types of instruments, were quickly able to recognize and understand the STM images without specialized training.

Binnig and his coworkers prepared this relief image to support the measurements made with the STM and to enhance its "credibility."[42] They went beyond normal reporting procedures by providing the raw data itself: the charts from the XY-recorder. By cutting, pasting, and taking pictures in an effective manner, they sought to dispel the skepticism of other surface scientists towards STM. After preparing this relief, they never undertook such a time- and labor-consuming effort again. Instead, they developed other methods to visualize their data to support the observations from their STM measurements.

Just what was the "credibility" that Binnig and Rohrer sought to achieve, as mentioned in their Nobel lecture? What is "credibility" for an instrument or an experiment? As I mentioned in the previous section, many researchers were suspicious of the results from the STM because of its unimaginable (at least for some) methodology. The STM inventors

[41] Technically, the researchers using STM still needed to be careful with the interpretation of the STM images, since these images represented the electron-density distribution on the surface, not the intrinsic surface of the atoms.

[42] Ibid., p. 620.

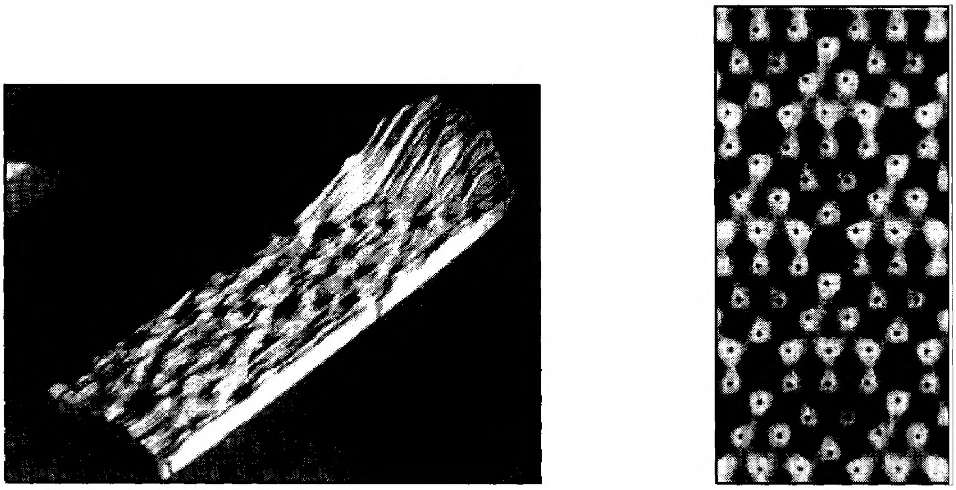


Figure 8. (a) A relief of the STM image, (b) A processed STM image. Reprinted figures with permission from G. Binnig, H. Rohrer, Ch. Gerber, and E. Weibel, "7×7 Reconstruction on Si(111) Resolved in Real Space," *Physical Review Letters*, 50 (1983): 120–123. Copyright (1983) by the American Physical Society.

made efforts to persuade their suspicious colleagues of the veracity of the STM by showing the raw chart data from the XY-recorder. The computer-processed image was regarded as enhanced, modified or idealized one. Since the IBM was a computer manufacturer, criticism of the processed STM image might not be a surprise (in fact, Binnig and his colleagues heard such rumor). The inventors might want to avoid such criticism as well. Moreover, in this case, the relief image visualized the process of STM data processing. This visualization played a crucial role for the credibility of the STM.

The inventors of STM also prepared a second figure in the Si(111)7×7 paper for "analysis and discussion" (Figure 8(b)).[43] This STM image was a computer-processed image and showed the arrangement of atoms on the top layer. The manually constructed relief and this STM image were based on the same original measurements, but the methods of representing the data were different. In Figure 8(b), contrast was applied to show vertical positioning displaying higher positions on the surface by brighter areas on the image. Consequently, this type of representation became the standard mode for displaying STM images, which proved to be suitable for showing the top surface layers for analysis and discussion.

The results of this STM experiment invalidated many of the models of the Si(111) 7×7 surface described in the previous section. However, the results also clearly supported a group of adatom models. For the first time, convincing and definitive results on the silicon surface established the STM as an effective instrument for surface characterization

[43] Loc.cit.

in surface science. From their results, Binnig and his colleagues proposed a modified adatom model in their paper. The structure of Si(111)7×7, however, still remained to be an unsolved problem, and the 7×7 studies continued.

This STM accomplishment attracted several researchers' interests for mainly two reasons: First was that STM revealed the fundamental contour of the surface structure of Si(111)7×7 successfully. Second was that the newly developed instrument enabled one to image or "see" individual atoms without physical contact, to investigate the surface electronic structure, and to possess the capablity of surface fabrication at atomic level.[44]

After the observation of the 7×7 surface by STM, papers on the Si(111)7×7 reconstructed surface began to cite the STM results. Citing the experiment or instrumentation in scientific papersonly indicates that the instrument is still under investigation by the scientific community. Its acceptance only comes after it can be replicated elsewhere and the results independently confirmed by others, as I describe in the following section. After this has occurred, one often sees such instrumentation turn into a black-box, an accepted device that can be used by the average researcher with the results accepted as reputable scientific research.

4. Post-STM Relief of Si(111)7×7 and the Settlement of Controversy

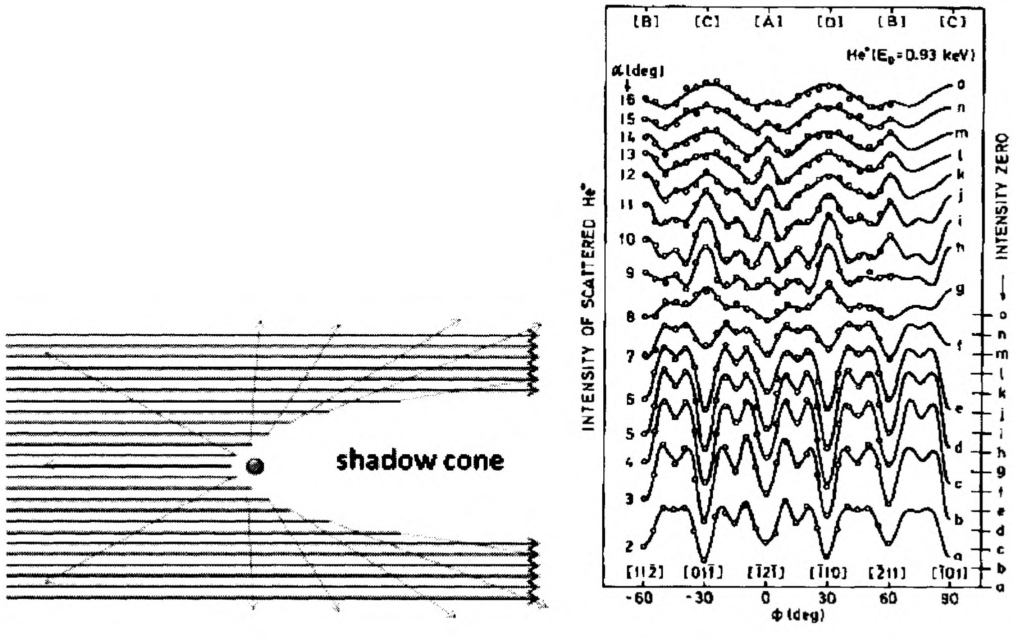
4.1. Another Real-Space Observation: Ion Scattering

The STM accelerated studies of the Si(111)7×7 surface and spawned a variety of proposed models. The STM real-space image provided a highly suggestive patterned and indented surface. To determine the structure of the 7×7 surface, further research was still needed, because the lateral resolution of STM was not sufficient to resolve adjacent atoms except in special circumstances, and the structure underneath the surface could not be imaged.

Several new methods for studying surfaces appeared in the early 1980s. Among them, we will draw attention two methods, impact-collision ion scattering spectroscopy (ICISS) and Transmission Electron Diffraction (TED). The former provided geometrical information in real space and the latter took a crucial role in resolving the controversy of the surface structure of Si(111)7×7. Before we proceed with the history of Si(111)7×7 by STM, I would like to compare the two real space observations by STM and ICISS.

In the early 1980s, ICISS, which was a specialized technique of ion scattering spectroscopy, also produced a real-space observation of the Si(111)7×7. When an ion beam at several hundred eV strikes a target atom, the interaction between the ion and the atom generates a region about the radius of the atom where the ion cannot intrude, called the "shadow cone," (Figure 9(a)). When there is an adjacent atom within the shadow cone, there is no ion-scattering signal by that atom. However, when there is an adjacent atom at the edge of the shadow cone, the intensity of ion scattering dramatically increases. Utilizing these effects, ICISS enables one to determine the relative position of atoms on the

[44] H. Iwasaki and S. Nakamura, "Sosa Tonneru (Koka) Kenbikyo niyoru Hyomen Genshi Kozo no Jitsukukan Kansatsu-Si(111)7×7 Chokozo Hyomen (Surface Atomic Structure in Real Space by Scanning Tunneling Microscopy: Si(111)7×7 Surface Superstructure) [in Japanese]," *Kotai Butsuri*, 18 (1983): 351–356.



(a) (b)

Figure 9. (a) The shadow cone (the author's diagram). (b) Intensity of He^{+} ions scattered from the Si(111)7×7 reconstructed surface. The symbols of [A][B][C][D] indicate the directions of the surface. α and ϕ are the polar and azimuthal angles of incidence direction of primary helium ions. Reprinted figures with permission from M. Aono, R. Souda, C. Oshima, and Y. Ishizawa, "Structure Analysis of the Si(111)7×7 Surface by Low-Energy Ion Scattering," *Physical Review Letters*, 51 (1983): 801–804. Copyright (1983) by the American Physical Society.

surface.[45] The ICISS analysis gave real-space information on the location of atoms on the 7×7 surface, and was consistent with STM images (Figure 9(b)).

While the ICISS experiment received attention from the surface science community as a successful technique, it did not have the same degree of impact as that from the STM, even though both ICISS and STM are real-space experiments. Why were these two experiments received so differently by the surface science community? I point to two notable differences between the ICISS and STM experiments. First, the ICISS experiment, had direct continuity with the existing method, ion scattering spectroscopy. The STM had no predecessor. This gap between the unexpected observation with a newly invented instrument and the initial skepticism of most researchers could make the impact of the STM stronger, provided that the STM inventors could overcome the skepticism from within the

[45] M. Aono, C. Oshima, S. Zaima, S. Otani, and Y. Ishizawa, "Quantitative Surface Atomic Geometry and Two-Dimensional Surface Electron Distribution Analysis by a New Technique in Low-Energy Ion Scattering," *Japanese Journal of Applied Physics*, 20 (1981): L829–832. M. Aono, R. Souda, C. Oshima, and Y. Ishizawa, "Structure Analysis of the Si(111)7×7 Surface by Low-Energy Ion Scattering," *Physical Review Letters*, 51 (1983): 801–804. The researchers proposed a pyramidal model, which has three lower adatoms at the on-top sites of three adjacent underlying first layer atoms, and the one upper adatom is located at the hollow site of the three lower adatoms.

surface science community.

The second difference was the method of data representation. The ICISS graphically represented its scattering distribution and intensity. On the other hand, the STM represented the electron cloud around the atom with the tunneling current between the tip and the sample, as it were, revealing the bare skin of the bulk. The impact of this representation is worthy of attention. The STM image showed the arrangement of the atoms directly to any viewer, while the ICISS distribution could be intuitively read only by experienced ISS users. It can be argued that these differences between the two real-space observations were responsible for the impact created by the advent of the STM.

4.2. Observations of the Si(111) 7×7 Reconstructed Surface by TED

There were several sources of diffraction data of the Si(111)7×7 surface, including LEED, RHEED, X-ray photoelectron diffraction, and TED. Of them, the application of TED provided significant insight to the problem of the Si(111)7×7 surface structure. In the early 1980s, though several researchers developed TED, the number was limited because TED also required the use of an ultrahigh vacuum high-resolution electron microscope, UHV-HREM (Figure 10 shows the TED pattern of the 7×7 surface).

Two probable models for the 7×7 surface came from TED research. One was a triangle-dimer model, which had the unit cell containing nine dimers (paired atoms) or pairs of dimers bordering a triangular subunit of the unit cell. This model gave good, though not perfect, agreement with TED patterns. The other was a dimer adatom stacking-fault model or DAS model, which consisted of twelve adatoms in the top layer, six rest atoms, which were not connected to adatoms, in the second layer, nine dimers in the third layer, one corner hole, and one pair of stacking-faulted/ unfaulted triangle regions in the unit cell (Figure 11).[46] Kunio Takayanagi and his coworkers extended their TED analysis with the DAS model to include data from LEED, RHEED, ICISS, and STM experiments. All of these results were consistent with the DAS model.

Additional experiments, such as high-energy helium ion scattering and Ultraviolet Photoemission Spectroscopy, were performed to observe the Si(111)7×7 surface, which in turn generated even more models.[47] These new models were investigated by testing them against existing experiments and with computational studies. At the time, all of these new

[46] K. Takayanagi, Y. Tanishiro, M. Takahashi, and S. Takahashi, "Structural Analysis of Si(111)7×7 by UHV-Transmission Electron Diffraction and Microscopy," *Journal of Vacuum Science and Technology A*, 3 (1985): 1502–1506; K. Takayanagi, Y. Tanishiro, S. Takahashi, and M. Takahashi, "Structure Analysis of Si(111)7×7 Reconstruction Surface by Transmission Electron Diffraction," *Surface Science*, 164 (1985): 367–392. Takayanagi and his collegues had been working with the Transmission Electron Microscope, captured a dark-field transmission electron microscopic image of the Si(111)7×7 surface, but it did not show the arrangement of atoms on the surface.

[47] For high-energy helium ion scattering, see R. J. Culbertson, L. C. Feldman, P. J. Silverman, and R. Haight, "Hydrogen Adsorption on Si(111)-(7×7)," *Journal of Vacuum Science and Technology*, 20 (1982): 868–871. For ultraviolet photoemission spectroscopy, see J. E. Demuth and A. J. Schell-Sorokin, "Rare Gas Titration Studies of Si(111) Surfaces," *Journal of Vacuum Science and Technology A*, 2 (1984): 808–811. For computation, see T. Yamaguchi, "Si(111) 7×7 Reconstruction: Strain in the Adatom Model," *Physical Review B*, 30 (1984): 1992–2000; T. Yamaguchi, "Restricted Role of Experiments in Real Space in Determination of the Si(111)7×7 Reconstructed Structure," *Physical Review B*, 32 (1985): 2356–2370.

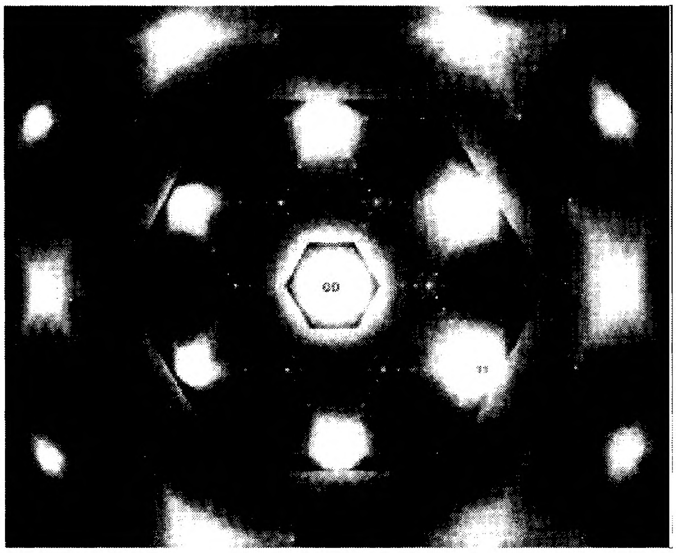


Figure 10. TED pattern of the Si(111)7×7 surface. Reprinted from K. Takayanagi, Y. Tanishiro, S. Takahashi, and M. Takahashi, "Structure Analysis of Si(111)7×7 Reconstruction Surface by Transmission Electron Diffraction," *Surface Science*, 164 (1985): 367–392. Copyright (1985), with permission from Elsevier.

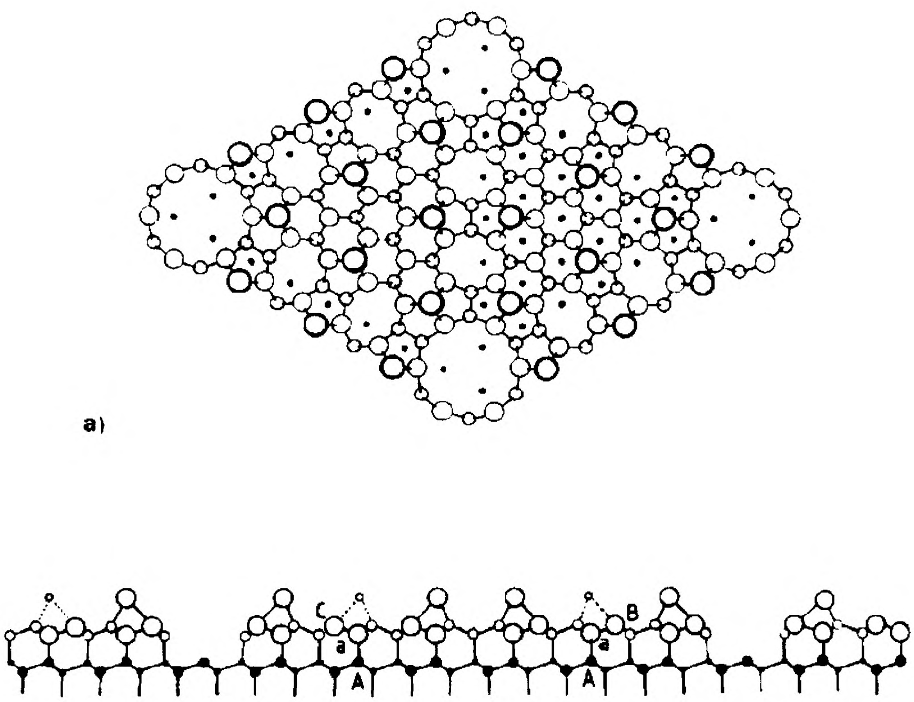
models were much more complex than the models developed prior to the publication of the constructed STM relief. Nevertheless, the controversy over the 7×7 surface still continued.

4.3. Revisiting Si(111) 7×7 by the STM Inventors

The STM inventors decided to reexamine the 7×7 surface with STM four years after their original experiment.[48] While the first observation of the 7×7 with STM was a significant advance, there were still some ambiguities in the observations that required clarification. To eliminate the issue of surface contamination, they used Auger Electron Spectroscopy, AES, to monitor the cleaning process of the surface before making their observations by STM. AES was one of the most common analysis methods for elemental composition and was sensitive to surface contaminants.

The issue of contamination was a known problem that had raised suspicions during early experiments on the 7×7 reconstructed structure. This was on the minds of the STM inventors. They took care to describe the cleanness of the surface in order to remove all doubts about contamination corrupting the STM images. Contamination had been a persistent nuisance in surface studies, and scrupulous cleaning of the surface was required for any study to be universally accepted. Eliminating the possibility of surface contamination was indispensable for the acceptance or agreement with a newly discovered observation.

[48] G. Binnig, et al., "7×7 Reconstruction,"op.cit. (note 39); G. Binnig, E. Rohrer, F. Salvan, Ch. Gerber, and A. Baro, "Revisiting the 7×7 Reconstruction of Si(111)," *Surface Science*, 157 (1985): L373-L378. Hennig describes and discusses the changing of the STM images from 1980–1990 in detail (Hennig, "Changes in the Design," op.cit. (note 3)).



a)

b)

Figure 11. The top view (a) and the side view (b) of the DAS model by Takayanagi and his colleagues. Reprinted with permission from K. Takayanagi, Y. Tanishiro, M. Takahashi, and S. Takahashi, "Structural Analysis of Si(111)7×7 by UHV-Transmission Electron Diffraction and Microscopy," *Journal of Vacuum Science and Technology A*, 3 (1985):1502–1506. Copyright (1983), American Vacuum Society.

After cleaning, they also measured the LEED pattern of the same sample to confirm the quality of the silicon surface over a wide area. Thus, they could make AES, LEED, and STM measurements on the same sample without having to remove it from the UHV chamber. The degree of meticulous care for the sample in this set of experiments suggested that there could have been some problems or anomalies with the original STM measurements. However, the STM images from these latest experiments contained all of the main features that were seen in the earlier measurements. Incorporating existing established methods, such as LEED and AES, added to the reliability of measurements with newly invented instruments, like STM. This process should have enhanced the credibility of the STM.

The STM inventors hoped that this careful reexamination of the surface would dispel all suspicions and concerns about the use of STM for observation of surface structures. Nevertheless, there was still some skepticism about STM. As Binnig and Rohrer mentioned in their Nobel lecture, "Rumors reached us that scientists would bet cases of champagne that our results were mere computer simulations! The bets were probably based on the fact the STM was already three years old, and atomic resolution was still our exclusive property." The replications of STM played a crucial role in the history of Si(111)7×7, as I describe in the following section.

4.4. Replication of STM

After the first report heralding the invention of STM in 1981, a few researchers took up the challenge to replicate STM instruments in their own laboratories. It took over three years to catch up with the STM inventors. Some researchers also chose to observe the 7×7 surface with their own STM instruments, and their observations supported the DAS model.[49]

The researchers, who replicated the STM, up until late 1986, often created relief-like images from X-Y recorders.[50] The images had the X-Y recorder traces and some thickness for the z-axis direction. The relief-like images played a key role in demonstrating the degree of success of STM replication and thus an important first step in establishing the credibility of the STM measurements. This was, of course, important in cementing their credentials to the STM community by demonstrating the performance of their own STM. But this was just the first step for establishing the credibility and importance of the STM. The second step was to make a significant contribution to the scientific research.

The researchers, who replicated STMs, developed technology and invented new techniques, such as Current-Imaging Tunneling Spectroscopy (CITS). This method overcame some of the inherent problems of previous STMs and allowed for real-space imaging of surface electronic states. One could now separate geometric and electronic information during the course of a scan. This CITS-STM image displayed both filled and empty surface states of the 7×7 surface at a glance (Figure 12).[51]

The STM experiments by Joseph Demuth's group settled the more than twenty-five-year-old question of the 7×7 surface structure in 1986.[52] The new STM created different images depending on the applied voltage at the sample surface. Viewing the surface at negative sample bias voltages resulted in enhanced imaging of the adatoms in the faulted half, while images recorded at positive sample bias voltages revealed twelve adatoms of equal height in each unit cell.[53] It was initially difficult to separate the structural and electronic contributions to the STM images, and this difficulty was an impediment to quantitatively interpreting the images.

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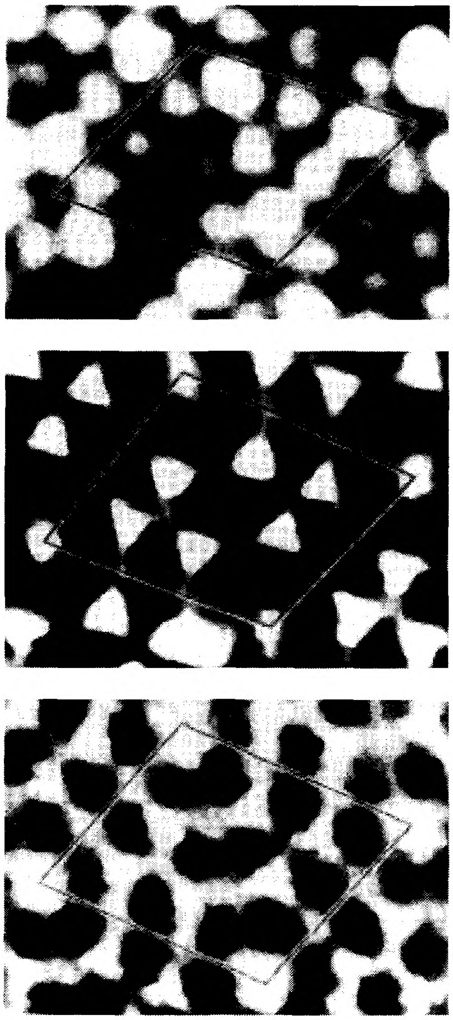
[49] R. S. Becker, J. A. Golovchenko, D. R. Hamann, and B. S. Swartzentruber, "Real-Space Observation of Surface States on Si(111)7×7 with the Tunneling Microscope," *Physical Review Letters*, 55 (1985): 2032–2034; R.J. Hamers, R.M. Tromp, and J.E. Demuth, "Surface Electronic Structure of Si(111)-(7×7) Resolved in Real Space," *Physical Review Letters*, 56 (1986): 1972–1975.

[50] For example, P. Muralt, D. W. Pohl, and W. Denk, "Wide-range, Low-operating-voltage, Bimorph STM: Application as Potentiometer," in *IBM Journal of Research and Development*, 30(1986): 443–450; J. K. Gimzewski and A. Humbert, "Scanning Tunneling Microscopy of Surface Microstructure on Rough Surfaces," ibid.: 472–477; M. Ringger, B. W. Corb, H. R. Hidber, R. Schlögl, R. Wiesendanger, A. Stemmer, L Rosenthaier, A. J. Brunner, PC. Oelhaften, and H.-J. Güntherodt, "STM Activity at the University of Basel," ibid.: 500–508; H. Tokumoto, H. Bando, W. Mizutani, M. Okano, M. Ono, H. Murakami, S. Okayama, Y. Ono, K. Watanabe, S. Wakiyama, F. Sakai, K. Endo, and K. Kajimura, "Observation of Atomic Image of 2H-$NbSe_2$ Surface by Scanning Tunneling Microscope," *Japanese Journal of Applied Physics*, 25 (1986): L621-L623.

[51] Hamers, et al., "Surface Electronic Structure," op.cit. (note 48).

[52] R. M. Tromp, R. J. Hamers, and J. E. Demuth, "Atomic and Electronic Contributions to Si(111)-7×7 Scanning-Tunneling-Microscopy Images," *Physical Review B*, 34 (1986): 1388–1391.

[53] Ibid.



(a)

(b)

(c)

Figure 12. CITS images of Si(111)7×7: (a) adatom state at −0.35V; (b) dangling-bond state at −0.8V; (c) backbond state, state of a chemical bond between an atom in the first layer and an atom in the second layer, at −1.7V. Reprinted figure with permission from R. J. Hamers, R. M. Tromp, and J. E. Demuth, "Surface Electronic Structure of Si(111)-(7×7) Resolved in Real Space," *Physical Review Letters*, 56 (1986): 1972–1975. Copyright (1986) by the American Physical Society.

Demuth and his coworkers turned to calculations using the atomic charge superposition method to create theoretical images of the surface, but these images did not contain specific details on surface-induced electronic states. They only reflected the atomic geometry of the structure.[54] They found quantitative agreement between calculated and measured images for the Si(111)7×7 surface, which established the value of such calculations. To compare the calculated images with the results from their experiments, the Demuth group

[54] J. Tersoff and D. R. Hamann, "Theory of the Scanning Tunneling Microscope," *Physical Review B*, 31 (1985): 805–813.

simulated five models of the 7×7 reconstruction (Figure 13). The experimental images were consistent with simulation images of Binnig (a) and Takayanagi's (DAS) model (e).

The top views of both models were in good agreement with the experimental data, but from the cross-section view, the DAS model was clearly the one that has the best agreement with the experimental data. This indicated that the observations from the STM experiments were needed to support the DAS model and thus led to the settlement of the controversy over the Si(111)7×7 surface structure. Subsequent to this determination, X-ray diffraction

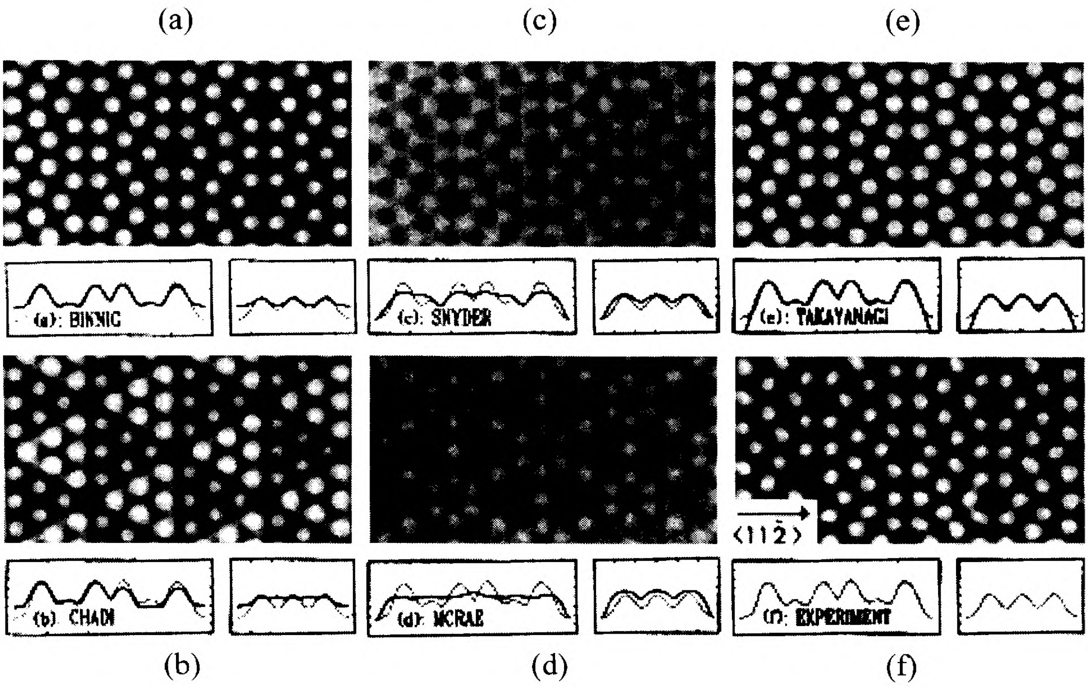


Figure 13. (a) Binnig model, which is a modified adatom model from STM observation. (b) The Chadi model was derived from the energy-minimization calculations on various types of adatom models to compare with the seven experimental data sets available.[55] (c) Snyder and his colleagues proposed a modified milk-stool model that employed a quantum chemistry computer program, GAUSSIAN-80, to reexamine the milk-stool model.[56] (d) From the results of their TED work, McRae and Petroff proposed a triangle-dimer model, which has the unit cell containing nine dimers or pairs of dimers bordering a triangular subunit of the unit cell.[57] (e) Takayanagi's (DAS) model derived from TED work. (f) The experimental image obtained with +2V. Simulated STM images and cross-section views of five models and the result of the experiment. In the cross-section view, the left panel shows the longer diagonal of the unit cell, while the right shows the shorter diagonal. Reprinted figure with permission from R. M. Tromp, R. J. Hamers, and J. E. Demuth, "Atomic and Electronic Contributions to Si(111)-7×7 Scanning-Tunneling-Microscopy Images," *Physical Review B*, 34 (1986): 1388–1391. Copyright (1986) by the American Physical Society.

[55] D. J. Chadi, "New Adatom Model for Si(111)7×7 and Si(111)-Ge 5×5 Reconstructed Surfaces," *Physical Review B*, 30 (1984): 4470–4480.

[56] L. C. Snyder, "Modified Milk-Stool on Wurtzite Layer Model for Si(111)7×7 Surface Reconstruction," *Surface Science*, 140 (1984): 101–107.

[57] E. G. McRae and P. M. Petroff, "Test of Si(111)7×7 Structural Models by Comparison with Transmission Electron Diffraction Patterns," *Surface Science*, 147 (1984): 385–395.

and ion-scattering measurements provided further support for the DAS model.[58]

5. Conclusion

Farnsworth and his colleagues discovered the 7 × 7 structure of Si(111) reconstructed surface with LEED. The Farnsworth type and Germer type of LEED merged and the combination invigorated surface science. LEED also established the method of using reciprocal space images in surface science. After the 1960s, along with the improvement of generating ultra-high vacuum, numerous instruments were developed and applied to the analysis of surface structures. However, the Si(111)7×7 reconstructed surface was too large and complicated, and the existing methods were insufficient to determine its structure. The STM provided crucial information for solving this long standing problem. It was an improved STM that ultimately identified the arrangement of atoms of the DAS model, which was derived from TED research.

When a new observation was reported, such as the discovery of the 7×7 structure and the observation with the STM, surface scientists casted suspicious eyes and reexamined it with other existing methods in order to ensure the sample was not contaminated and confirm the observation. To solve a particular problem in surface science, information from other laboratories using different experimental methods, providing required as the data from various types of experiments, were needed for comparison with their data. The instruments for surface science were thus shown to have a mutually complementary relationship. While the STM settled the controversy of the Si(111)7×7, it was not solely the STM that led to the settlement, but rather an accumulation of the research on the Si(111)7×7 surface over a number of years.

The STM inventors took a strategy to persuade scientific community and to establish its firm position in the field of surface science. Their strategy was choosing a hot topic and to create images of the surface with ingenuity. The Si(111)7×7 problem was challenging, had been tackled by many other researchers without resolution, and would therefore continue to attract researchers. Moreover the inventors convinced the STM would contribute to that controversy.

Binnig and his colleagues created the two kinds of the STM images to establish its credibility. The first one was the relief with a three-dimensional representation. The relief had double role for representation. The one contributed to observe the image intuitively, and the other was the process of imaging. The latter helped its credibility. The STM inventors never created the relief again, but other researchers, in replicating STMs, created this relief-type representation to show the level of performance of their own STMs. The role of the relief image changed from its original purpose. The second computer-processed

[58] H.-J. Gossmann, J. C. Bean, L. C. Feldman, E. G. McRae, and I. K. Robinson, "7×7 Reconstruction of Ge(111) Surfaces under Compressive Strain," *Physical Review Letters*, 55 (1985): 1106–1109; I. K. Robinson, W. K. Waskiewicz, P. H. Fuoss, J. B. Stark, and P. A. Bennett, "X-ray Diffraction Evidence of Adatoms in the Si(111)7×7 Reconstructed Surface," *Physics Review B*, 33 (1986): 7013–7016; R. M. Tromp and E. J. van Lohnen, "Ion Beam Crystallography of Silicon Surfaces III, Si(111)-(7×7)," *Surface Science*, 155 (1985): 441–479.

image, was for the discussion and analysis, presenting the locations of each atom with contrasting density. It was easy to recognize individual atoms without well-trained eyes and complicated calculations as analysis of reciprocal space patterns and scattering patterns. The STM inventors' strategy for acceptance of the STM was successful and established a firm position as one of the most powerful instruments in surface science research.

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