My Life With Erwin
The Beginning of an Atom-Probe Legacy

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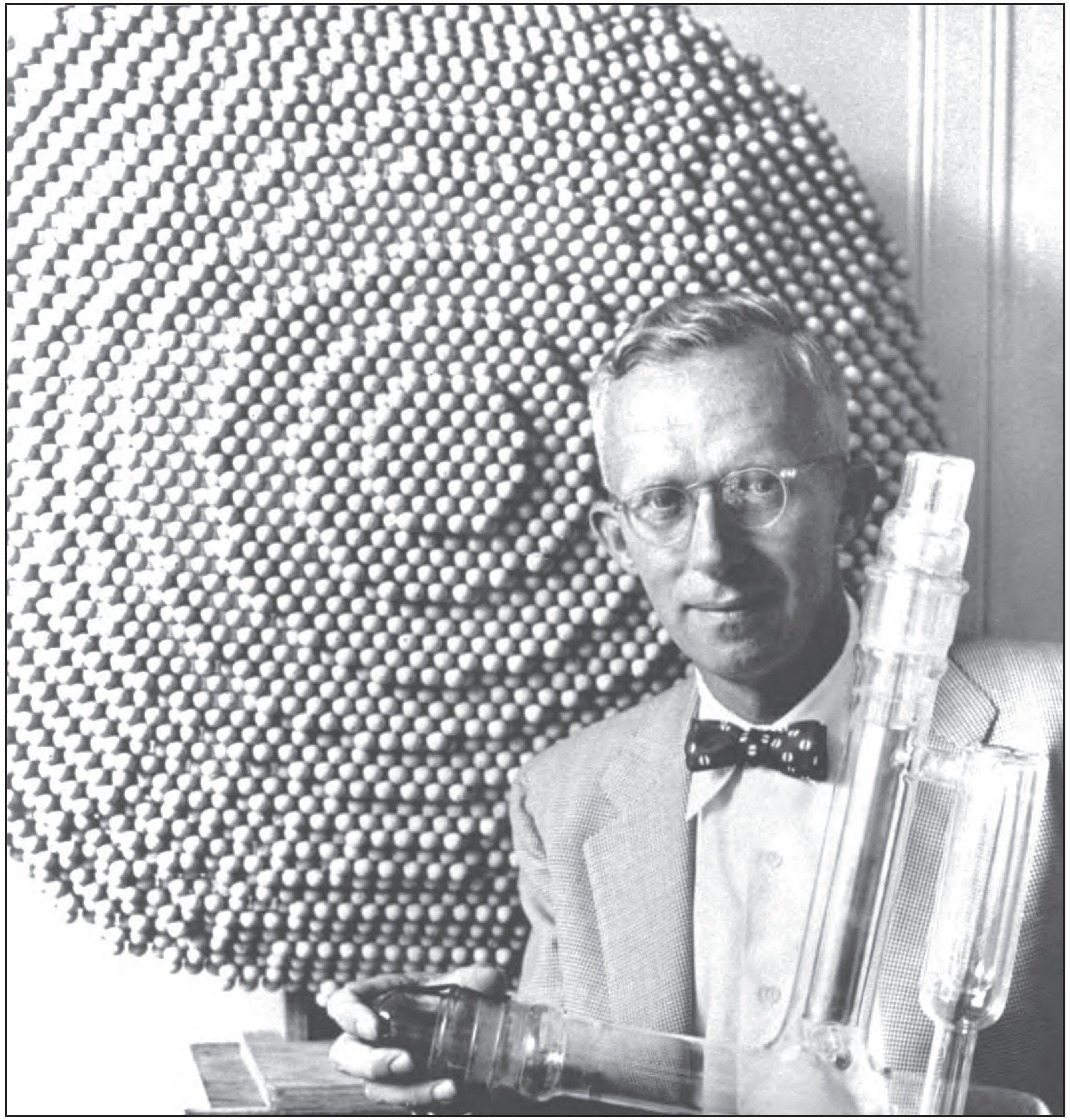
The 21 Club
The University of New Mexico

Centennial Engineering Center
Stamm Room
November 13, 2018

The Atom-Probe was Erwin Wilhelm Müller's last major achievement



1911: Born June 13 in Berlin, Germany.

1936: Degree with Nobel Laureate Gustav Ludwig Hertz.

1937: Invented the Field Emission Microscope.

1941: Discovered Field Desorption.

1951: Invented the Field Ion Microscope.

1952: Joined the Penn State faculty.

1956: First observation of individual atoms.

1966: Invented the Atom-Probe Field Ion Microscope.

1975: Elected to the National Academy of Science*.

1975: Elected to the National Academy of Engineering.

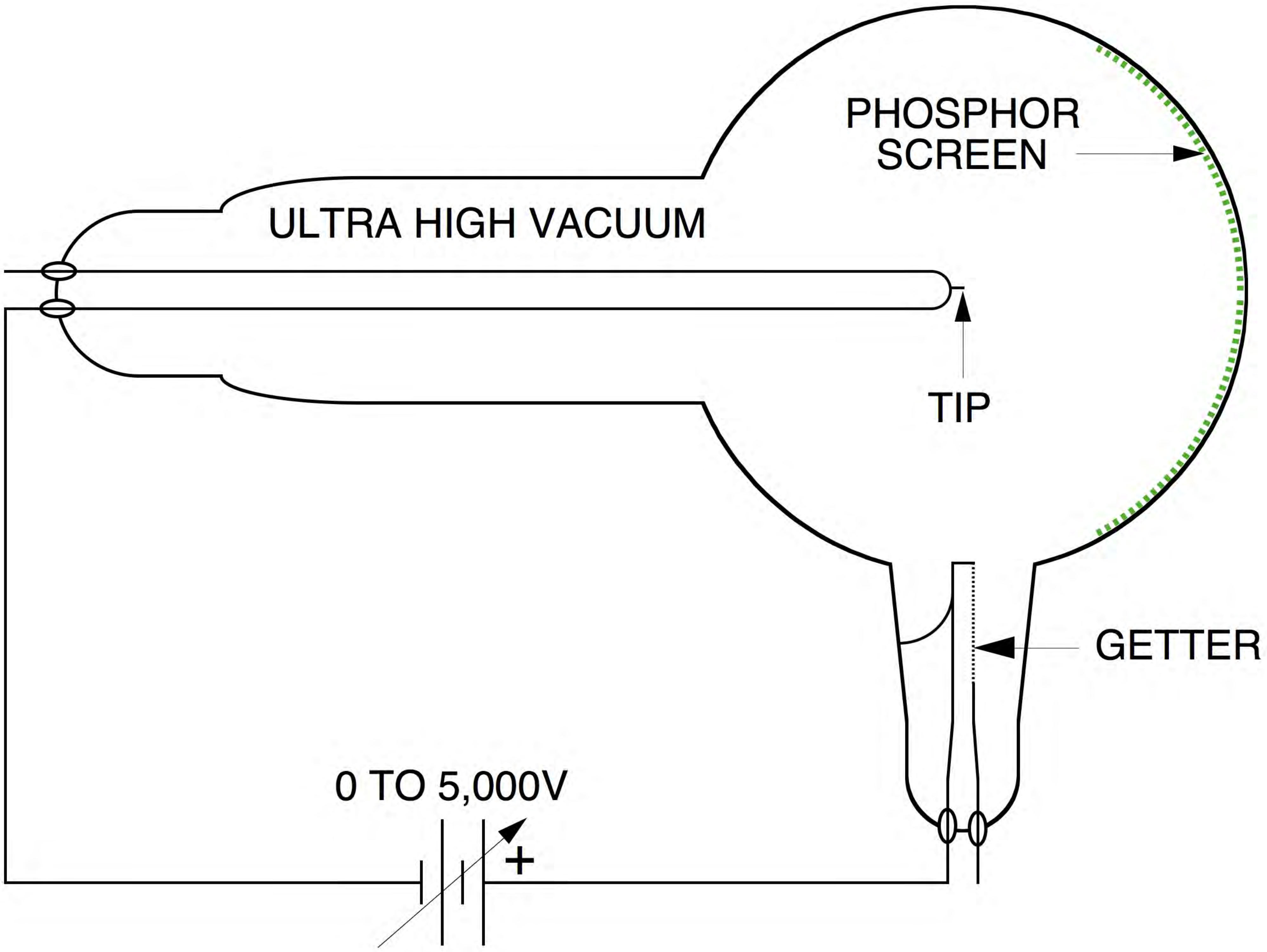
1975: Nominated for the National Medal of Science.

1977: Died, May 17 of a heart attack.

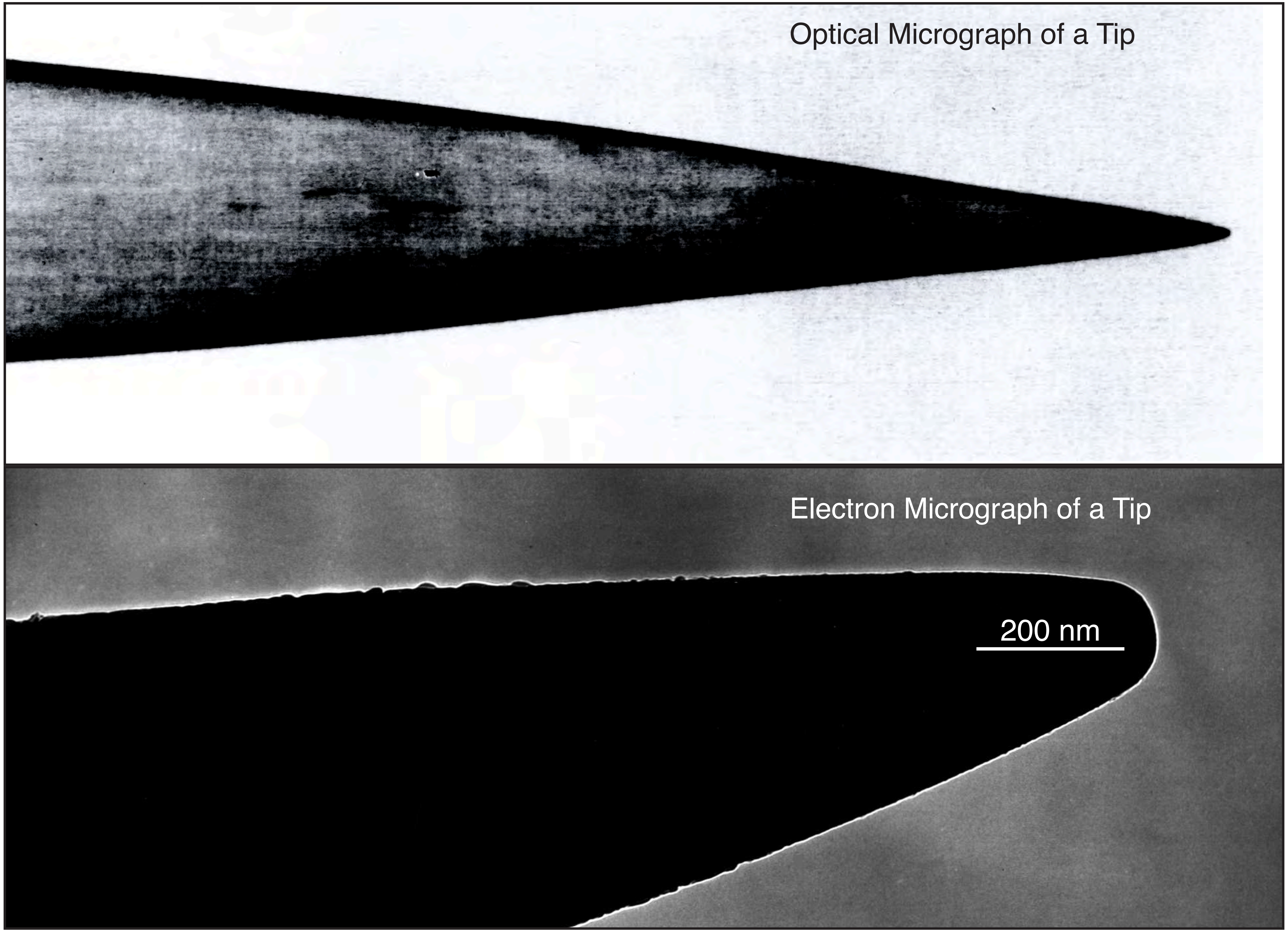
1977: Awarded the National Medal of Science posthumously (in November 22 by President Carter at the White House).

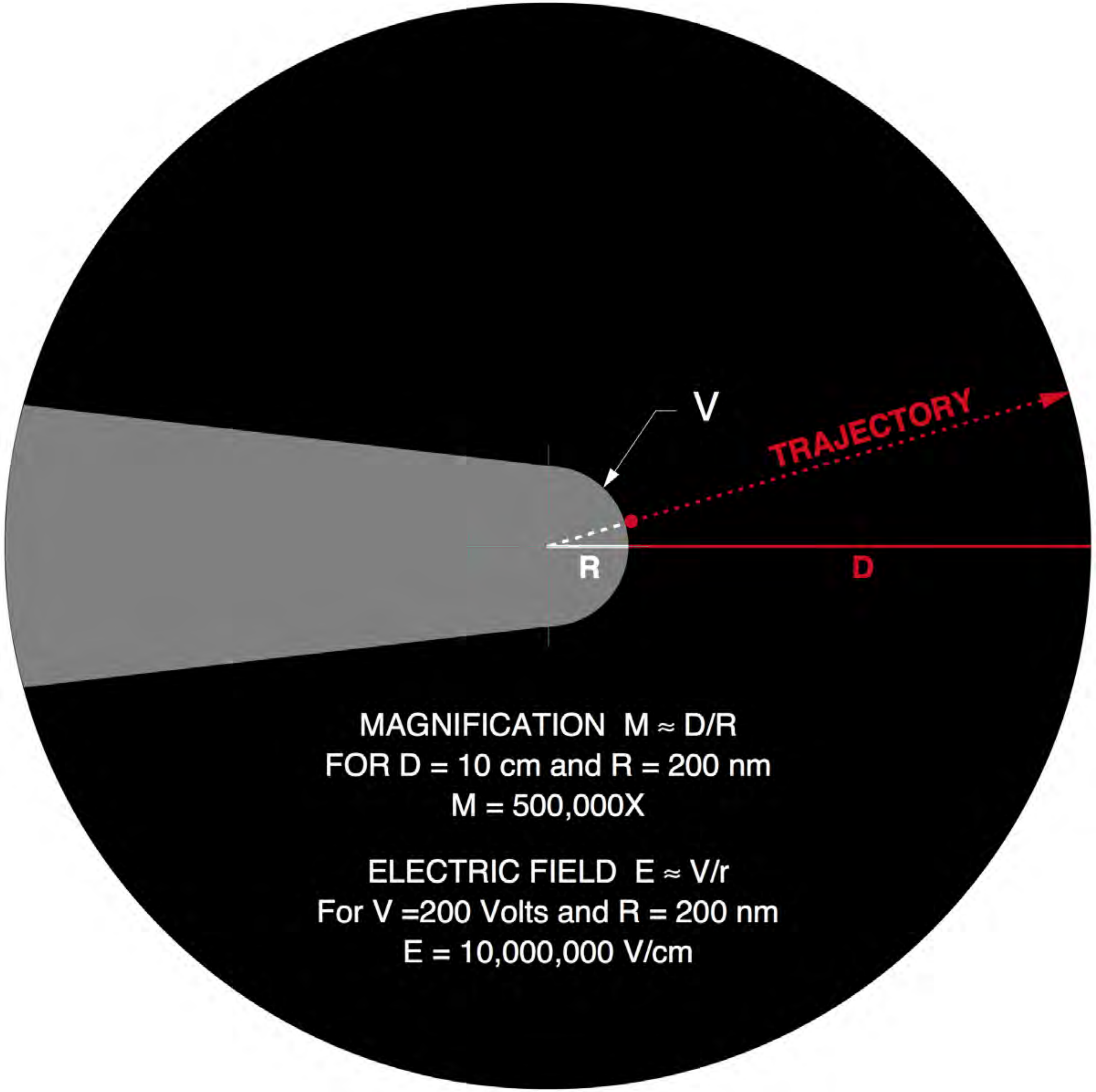
*Under consideration for a Nobel Prize in Physics for the Atom-Probe Field Ion Microscope (with Ernst Ruska for the electron Microscope).

The progenitor of the Atom-Probe was the Field Emission Microscope

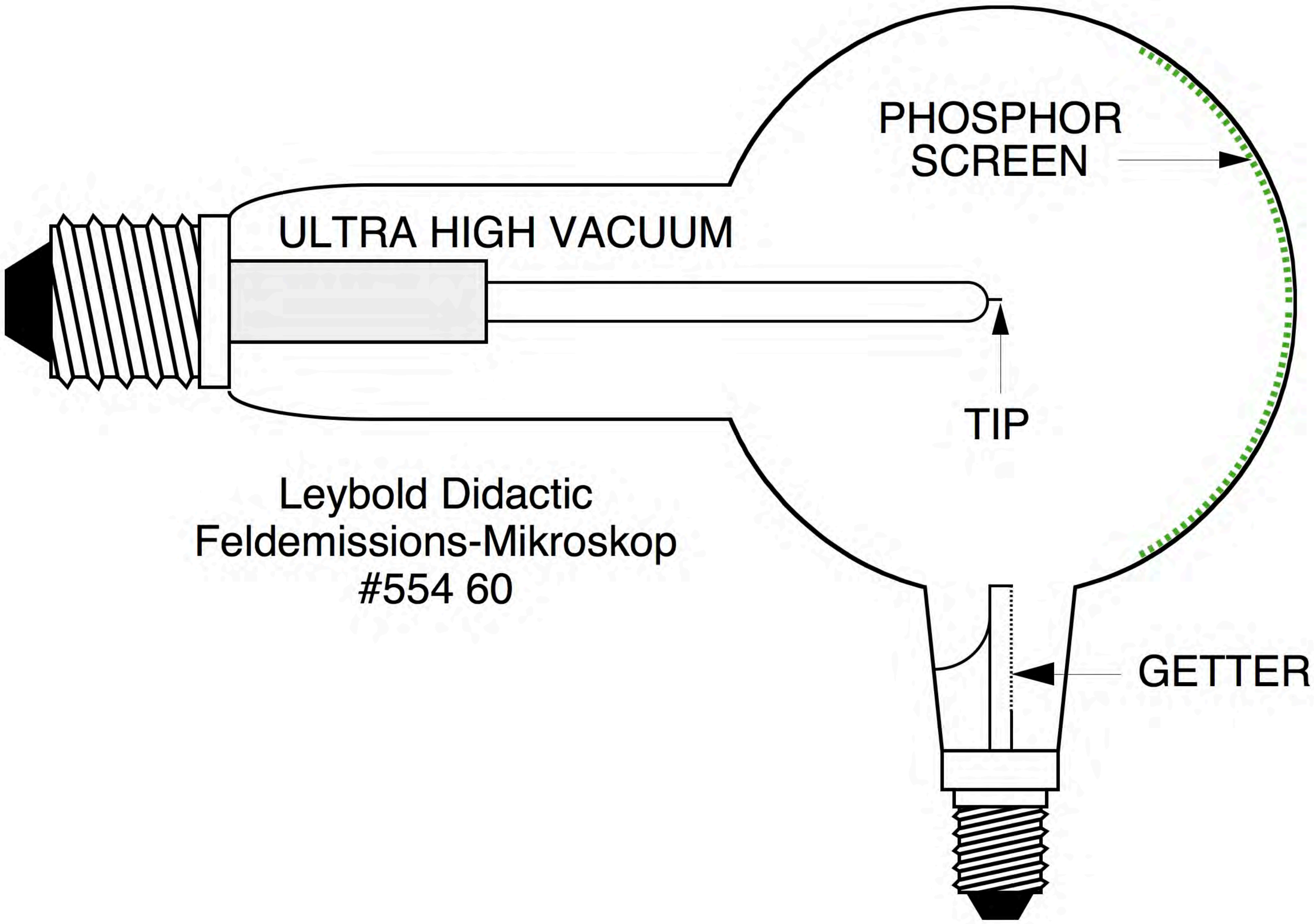


The apex of the tip is approximately spherical




V
TRAJECTORY
R
D
MAGNIFICATION M ≈ D/R
FOR D = 10 cm and R = 200 nm
M = 500,000X
ELECTRIC FIELD E ≈ V/r
For V =200 Volts and R = 200 nm
E = 10,000,000 V/cm

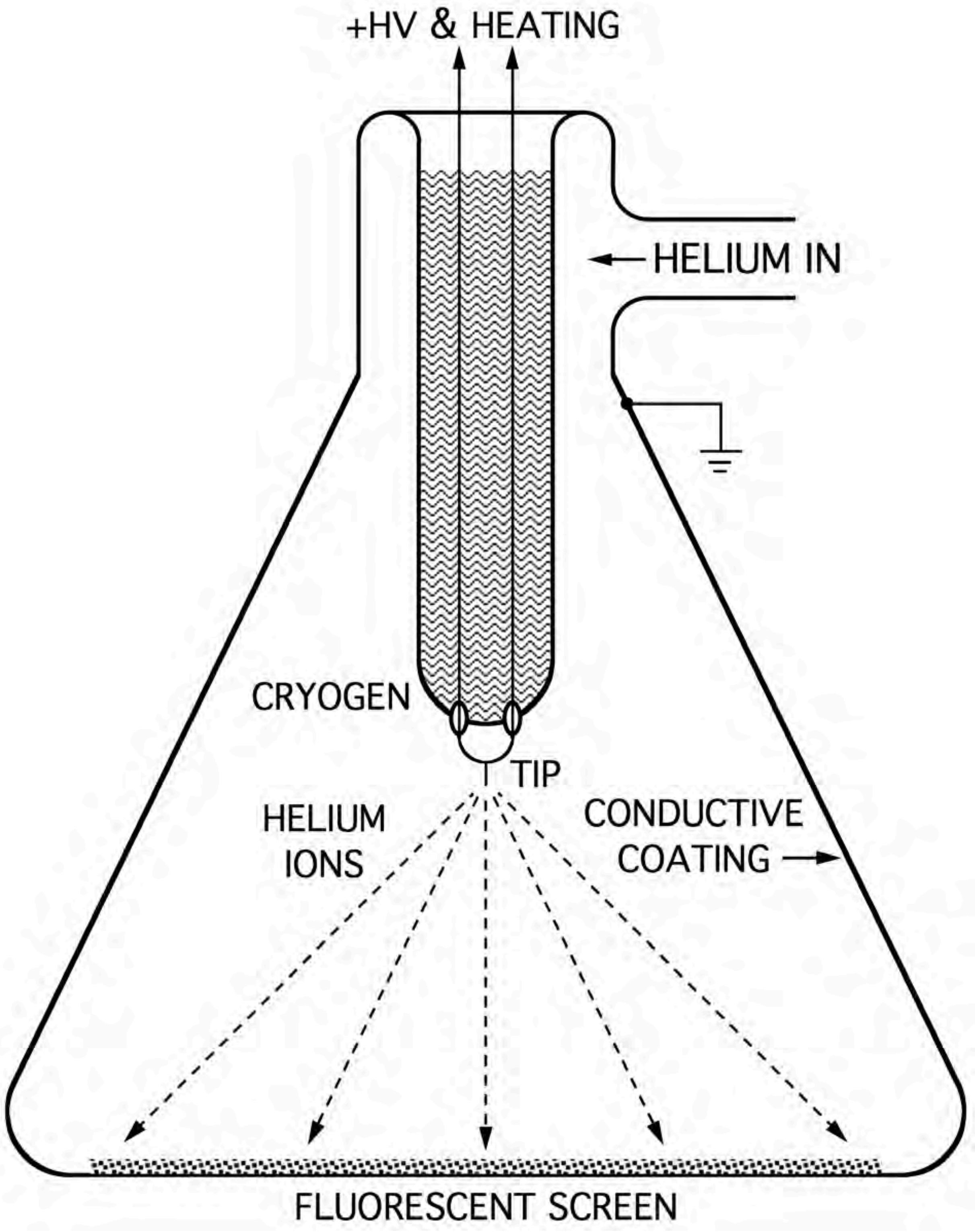
A commercial Field Emission Microscope is available

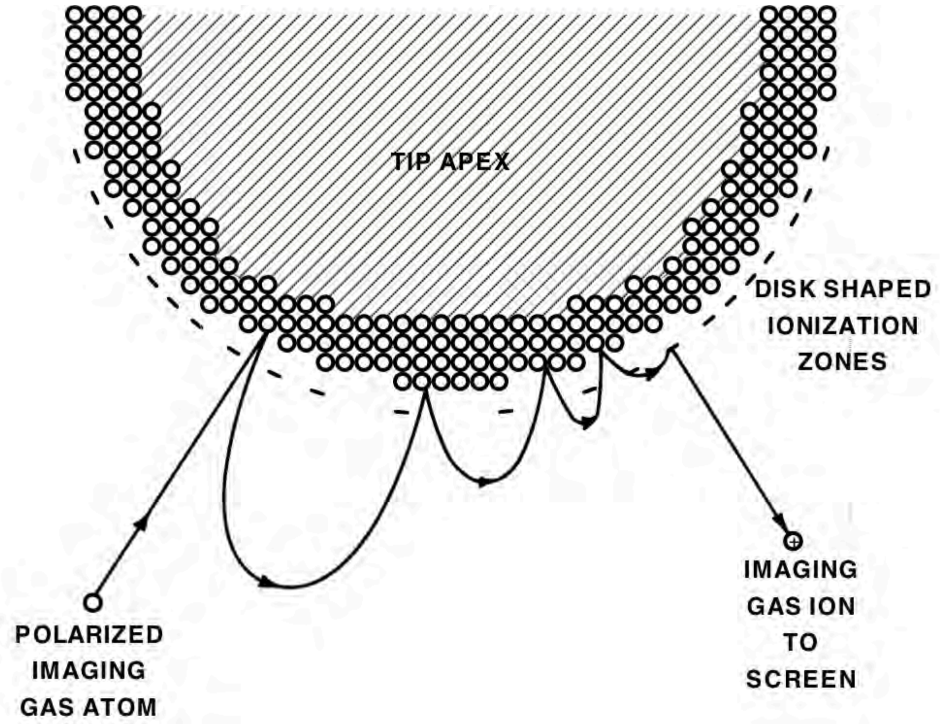


Leybold Didactic
Feldemissions-Mikroskop
#554 60

My Atom Probe legacy began in 1966 at Penn State in the Field Emission Laboratory

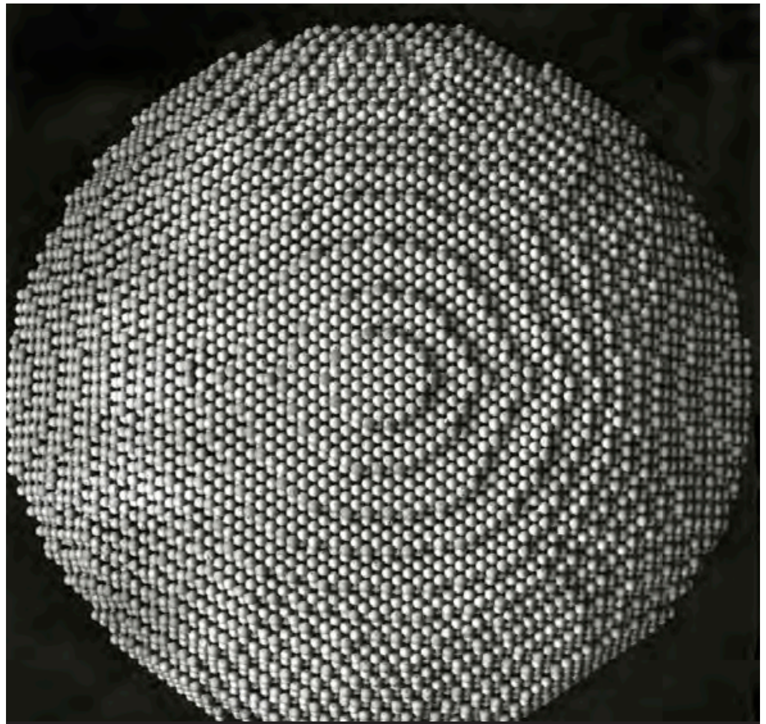
1. **John Panitz**
2. **Douglas Barofsky**
3. Klaus Rendulic
4. **Brooks McLane**
5. Jay Politzer
6. Myron Hicks
7. Tien Tzou Tsong
8. **Gerry Fowler**
9. Sandy Mori (Secretary)
10. Osamu Nishikawa
11. **Professor Müller**
12. Albert Nishikawa



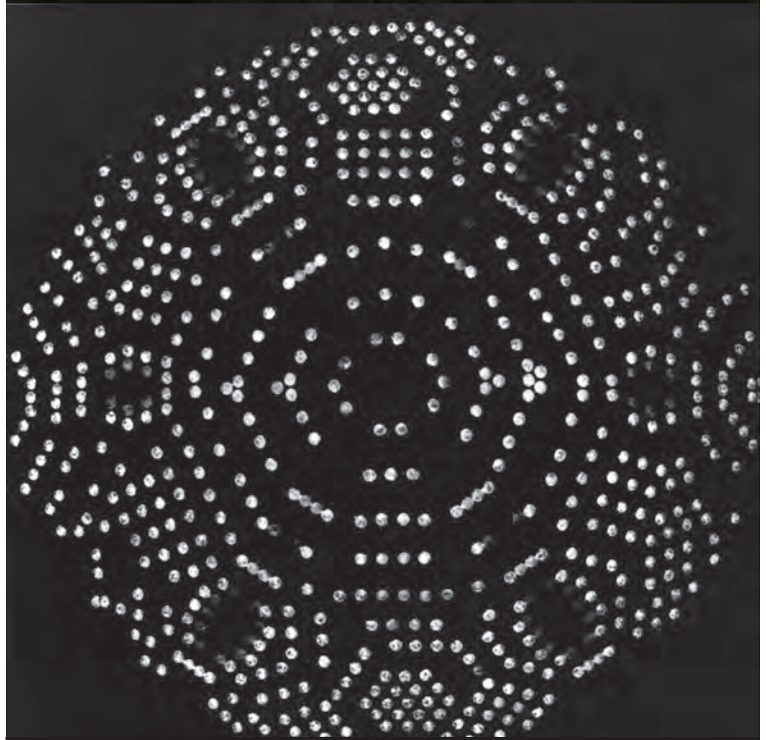


Gas phase helium atoms in a Field Ion Microscope are polarized and attracted to the tip apex where they thermally accomodate to the tip temperature in a series of hopping motions. When a polarized atom passes through an ioniztion zone located ≈ 4Å above each protruding atom on the tip surface an electron will tunnel back into the tip, producing ions that accelerate radially from the tip apex to a fluorecent screen form an image. When a critical field strength is reached, protruding atoms are removed from the surface as positive ions that accelerate from the apex to produce an atomically smooth surface. This is the ultimate cleaning procedure!

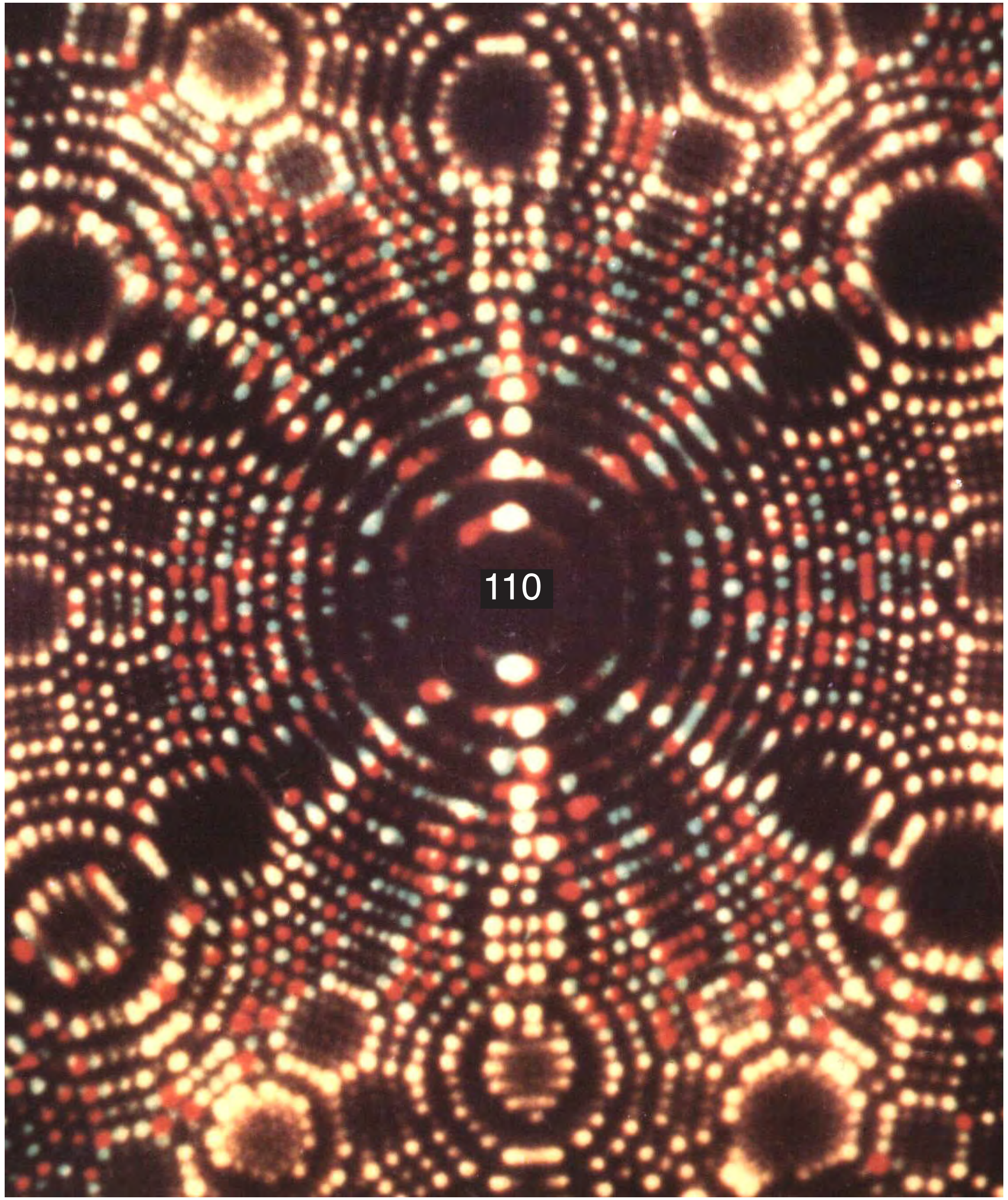
Atomic resolution imaging in the Field Ion Microscope was a routine procedure



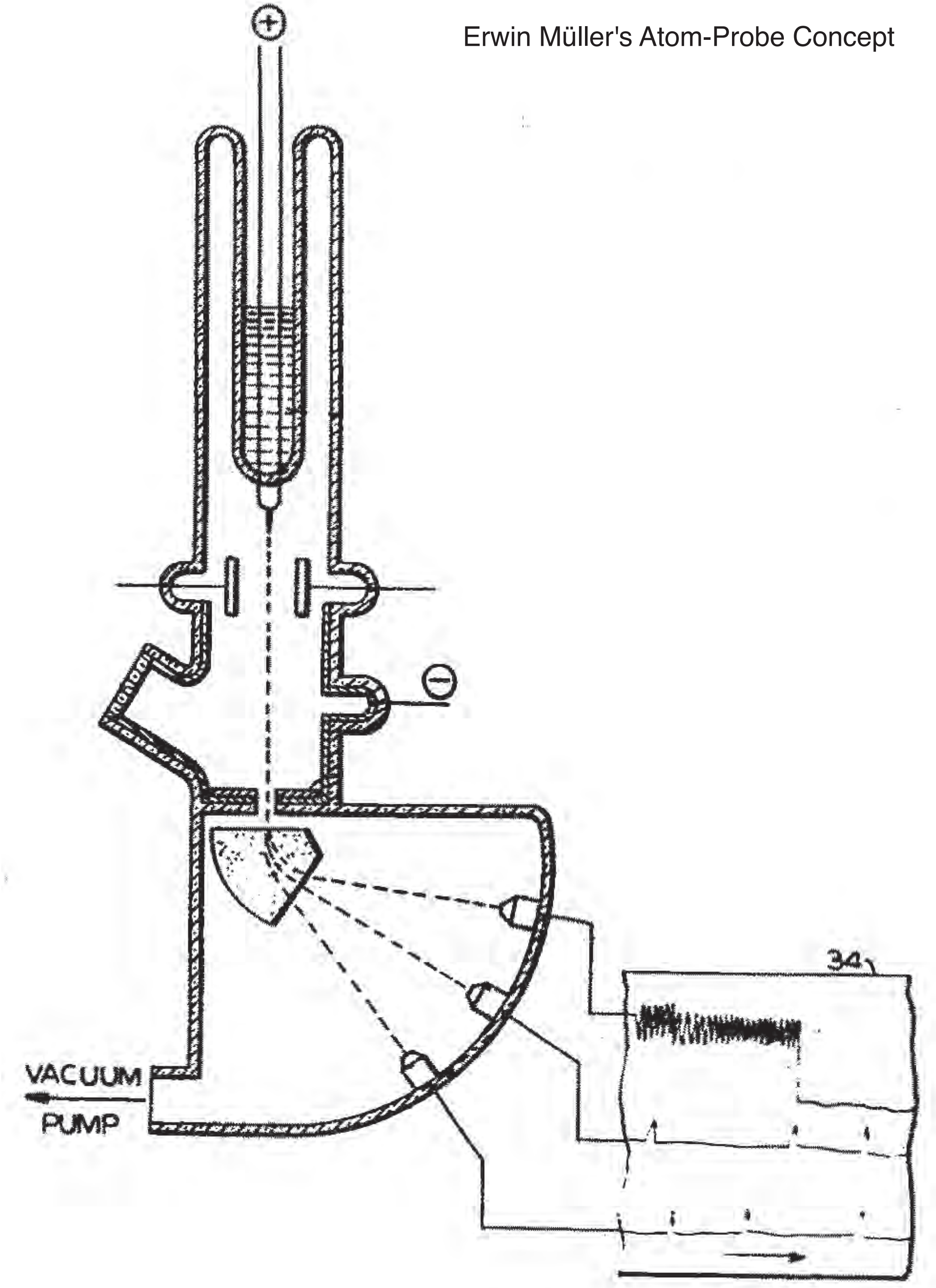
Ball model of a 110 oriented tip apex



Protruding atoms on the ball model



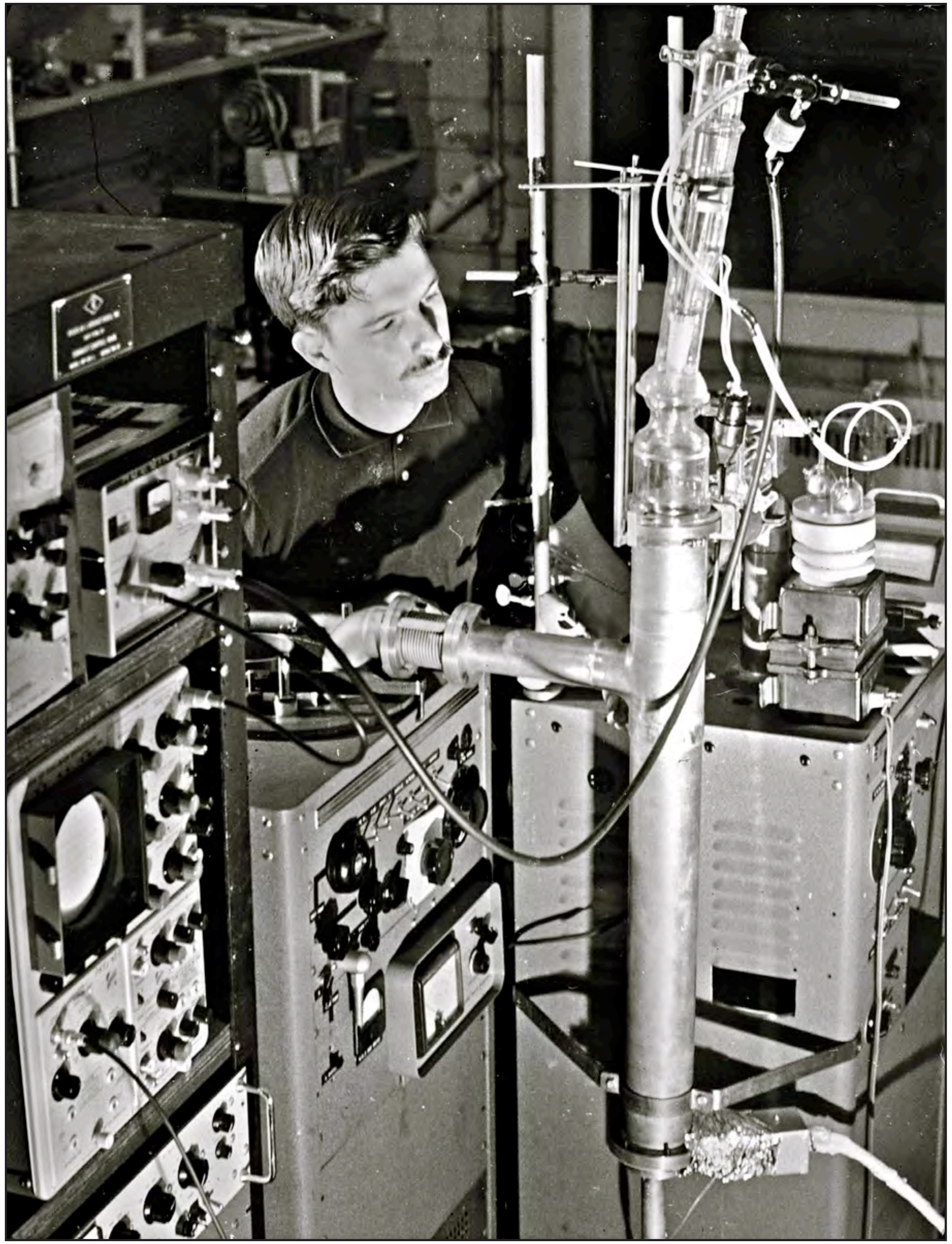
- A ball model of the tip apex can reflect the appearance of a Field Ion Microscope image.
- Changes in the tip apex could be observed by an optical color comparison of two black and white images projected through red and green filters. An optical color comparison of a tungsten image before ion implantation (red) and after ion implantation (green) is shown at the left Yellow (red + green that overlap) indicate no change in the surface of the tip apex.
- The chemical identity (m/z) of an atom in an image could not be determined.

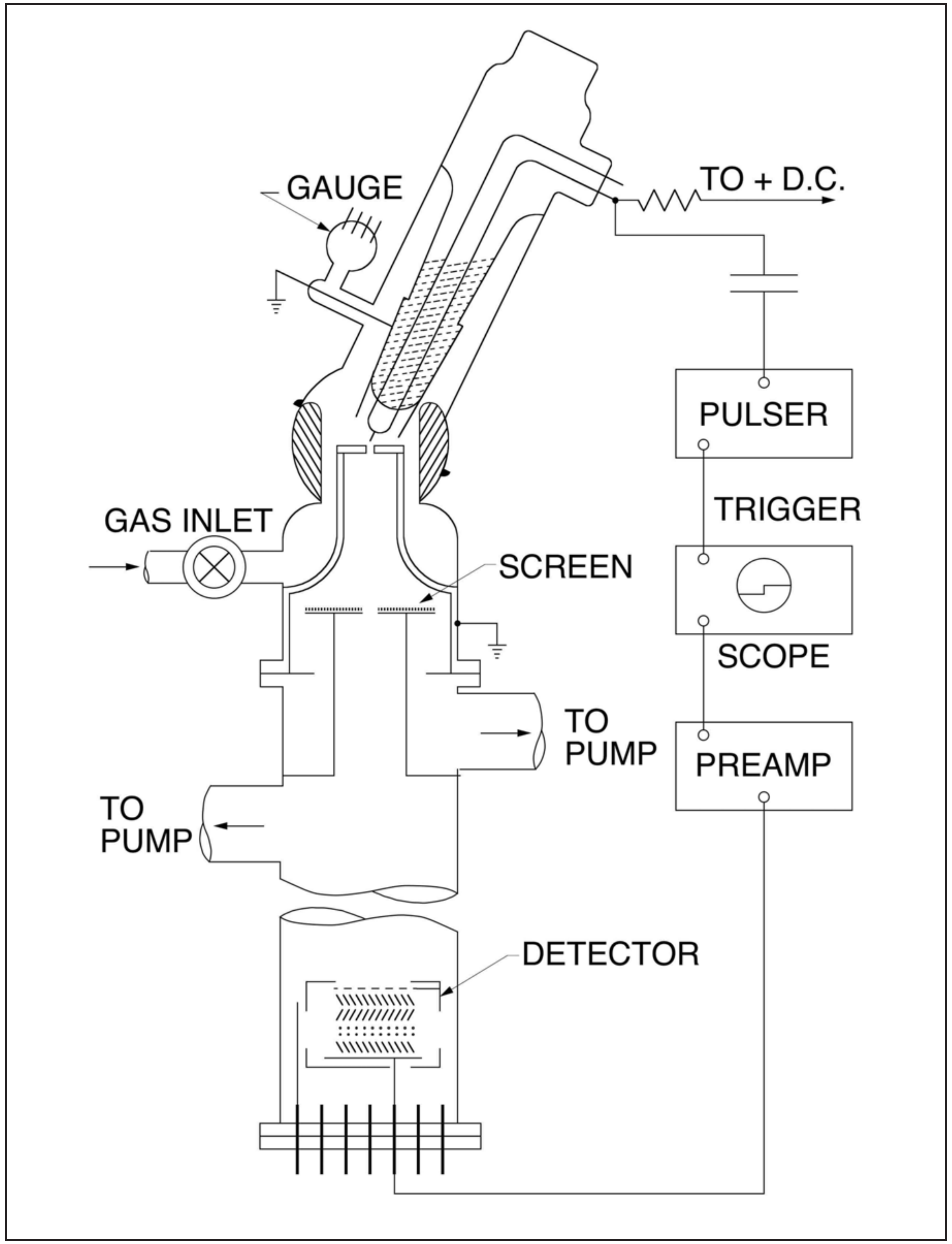


- Erwin had returned from european travel and suggested the concept of an Atom Probe Field Ion Microscope.
- Erwin called the device an "Atom Probe" in analogy with Robert Castaing's "Electron Probe".[1]
- The chemical identity (m/z) of an individual image spot in a Field Ion Microscope image would be determined by mass spectrometry.
- John Panitz had completed an M.S. thesis with Erwin Müller.
- Douglas Barofsky was completing a Ph. D. thesis with Erwin Müller that used a magnetic sector mass spectrometer and suggested using a Time-of-flight (TOF) mass spectrometer for the Atom-Probe.
- A team was assembled: Erwin Müller (Provide Focus and direction) John Panitz (Create a single atom detector, a mass calibration technique and assist Gerry Fowler in constructing the instrument) Brooks McLane (Provide Electronic Support)

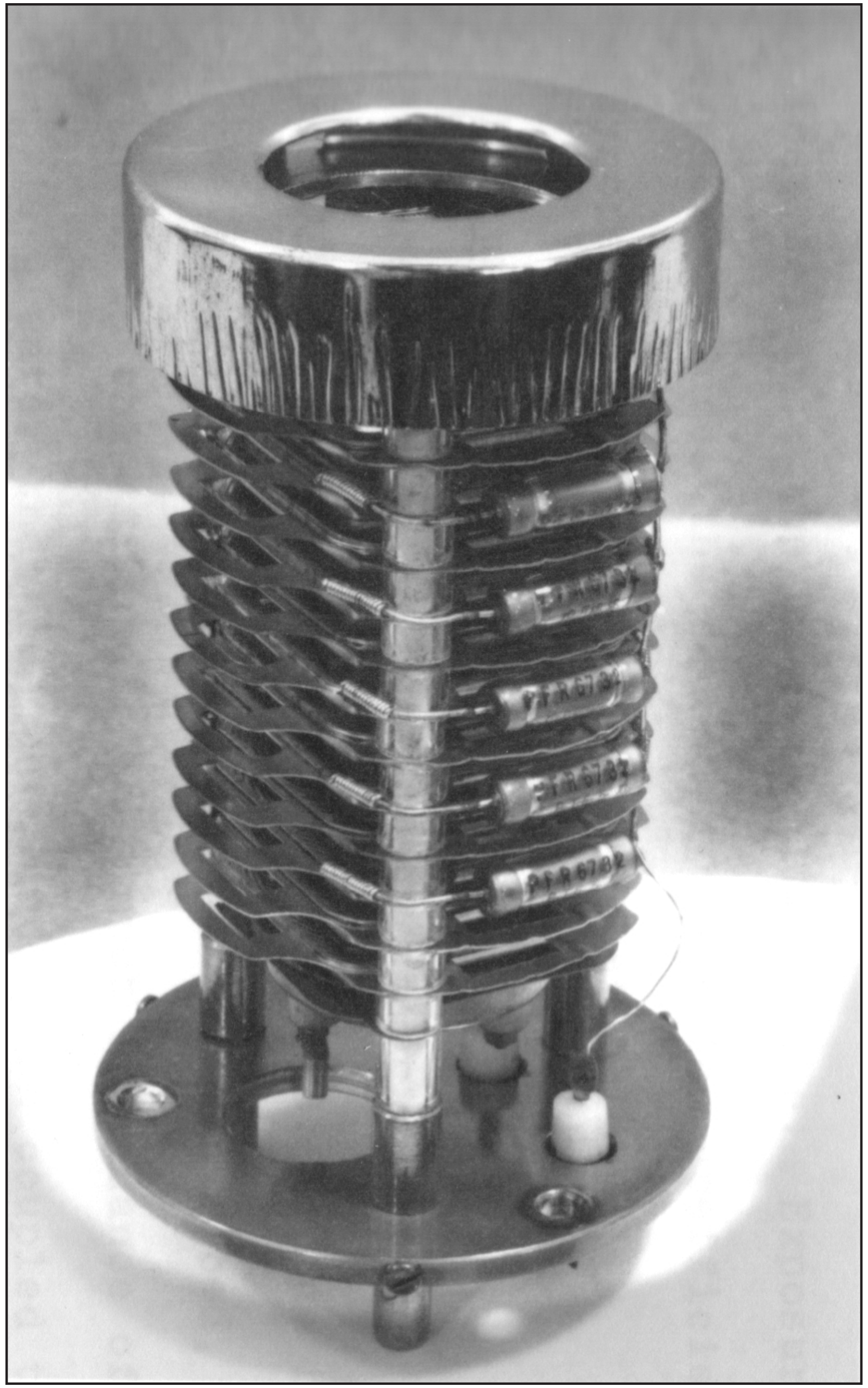
1. R. Castaing. *Electron Probe Microanalysis*. Adv. Electronics Electron Phys. 13 (Academic Press, NY 1960) 317.

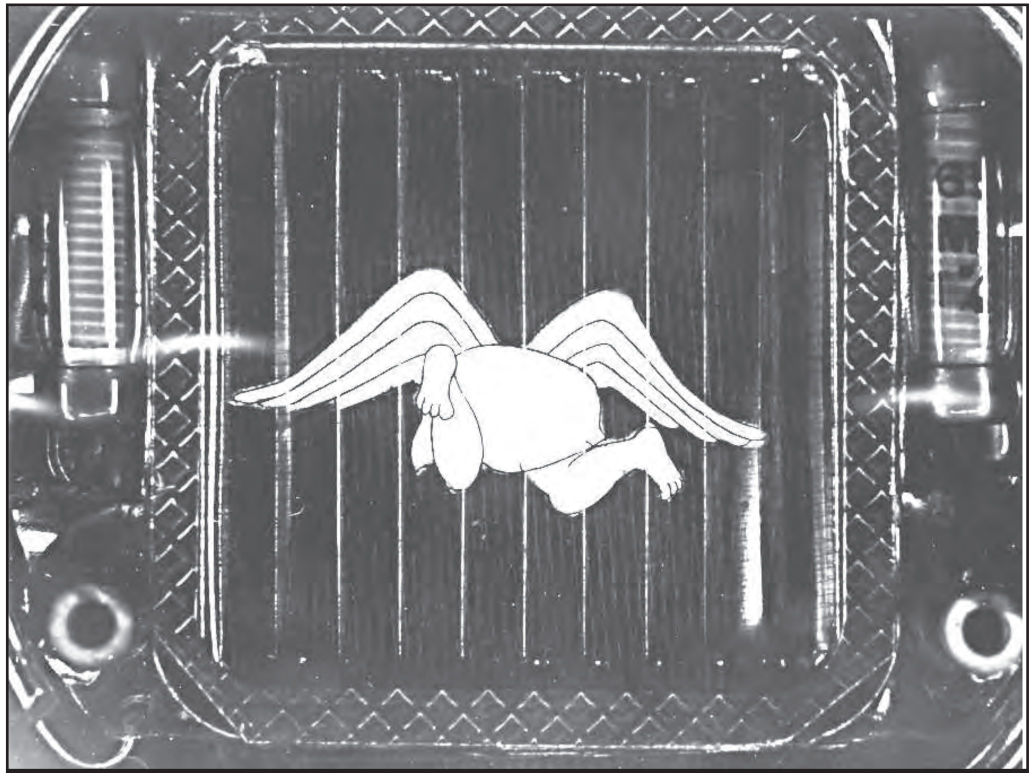
During 1966 Gerry Fowler and I assembled the first Atom-Probe Field Ion Microscope

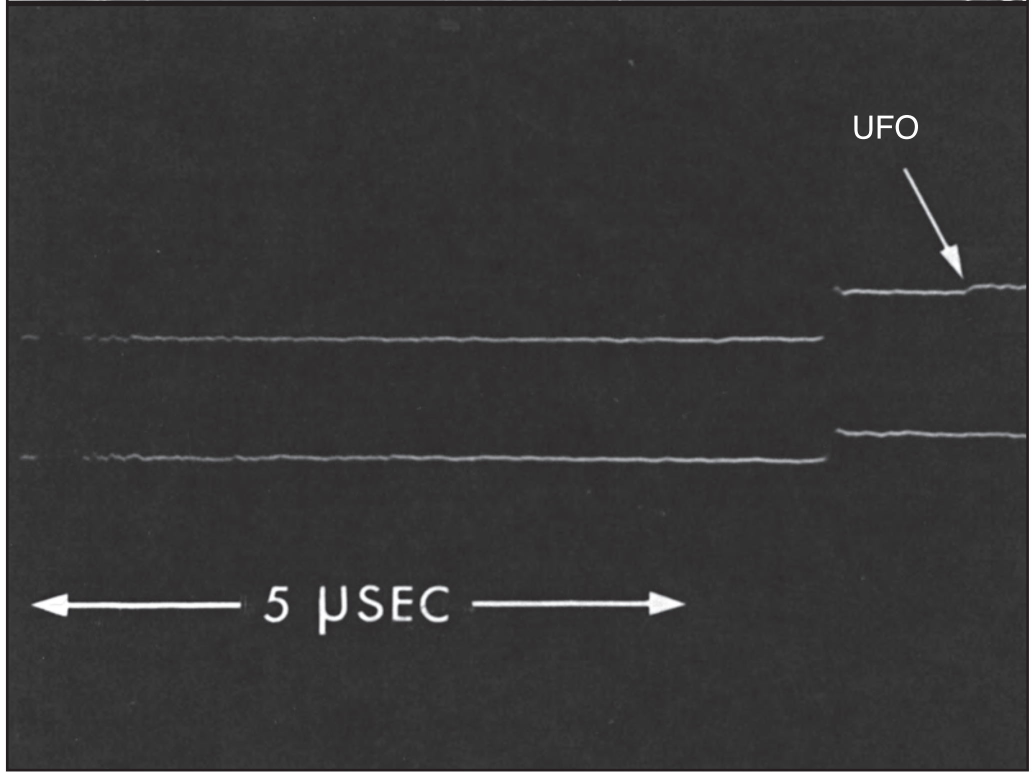


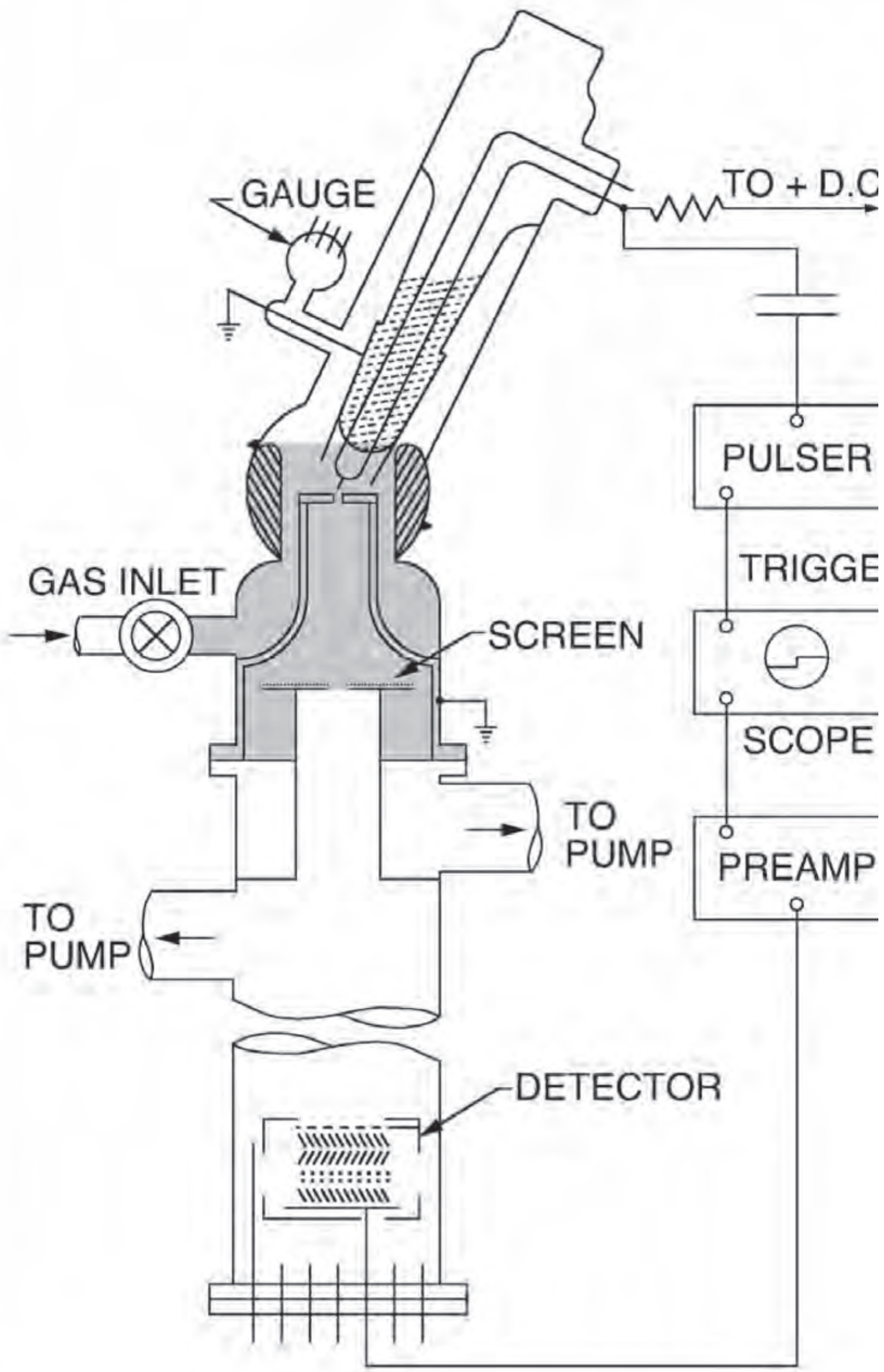


Single atoms and “UFOs” were found using the detector I had constructed

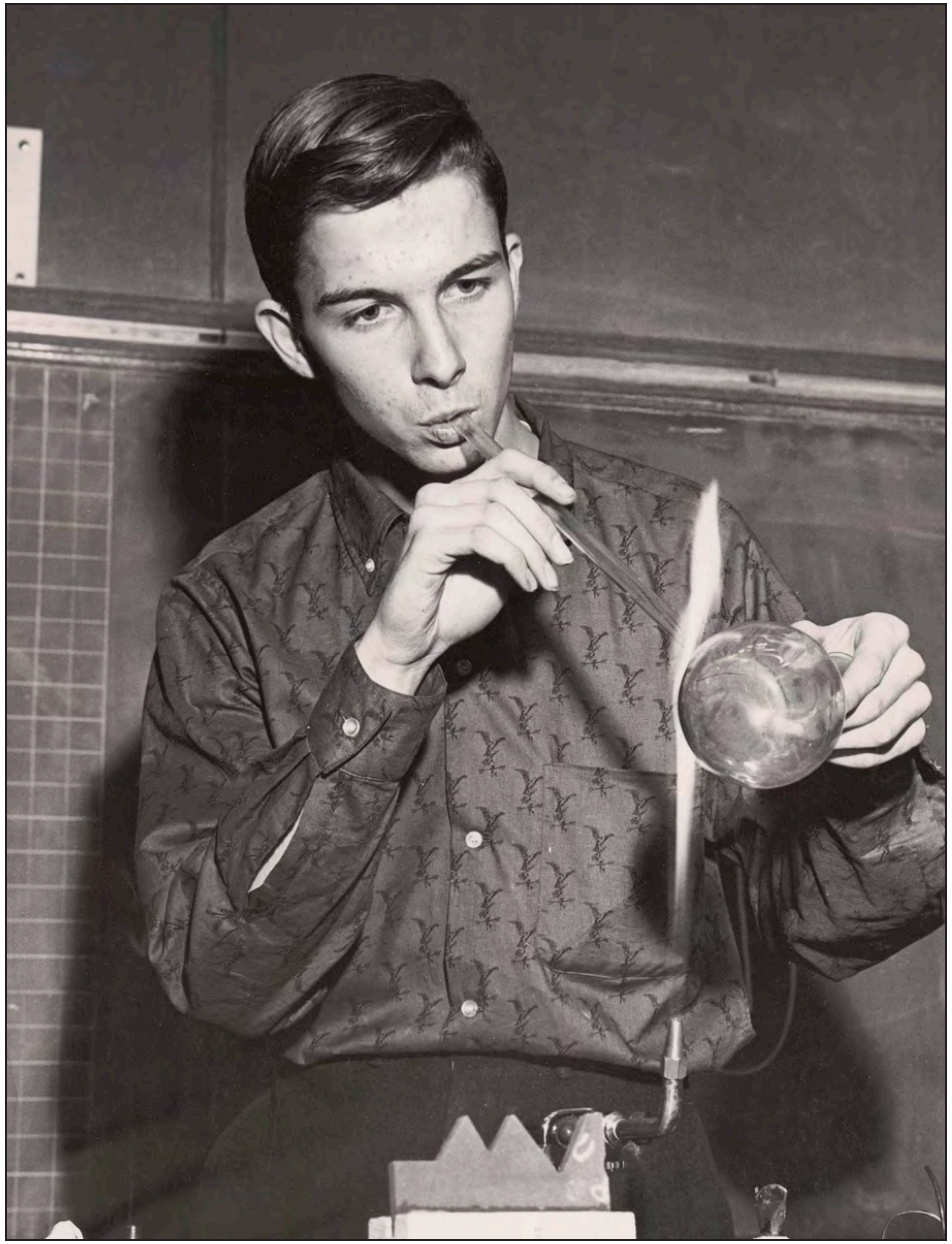


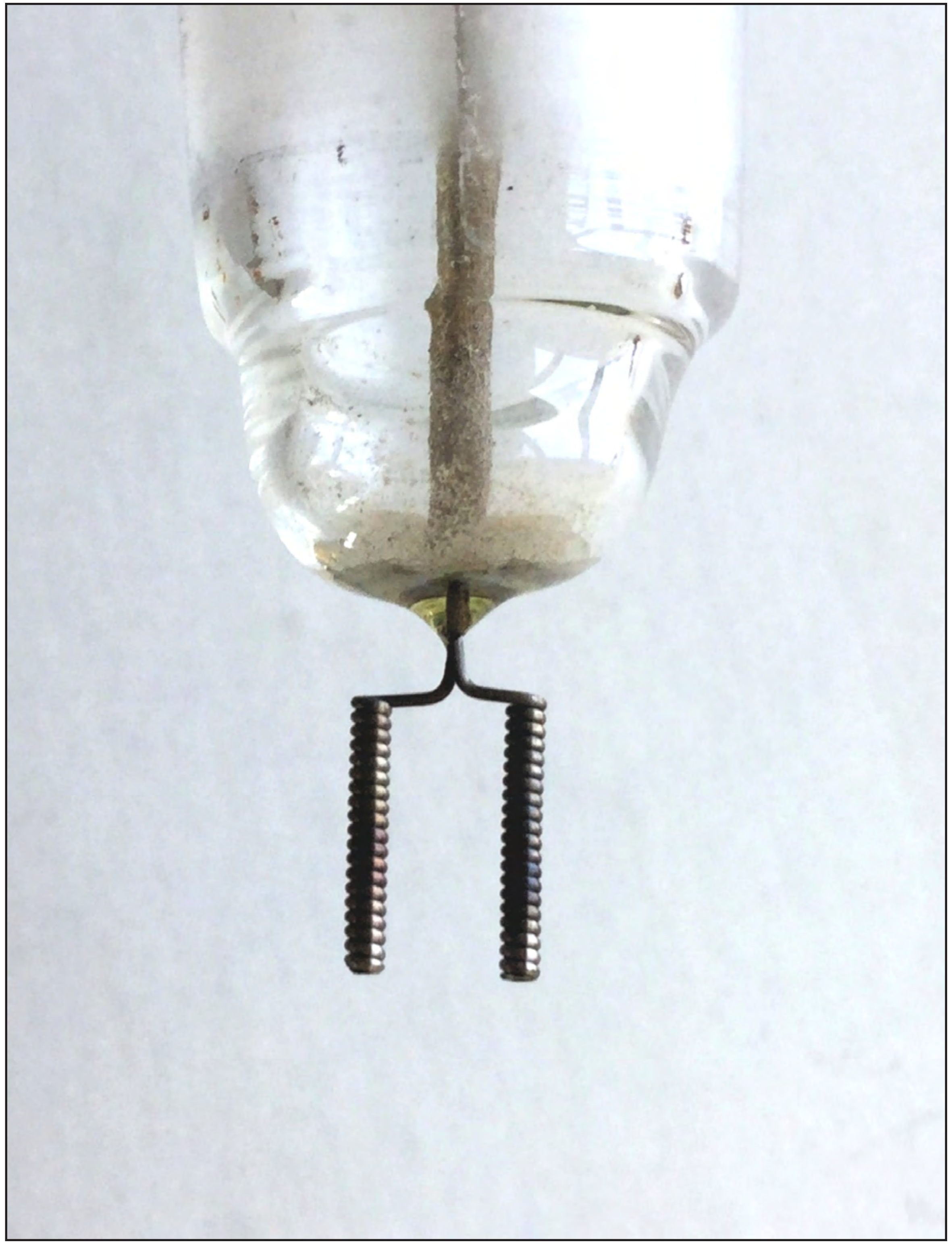




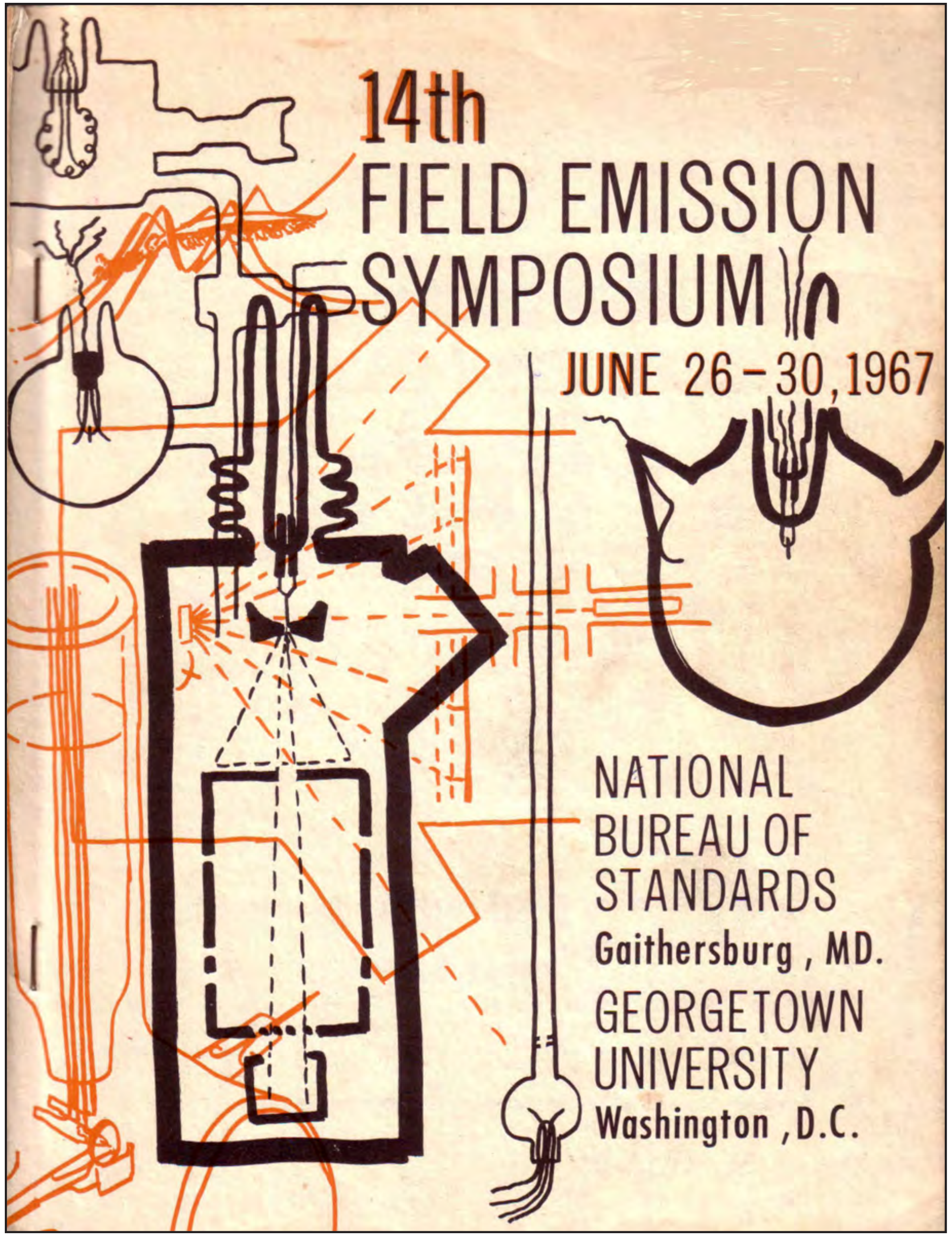

GAUGE
TO + D.C
PULSER
TRIGGE
GAS INLET
SCREEN
SCOPE
TO
PUMP
PREAMP
TO
PUMP
DETECTOR

And then there's the story of my glassblowing and Erwin's personality





In 1967 Erwin and I introduced the Atom-Probe Field Ion Microscope



The Atom-Probe Field Ion Microscope

Erwin W. Müller and John Panitz

Physics Department, The Pennsylvania State University

University Park, Pa.

A serious limitation of the field ion microscope has been its inability to identify individually imaged atoms. A newly conceived Atom-Probe FIM, consisting of a combination probe hole FIM and mass spectrometer having single particle sensitivity, will be described. During operation, the observer selects an atomic site of interest and places it over the probe hole. Pulsed field evaporation sends the chosen particle through the hole and into the spectrometer section which may be of the magnetic sector or time-of-flight type. Each has its own special advantages depending upon the particular application. These might include: identification of bright atom spots in the controversial adsorption experiments, investigation of the atomic nature of impurity and interstitial atom spots, analysis of segregations and precipitations, or the investigation of short range order in alloys. In some cases an adjustable probe hole covering an area of several atomic sites is advantageous. Experiences with a prototype time-of-flight instrument will be reported.

In 1968 the first paper was published but what about Gerry Fowler?

THE REVIEW OF SCIENTIFIC INSTRUMENTS VOLUME 39, NUMBER 1 JANUARY 1968

The Atom-Probe Field Ion Microscope*†

Erwin W. Müller, John A. Panitz, and S. Brooks McLane
Department of Physics, The Pennsylvania State University, University Park, Pennsylvania 16802

(Received 1 September 1967)

A serious limitation of the field ion microscope has been its inability to identify the chemical nature of the individually imaged atoms. The newly conceived atom-probe FIM is a combination probe-hole FIM and mass spectrometer having single particle sensitivity. During observation, the observer selects an atomic site of interest by placing it over a probe hole in the image screen. Pulsed field evaporation sends the chosen particle through the hole and into the spectrometer section. Preliminary results show that field evaporation of tungsten under poor vacuum conditions occurs as triply or quadruply charged WO, WN, WO_2, and WN_2 ions, while under better conditions doubly and possibly triply charged tungsten can be observed. Mo-Re alloys always produced doubly charged molybdenum and rhenium ions when examined in the atom-probe. Wide applications for the study of short range order in alloys, the chemical nature of precipitates and impurity atoms, and information regarding the imaging properties of various atom species, of both the substrate and adsorbed material, are foreseen.

I. INTRODUCTION

THE field ion microscope[1,2] provides an intimate view of the location of individual atoms on a metal surface. However, its inability to discriminate between different atomic species has been a serious shortcoming. While it is very certain that only one kind of atom is present on pure W, Ir, or Pt surfaces characterized by a highly perfect FIM pattern, it is not possible to unambiguously identify the different atomic species in the usually less regular FIM patterns which are obtained from alloys, crystals containing impurities, or adsorbates at the surface.

It is quite clear that a basic advance in field ion microscopy would be achieved with the identification of the atomic species associated with individual image dots. This difficult task now appears possible with a combination field ion microscope and mass spectrometer having single particle detection capability. Such a device may be called an atom-probe FIM in analogy with the well known electron microprobe developed by Castaing.[3] The latter instrument, a combination electron microscope and x-ray analyzer, can investigate the constituents of a small section of specimen which, although typically of the order of 1 μ in size, still contains some 10^{11} atoms. In the scanning electron microscope developed by Crewe[4] a very crude form of analysis is believed to be possible from the specific energy losses, but the number of atoms involved is still of the order of 10^5.

In the atom-probe FIM we wish to determine the nature of one single atom seen on a metal surface and selected from neighboring atoms at the discretion of the observer.

II. PRINCIPLE

The atom-probe consists of a field ion microscope modified so that the imaged atom chosen for analysis can be positioned over a probe hole in its fluorescent screen. This hole provides an entrance into the analyzer which in the present design is a time-of-flight spectrometer incorporating a detector having single particle sensitivity. After properly positioning the desired atom and pumping the imaging gas from the microscope, a high voltage pulse applied to the tip desorbs the atomic species which then travels through the probe hole. This pulse also initiates the horizontal sweep of an oscilloscope. When the ion reaches the detector a voltage pulse is fed to the oscilloscope, and thus the time-of-flight of the ion is determined. From these data its mass-to-charge ratio can be calculated since the ion acquires essentially its final velocity within a few tip radii, and since the potential difference through which it travels can be accurately measured. The desorption voltage V is, approximately, this potential difference, which is just the sum of the dc imaging voltage and the maximum pulse voltage, since the rise time of the pulse is negligible compared to the time of flight of the ion, and since the pulse amplitude is much less than the imaging voltage.

The mass-to-charge ratio of the detected ion is calculated from the relations

$$\tfrac{1}{2}mv^2 = neV \quad \text{and} \quad v = d/t,$$

where m is the mass of the ion, v is its velocity, ne is its charge, d is the distance between tip and detector, and t is the observed time of flight. One finds, for a tip-to-detector distance of 82 cm, that

$$m/n = 0.288Vt^2, \tag{1}$$

where m is measured in amu, V is in kilovolts, and t is in microseconds.

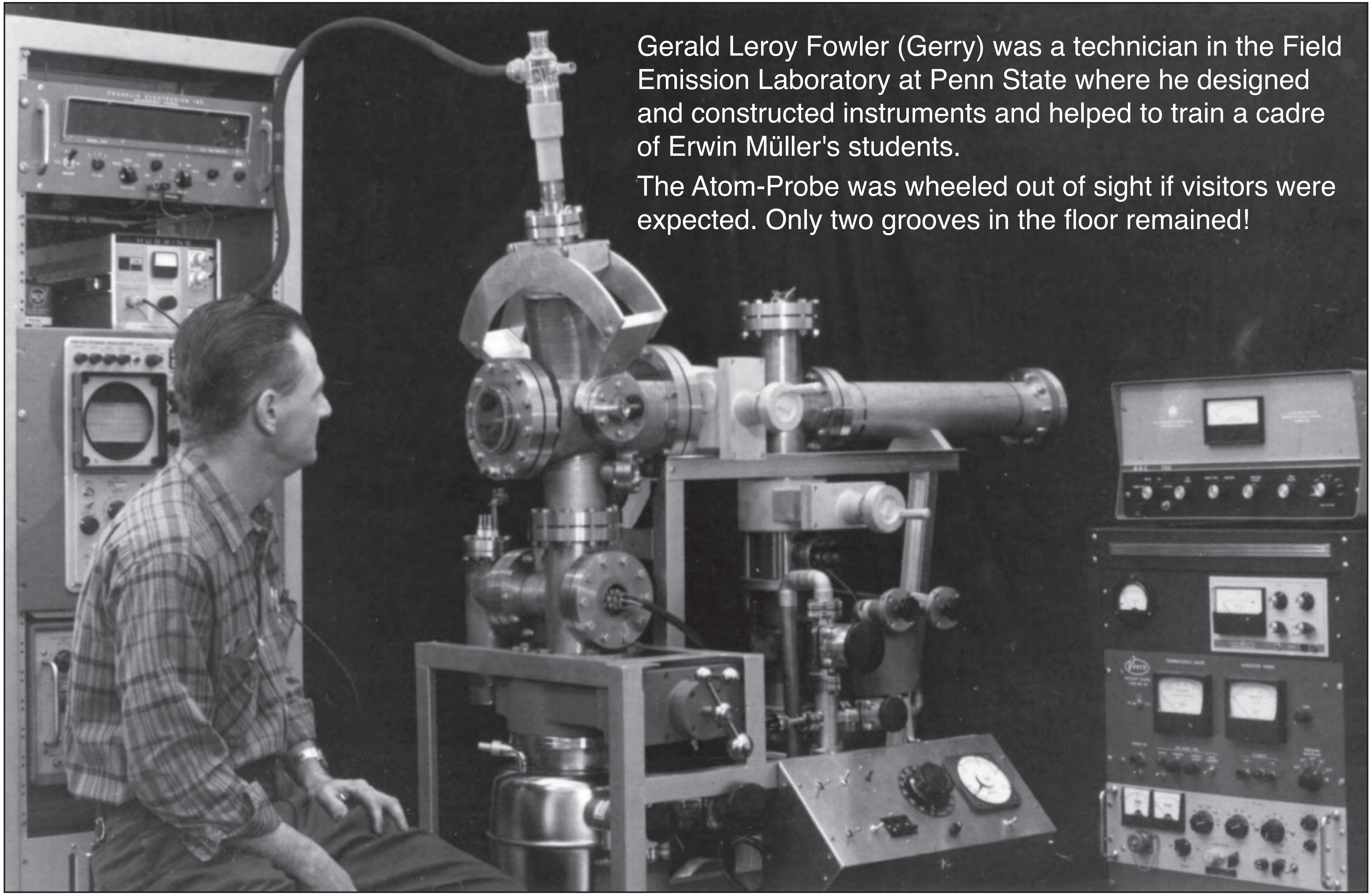
* Supported by the National Science Foundation.
† This paper was presented previously at the 14th Field Emission Symposium at the NBS, Washington, D. C. (June 1967).

[1] E. W. Müller, "Field Ionization and Field Ion Microscopy," in *Advances in Electronics and Electron Physics* (Academic Press Inc., New York, 1960), Vol. 13, pp. 83–179.
[2] E. W. Müller, Science **149**, 591 (1965).
[3] R. Castaing, "Electron Probe Microanalysis," in *Advances in Electronics and Electron Physics* (Academic Press Inc., New York, 1960), Vol. 13, pp. 317–384.
[4] A. V. Crewe, Science **154**, 729 (1966).

It is quite clear that a basic advance in field ion microscopy would be achieved with the identification of the atomic species associated with individual image dots. This difficult task now appears possible with a combination field ion microscope and mass spectrometer having single particle detection capability. Such a device may be called an atom-probe FIM in analogy with the well known electron microprobe developed by Castaing.

In the atom-probe FIM we wish to determine the nature of one single atom seen on a metal surface and selected from neighboring atoms at the discretion of the observer.

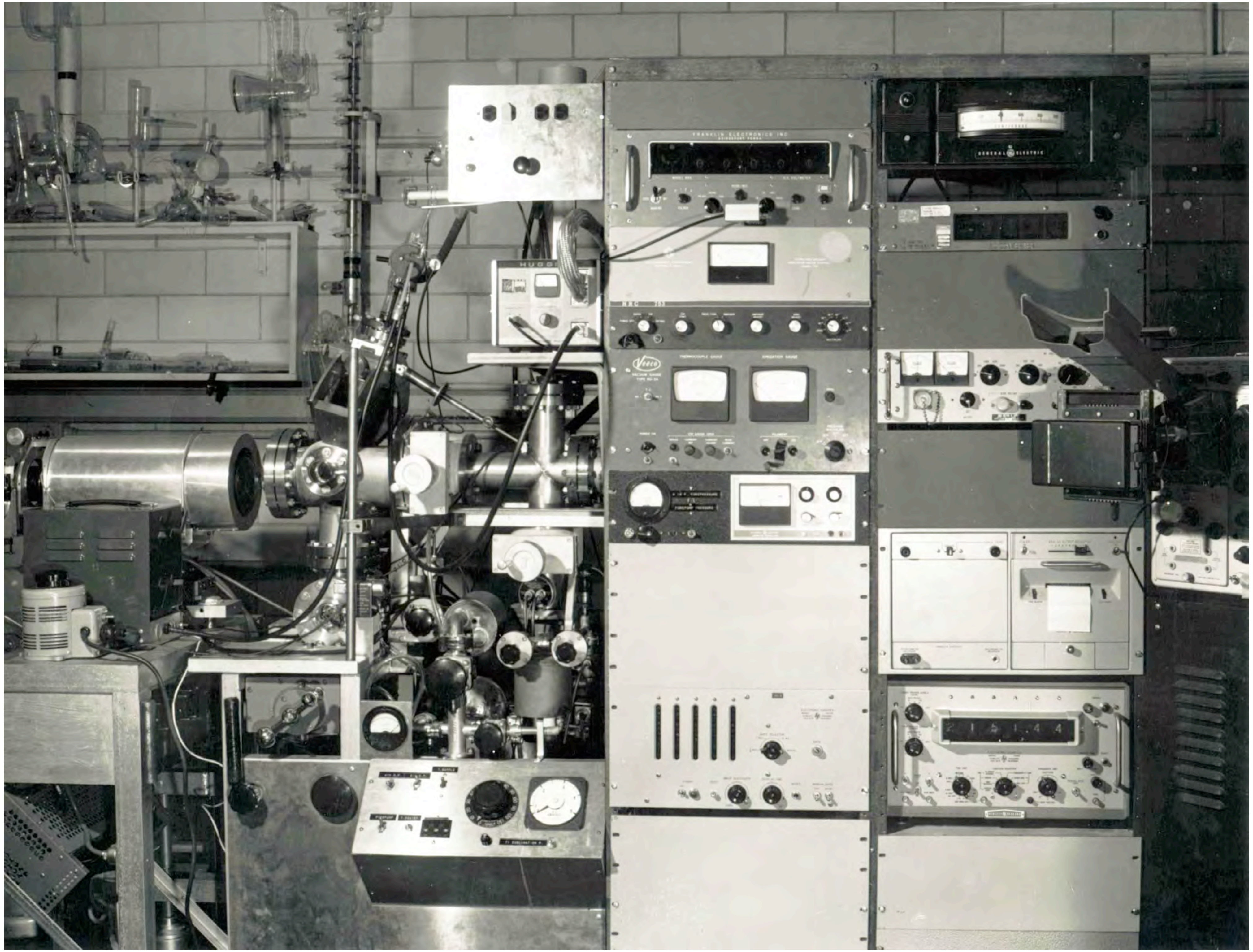
In 1968 Gerry Fowler constructed the second Atom-Probe



Gerald Leroy Fowler (Gerry) was a technician in the Field Emission Laboratory at Penn State where he designed and constructed instruments and helped to train a cadre of Erwin Müller's students.

The Atom-Probe was wheeled out of sight if visitors were expected. Only two grooves in the floor remained!

Erwin, Brooks and I collected data with the second Atom-Probe each Friday

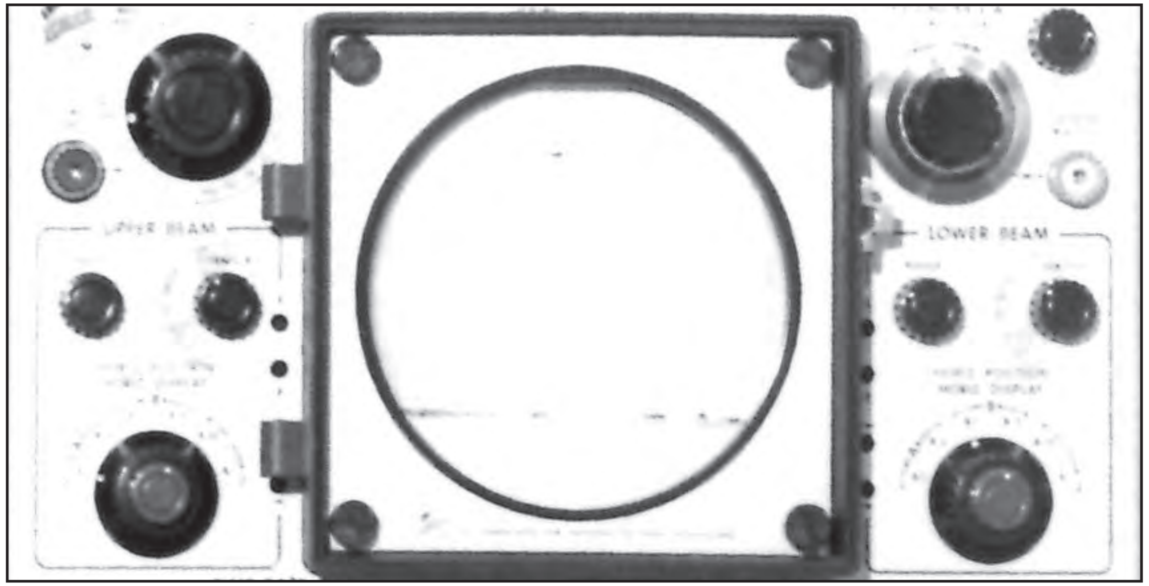


Essential Features

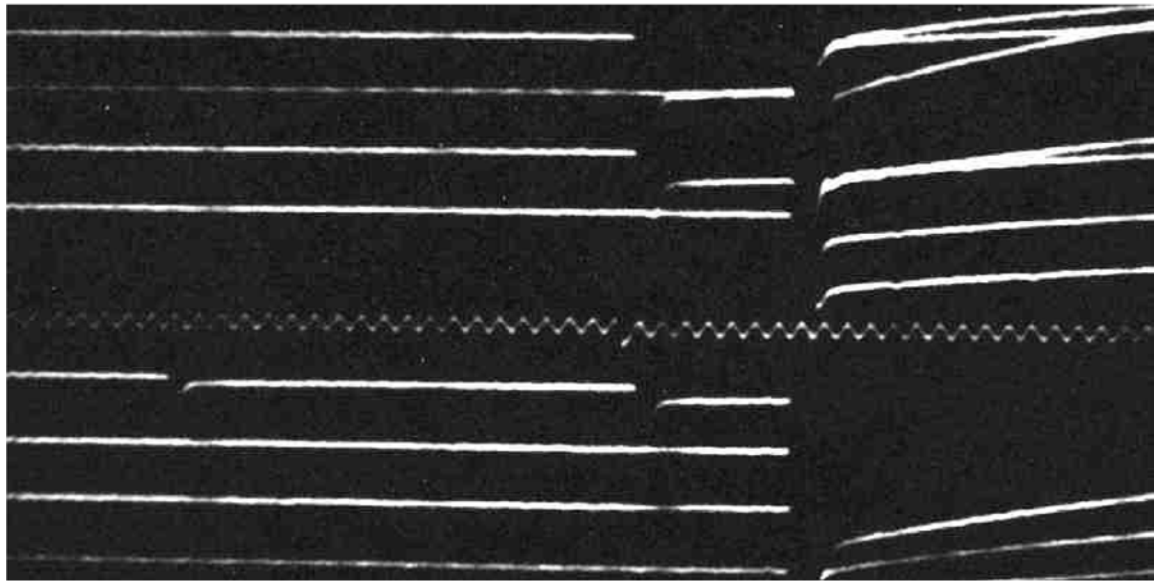
- An illuminated switch.
- A toggle switch.
- Knobs and dials.
- Always Available
- No wiring!

Useful Features

- Lightweight.
- Portable.
- Indestructible.

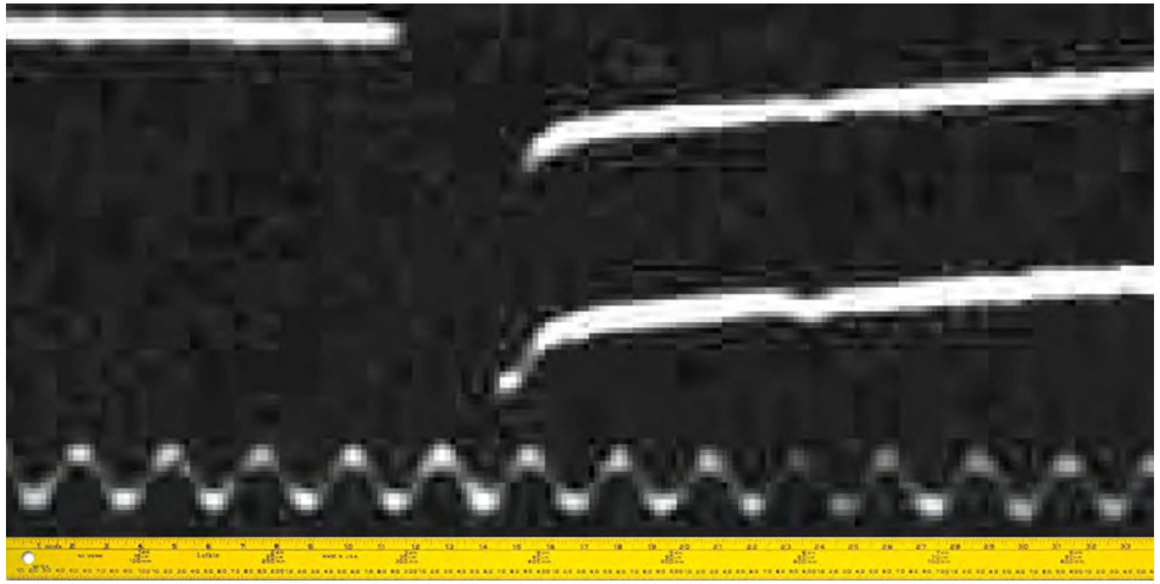


Record Ion Travel times on an Ocilloscope

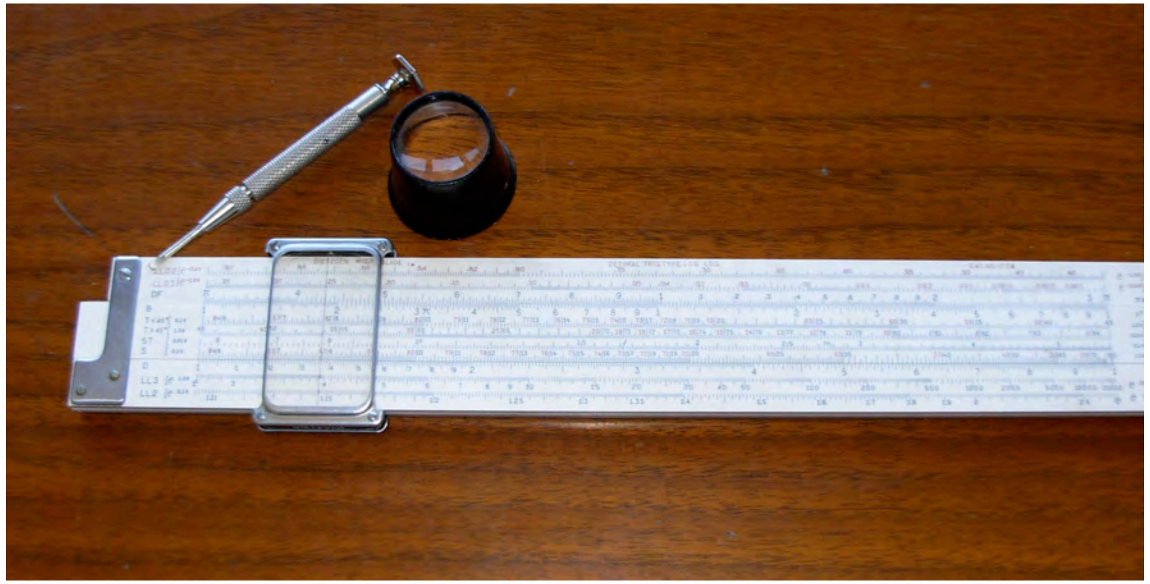


Take a Polaroid Print of the Oscilloscope Screen

Optically Magnify the Print



Use a Yard Stick to Find Travel Times from a Timing Trace



Calculate the Mass-to-Charge Ratio for Each Travel Time

Müller or My Data?

Erwin believed each event was significant but I used Penn State's IBM 360 to provide statistics

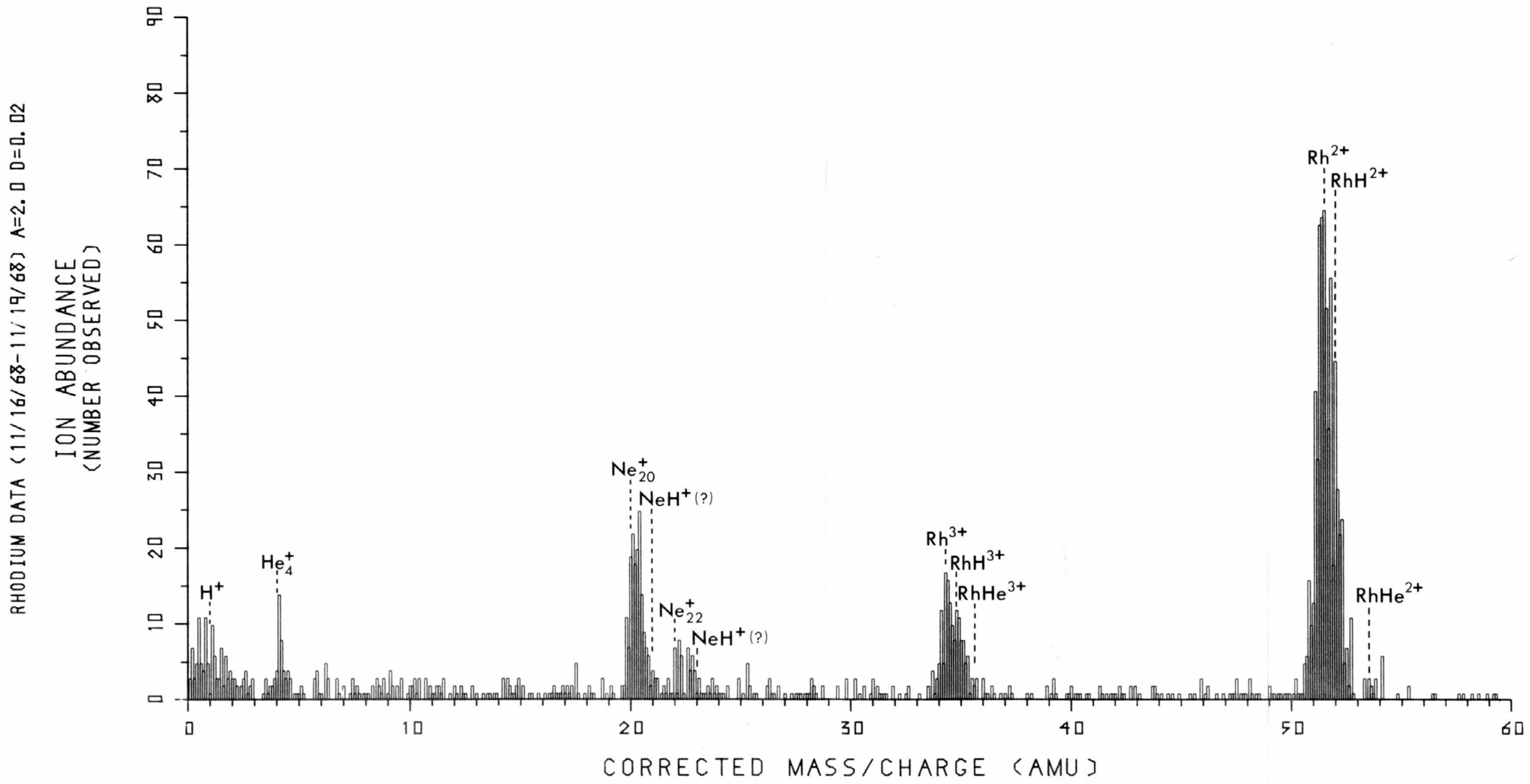
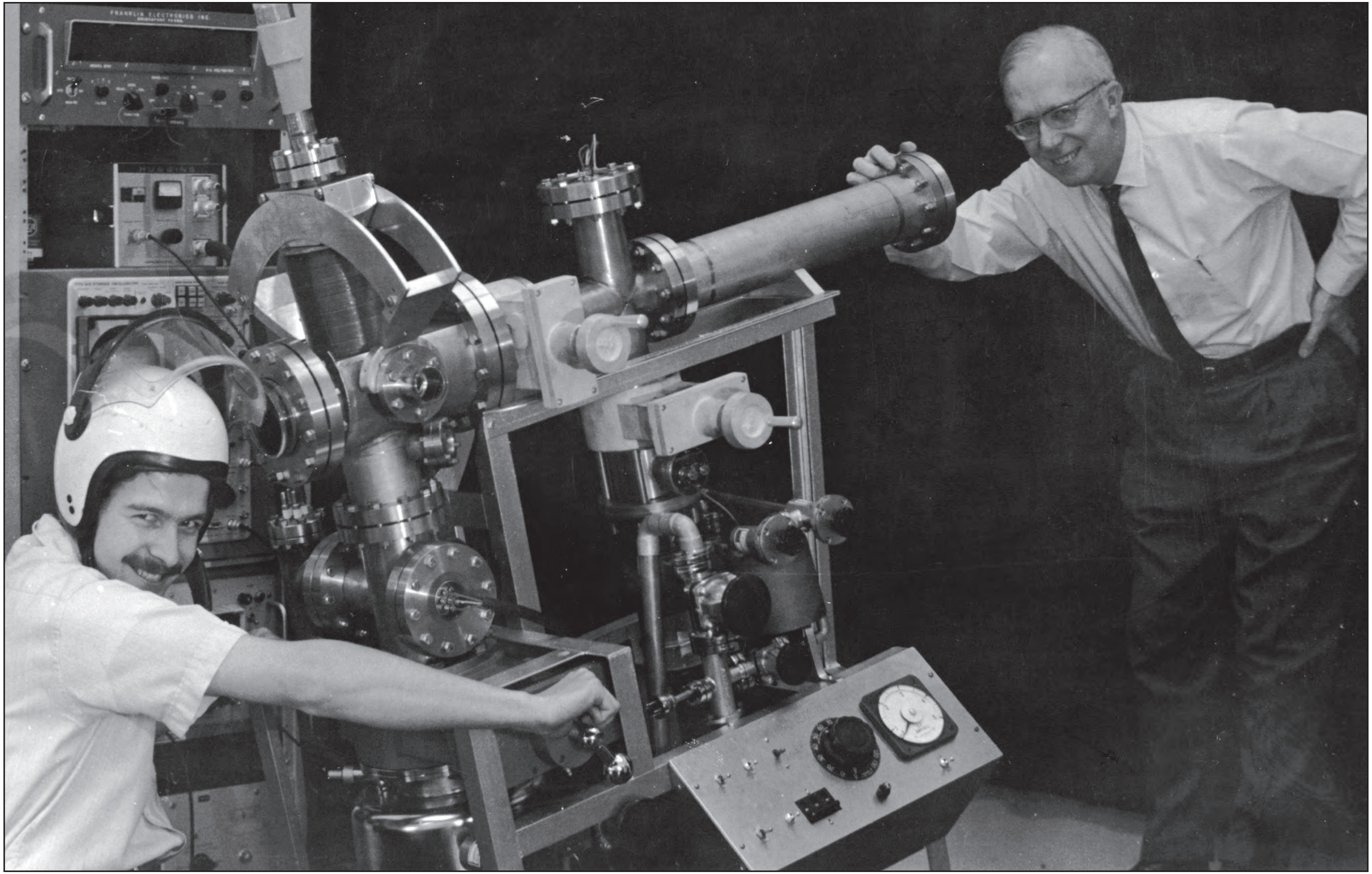
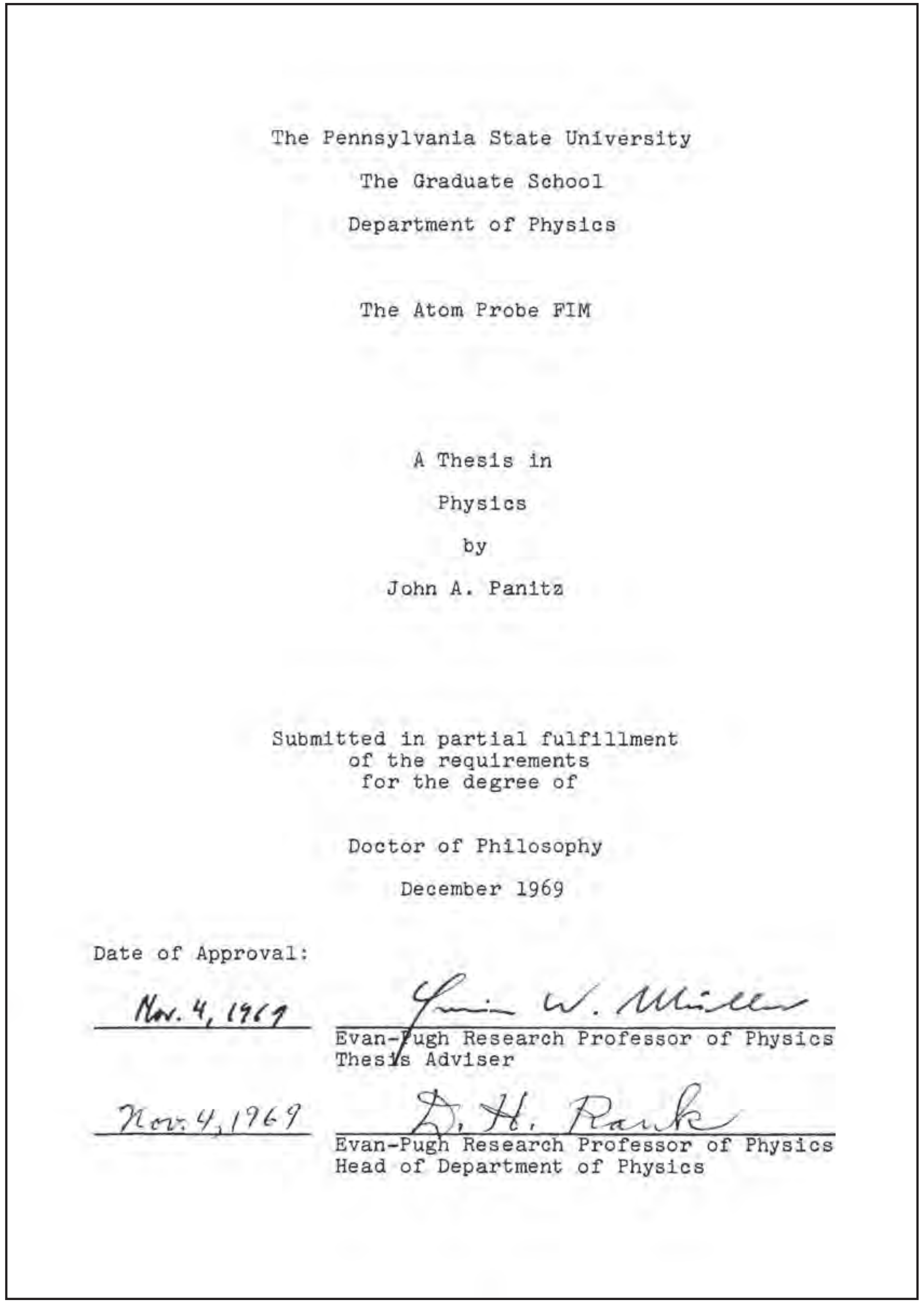


Figure 19. Abundance Histogram (α = 2.0, δ = 0.02)

In the end I saw myself differently



I graduated in 1969 and described an "Aiming Error"


The Pennsylvania State University

The Graduate School

Department of Physics

The Atom Probe FIM

A Thesis in

Physics

by

John A. Panitz

Submitted in partial fulfillment
of the requirements
for the degree of

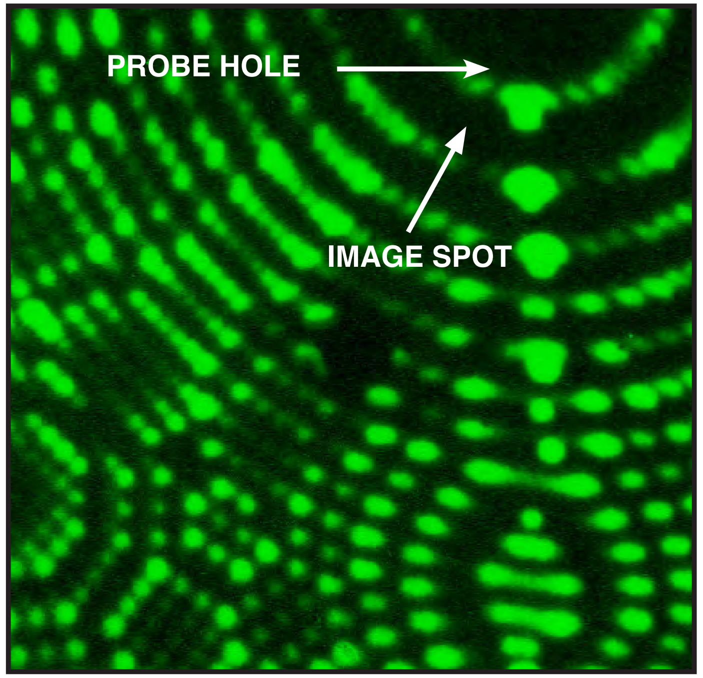
Doctor of Philosophy

December 1969

Date of Approval:

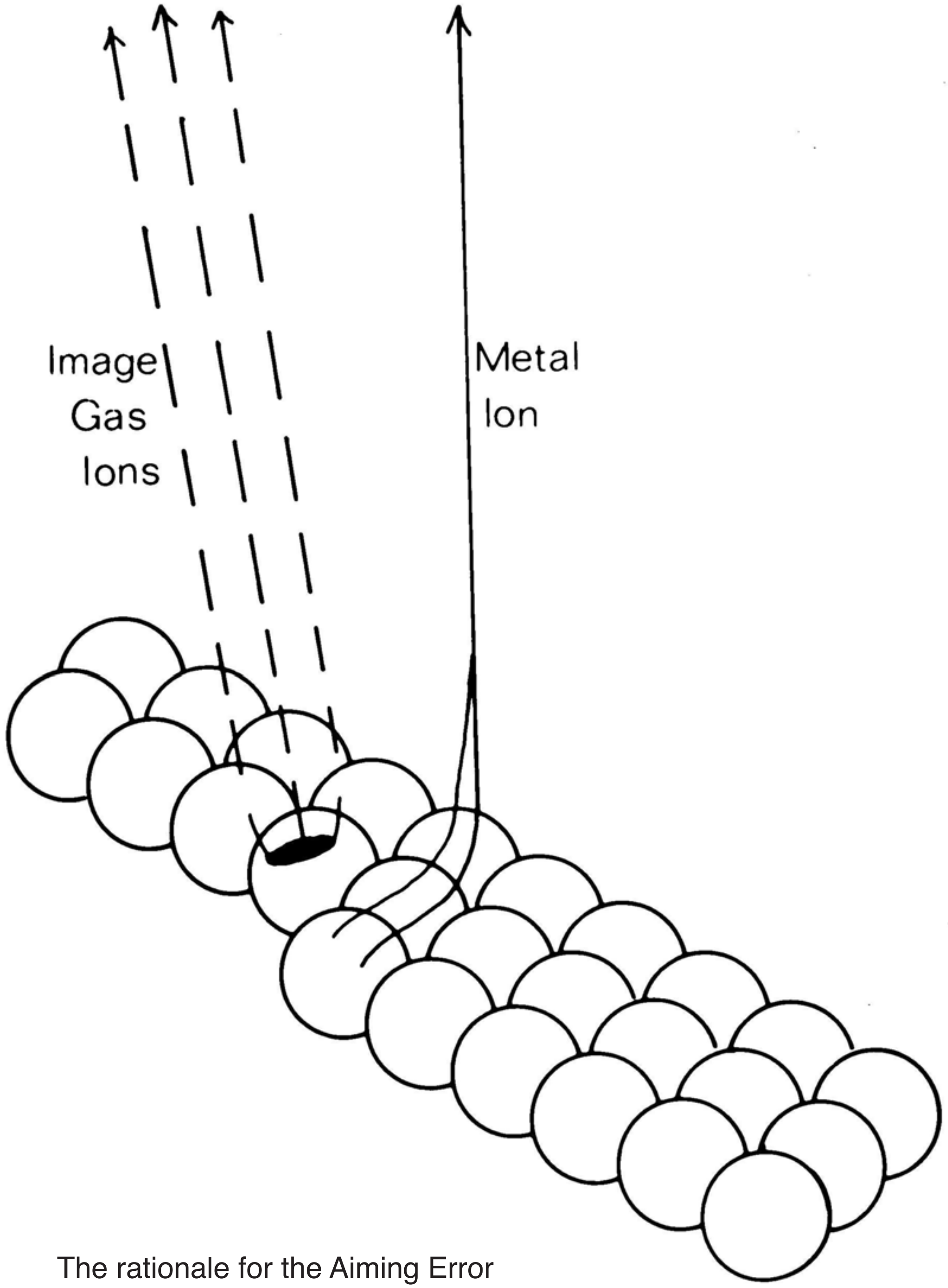
Nov. 4, 1969 — Evan-Pugh Research Professor of Physics
Thesis Adviser

Nov. 4, 1969 — Evan-Pugh Research Professor of Physics
Head of Department of Physics

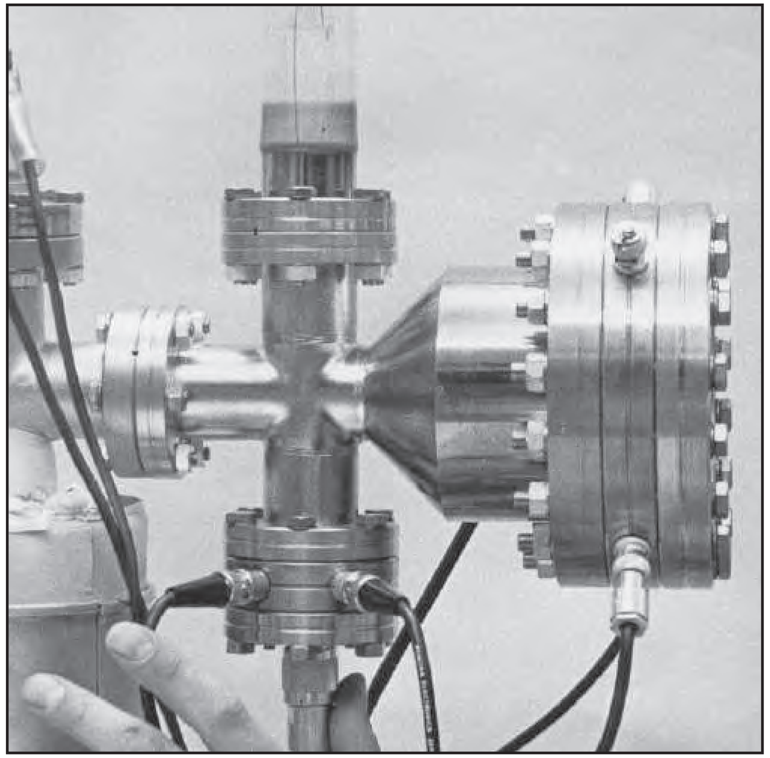


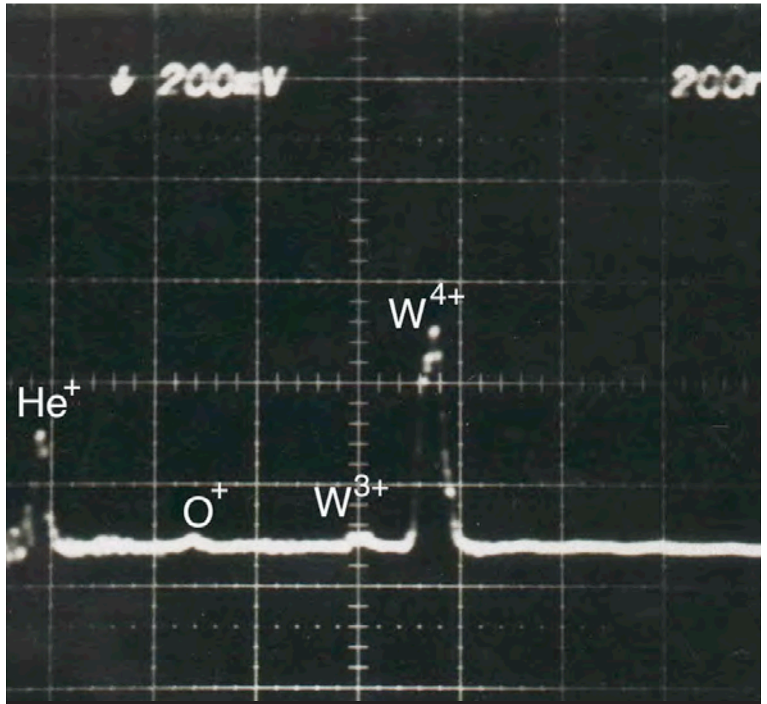
"If an image spot is placed over the probe hole, and a few atoms of the net plane edge are evaporated, no metal ion is ever detected However, if the probe hole is placed approximately one image spot diameter from the selected image spot toward the center of the plane, a metal ion is recorded during essentially every pulse."

As a result one could not "determine the nature of one single atom seen on a metal surface and selected from neighboring atoms at the discretion of the observer".

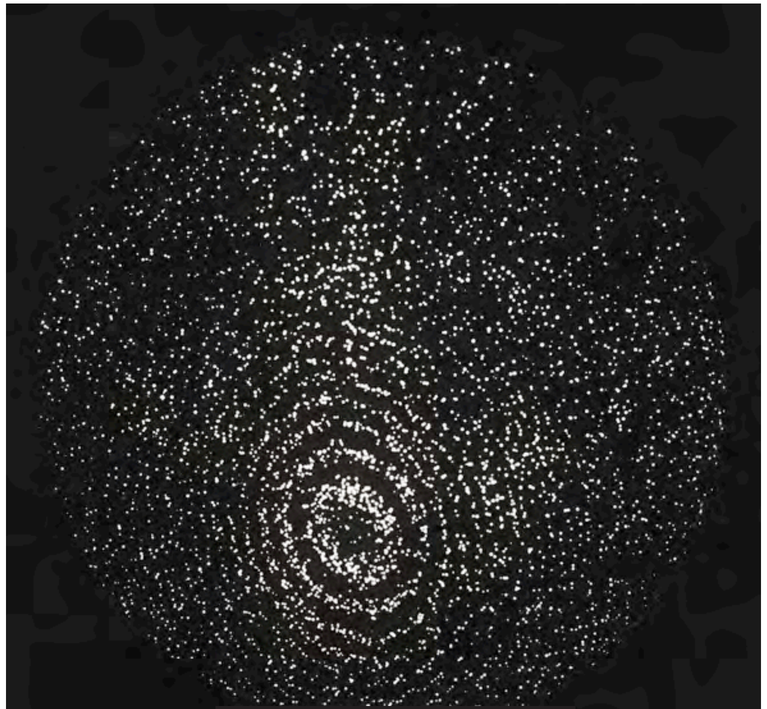


The rationale for the Aiming Error

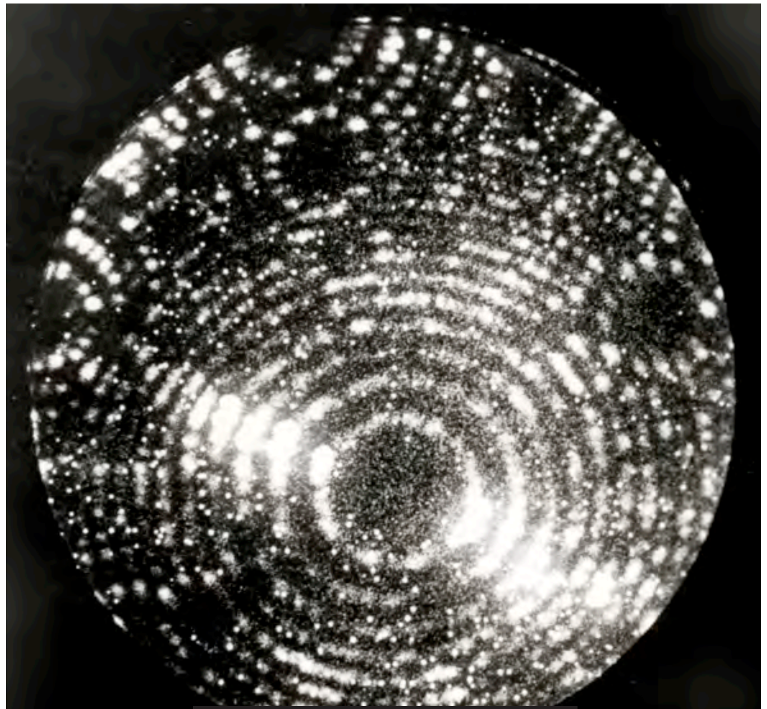




Mass Analysis



Time-Gating



The Aiming Error with W^{3+}

United States Patent [19] [11] **3,868,507**

Panitz [45] **Feb. 25, 1975**

[54] **FIELD DESORPTION SPECTROMETER**

[75] Inventor: **John A. Panitz**, Edgewood, N. Mex.

[73] Assignee: **The United States of America as represented by the United States Atomic Energy Commission,** Washington, D.C.

[22] Filed: **Dec. 5, 1973**

[21] Appl. No.: **422,048**

[52] **U.S. Cl.** **250/287,** 250/227, 250/306

[51] **Int. Cl.** **H01j 39/34**

[58] **Field of Search** 250/286, 287, 306, 307, 250/309

[56] **References Cited**

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3,141,105	7/1964	Courtney-Pratt	250/306
3,240,927	3/1966	Fite et al.	250/287
3,582,649	6/1971	Taylor	250/306

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The Atom Probe Field Ion Microscope by E. W. Muller from Naturwissenschaften, Vol. 57, No. 5, May, 1970, pp. 222–230. 250–309.

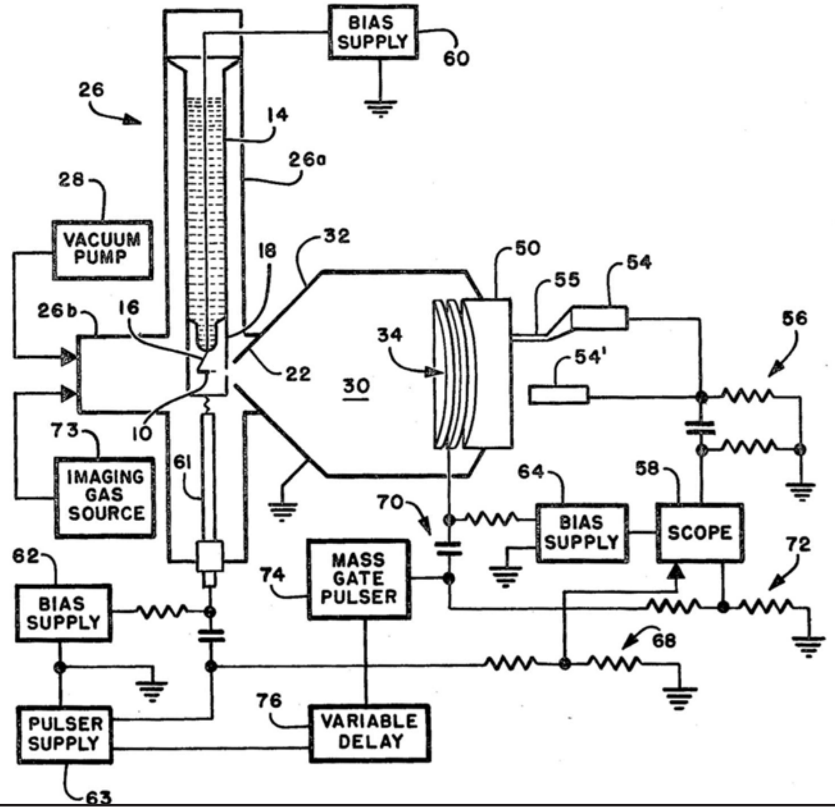
"Construction and Performance of an FIM-Atom Probe" by S. S. Brenner et al. from "Surface Science," North-Holland Publishing Co., Vol. 23, No. 1, 1970, pages 88–111.

Primary Examiner—James W. Lawrence
Assistant Examiner—C. E. Church
Attorney, Agent, or Firm—John A. Horan; Dudley W. King; Richard E. Constant

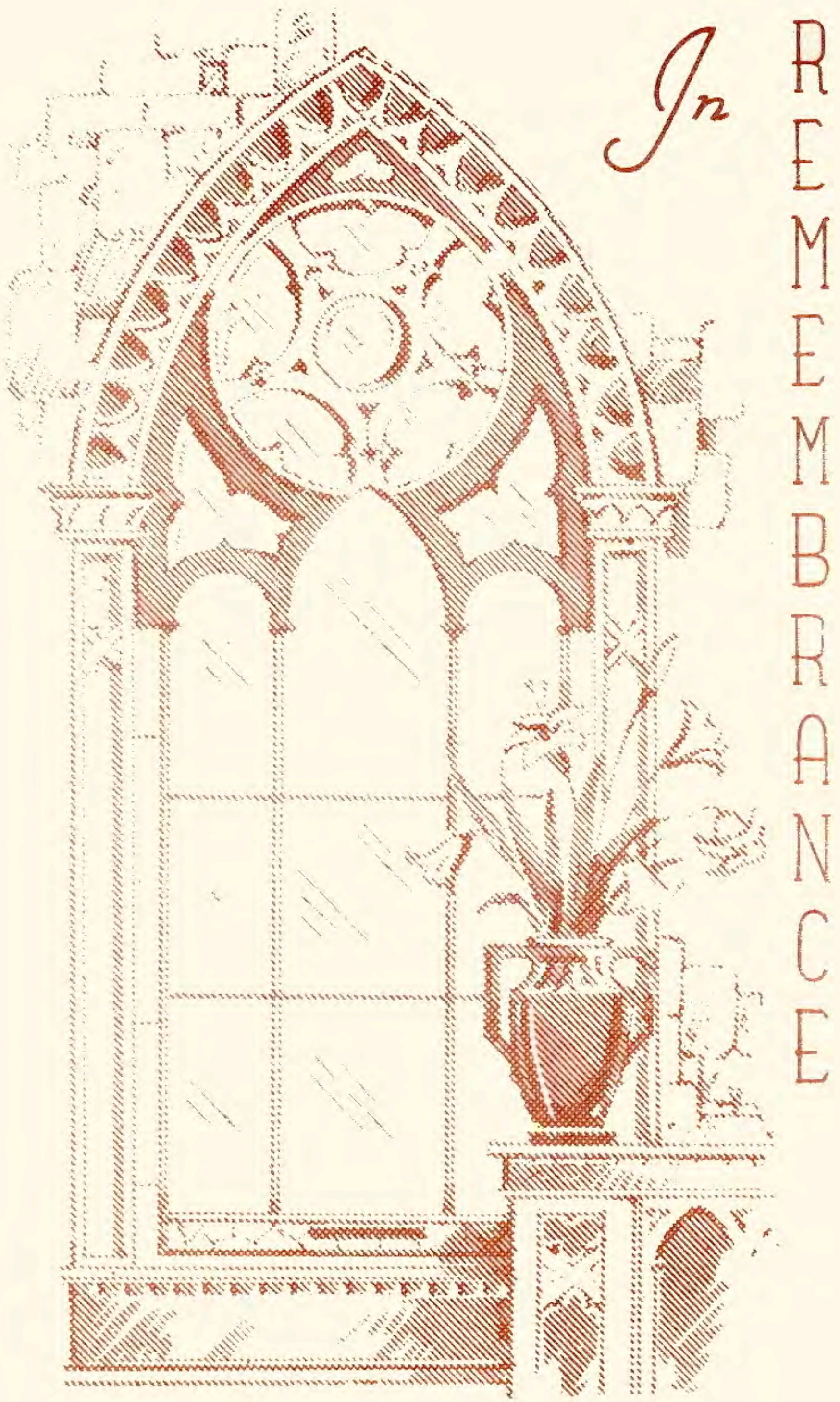
[57] **ABSTRACT**

A field desorption spectrometer which is capable of detecting and identifying one or more atoms of a specimen and/or all the atoms in an outer layer of the specimen or throughout the bulk of the specimen may comprise an apertured electrode for applying an electric field to field evaporate or field desorb ions from the specimen through the aperture, an apertured wall for blocking electric fields from the apertured electrode and specimen from a field free drift region and for transmission of the desorbed ions into the drift region, channel electron multiplier array means positioned in the drift region for intercepting of substantially all of said ions and for providing amplification of ion impacts by electron multiplication at locations corresponding with the locations where ions strike the array means, and means for sensing the multiplied electrons corresponding to these locations.

12 Claims, 8 Drawing Figures



In 1977 Erwin Wilhelm Müller died during a meeting of the National Academy of Science



May 20, 1977

Services for

Erwin W. Mueller

Born

Berlin, Germany
July 11, 1911

Died

Washington, D. C.
May 17, 1977

Services Held at

St. Andrew's Episcopal Church
1:30 p.m.

Minister

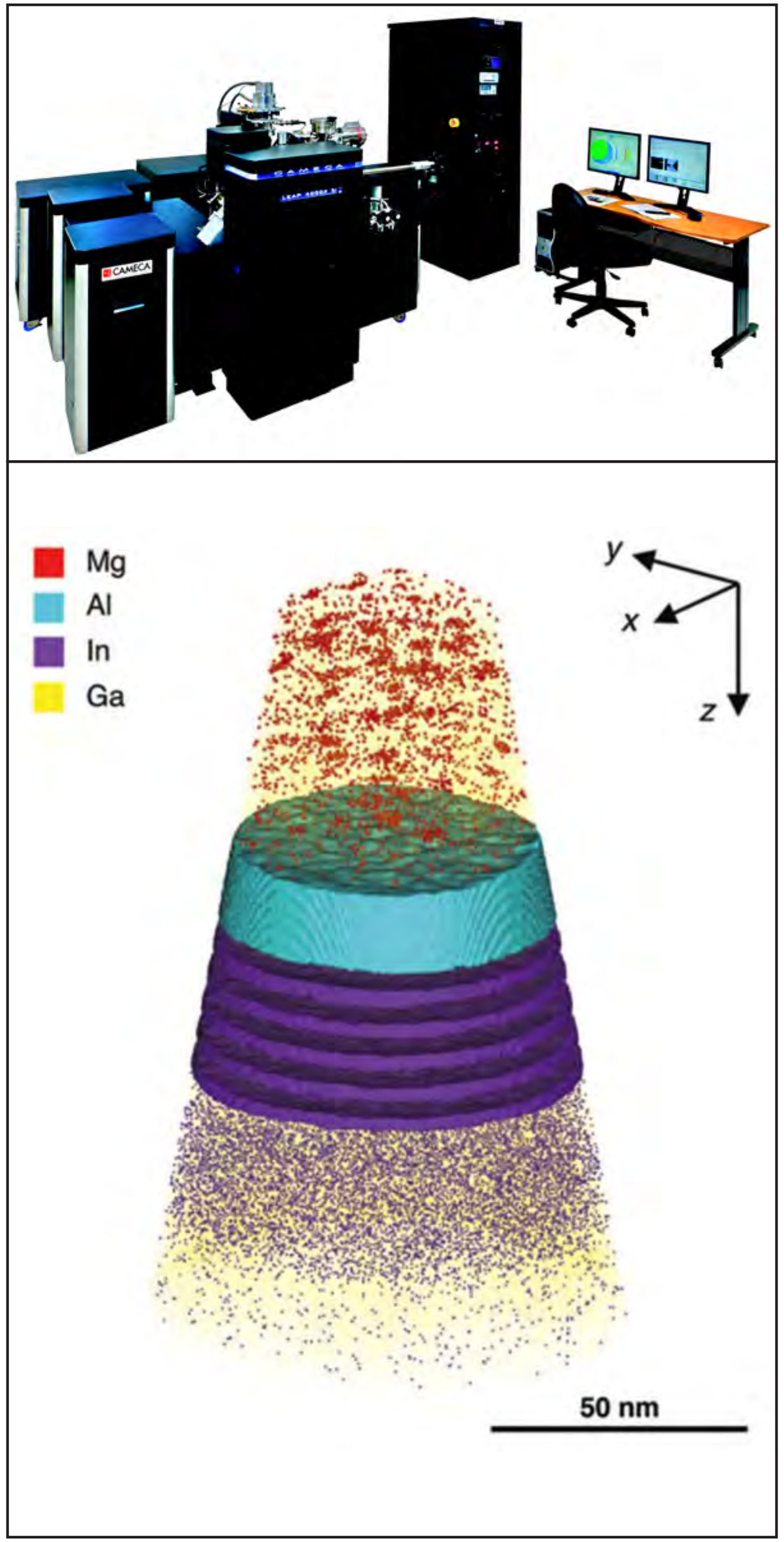
Rev. Arthur C. Parker

Interment

Centre County Memorial Park
State College, Pa.

Erwin once remarked "Here lies a great idea in a rotten brain"

Had Erwin envisioned the modern Atom-Probe?



In 1998 Thomas F. Kelly founded Imago Scientific Instruments. Using modern data aquisition technology with high voltage and picosecond laser pulsing, Imago created the Local Electrode Atom Probe (LEAP). It featured a position sensitive detector and a microextraction counter electrode and could acquire and display a mass resolved 3D tomographic image of metals, semiconductors and insulators in real time. The first instrument was shipped in 2003 to Oak Ridge National Laboratory. In 2010 Imago was acquired by Ametek, Inc and merged with its subsidiary Cameca SAS. A Cameca Atom-Probe Tomographic image and the instrument used to produce it is shown at the left.[1]

1. A. D. Giddings, et. al. *Reverse Engineering at the Atomic Scale: Competitive Analysis of a Gallium Nitride-Based Commercial Light Emitting Diode.* Microscopy Today. 22 (2014) 12.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Abstract

The Atom-Probe Field Ion Microscope was introduced in 1967. It was, and remains, the only instrument capable of determining the chemical identity of one single atom seen on a metal surface and selected from neighboring atoms at the discretion of the observer. The development of the Atom-Probe is a story of an instrument that one NSF reviewer called “impossible because single atoms could not be detected”. It is also a story of my life with Erwin Wilhelm Müller as his graduate student in the Field Emission Laboratory at the Pennsylvania State University in the late 1960s and his strong and volatile personality that arguably cost him the Nobel Prize in Physics, a personality perhaps fostered by his pedigree as Gustav Hertz’s student in the Berlin of the 1930s. It is the story that has defined my scientific career.