

Atomic-Scale Analytical Tomography

A comprehensive guide on Atomic-Scale Analytical Tomography (ASAT), this title discusses basic concepts and implications of the technique in areas such as material sciences, microscopy, engineering sciences, and several interdisciplinary avenues. The title explores how to successfully achieve ASAT at the intersection of transmission electron microscopy and atom probe microscopy. This novel technique can identify individual atoms in large volumes as well as in 3-D, with high spatial resolution. Written by leading experts from academia and industry, this book serves as a guide with real-world applications in cutting-edge research problems. Essential reading for researchers, engineers, and practitioners interested in nanoscale characterization, this book introduces the reader to a new direction for atomic-scale microscopy.

Thomas F. Kelly is a pioneer in the field of atomic-scale research. He was a professor of Materials Science and Engineering at the University of Wisconsin-Madison for 18 years prior to founding Imago Scientific Instruments (later acquired by CAMECA) to commercialize his invention, the local electrode atom probe (LEAP). Currently, Dr. Kelly is founder and CEO of Steam Instruments, Inc., and is a fellow of the Microscopy Society of America, the International Field Emission Society, the Microanalysis Society, and the Korean Society for Microscopy. He was recently elected to the US National Academy of Engineering.

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Advances in Microscopy and Microanalysis

Microscopic visualization techniques range from atomic imaging to visualization of living cells at near nanometer spatial resolution, and advances in the field are fueled by developments in computation, image detection devices, labeling, and sample preparation strategies. Microscopy has proven to be one of the most attractive and progressive research tools available to the scientific community and remains at the forefront of research in many disciplines, from nanotechnology to live cell molecular imaging. This series reflects the diverse role of microscopy, defining it as any method of imaging objects of micrometer scale or less, and includes both introductory texts and highly technical and focused monographs for researchers and practitioners in materials and the life sciences.

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Atomic-Scale Analytical Tomography

Concepts and Implications

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To Kanwar Bahadur and Erwin W. Müller, the first to image atoms.

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Foreword

The authors of this book are in a unique position to create this volume. In 2002, I received a phone call from a venture capital firm in California that wanted to know if a company called Imago, founded by Thomas F. Kelly, was a good investment. Imago wanted to manufacture a commercial atom probe. I explained that based on my experience there were many attempts to commercialize the atom probe over the past 35 years, but none had been particularly successful, so I advised against funding them. But Imago got their funding, produced and sold the first Local Electrode Atom Probe (LEAP), was purchased by the CAMECA division of Ametek in 2010, and the rest, as they say, is history.

I have known the first author of this book for many years. Besides being a forthright consummate entrepreneur, Tom has always been concerned with the history of the instruments he manufactured, and the LEAP was no exception. Its origin can be traced to the Atom-Probe Field Ion Microscope (APFIM), whose predecessor, the low-temperature Field Ion Microscope (FIM), was introduced in 1956 by Erwin Wilhelm Müller. With it he became the first person to “see” an atom on the apex of a sharply pointed needle-like metal substrate called a tip. In 1946, Erwin had discovered field desorption, a process that could now be seen to peel away the apex of the tip, layer by atomic layer, to expose the surfaces below. In 1967, Erwin suggested building the APFIM to determine the chemical identity of the atoms that were observed. In the APFIM, a tip is tilted to place the image of a single atom over a “probe hole” in the FIM’s fluorescent screen. When a nanosecond-long, high-voltage pulse was applied to the tip, the atom would field desorb as a positive ion and enter a mass spectrometer for analysis. Douglas Barofsky, Erwin’s postdoctoral student, suggested using a time-of-flight mass spectrometer, and the APFIM, with its ability to record surface species as a function of depth, became a reality. S. Sidney Brenner was the first metallurgist to design and build an APFIM to record the depth profile of species as a function of depth, determined by extracting the depth from a succession of field ion microscope images.

Today, we live in a different world where computers have replaced the individuals who recorded the painstaking and time-consuming process of imaging and analyzing individual atoms as a function of depth in the APFIM. But at what cost? Certainly, accuracy has been sacrificed for speed, because the current instrumental technique called “atom probe tomography” produces a map of species as a function of time but cannot determine the precise depth and lateral magnification, since FIM imaging is not

available. Instead, the depth and magnification must be calculated from a model of how the tip shape is expected to change with time. This limitation could be minimized if the atom probe were combined with a high-resolution electron microscope, an instrument currently under consideration. If successful, it would allow the change in tip shape to be determined as a function of time, and the rest *will* be history.

J. A. Panitz

Atomic-Scale Analytical Tomography (ASAT)

The ultimate microscopy would generate large images containing the location and identity of every atom in the sample, in three dimensions. The premise of this book is that this microscopy, Atomic-Scale Analytical Tomography (ASAT), will represent a profound scientific advance, and it is within reach. ASAT will require a dramatic jump in the resolving power of microscopy and in our technology and practice of data collection rate and data management. Today, the technologies underpinning transmission electron microscopy and atom probe microscopy are advancing rapidly, bringing us ever closer to ASAT; but the question remains: Could either of these approaches deliver this ultimate microscopy? This book explores the prospects for achieving true ASAT by *integrating* these seemingly disparate microscopy approaches. The authors present a road map for how this might be achieved and explore the scientific frontiers that will open with the advent of ASAT.

Preface

There are a lot of atoms in the universe, about 10^{80} . Our solar system has a mere 10^{55} atoms. What we seek in microscopy of materials is to gain knowledge of the microstructure down to the atomic scale. In this cosmic context, a three-dimensional dataset containing 1,000,000,000 (10^9) atoms is hardly impressive. It is really a very small amount of matter. But when viewed from the optics of history, it can be considered a massive accomplishment.

On the other end of the spatial range, an angstrom, or a hundred trillionths of a meter, is a difficult length scale for most people outside of the physical sciences to understand. It is approximately the distance between atoms in a solid. Measuring the identity and location of a single atom to better than an angstrom, again in three dimensions, might seem to be an act of folly or, at best, science fiction. However, for materials science, determining the identities and atomic locations of 1,000,000,000 atoms to that length scale, a technique we define as Atomic-Scale Analytical Tomography (ASAT), will soon be a necessity.

The kernels of ASAT are in the Imaging Atom Probe (IAP) and the Transmission Electron Microscope (TEM). We recognize that not all readers will have expertise in these subjects and do not feel it is the place of this work to completely educate readers on these individually remarkable techniques. We would recommend the excellent TEM texts of [1]–[4], among others. We would also recommend the texts of [5]–[11] for a thorough background on Atom Probe Tomography (APT). Our view remains that additional texts focused exclusively on the contemporary developments and applications of either TEM or atom probes, individually, will continue to have merit as these techniques continue to rapidly evolve. However, our particular interest and the focus of this book is to look at the intersections between the TEM and atom probes. Once we scratch away at this notion, we see that there are many opportunities for advances in materials science and many scientifically interesting and technologically important challenges for microscopy. We hope that this book helps to propel this discussion space. Our approach is to look at the motivations for coupling these techniques, the instrumentation challenges, the nature of these new experiments, and the many and varied issues arising from integration of the data: which data to correlate, which to integrate, and how the results might be used to do new materials science.

When John Panitz, working with Erwin Müller at the Pennsylvania State University, recorded the first atom probe data in 1967 [12], he manually recorded an oscilloscope trace on Polaroid™ film of the start pulse and impact pulse of tungsten atoms flying

down a 1-meter tube. He was able to record about 100 such events in one night's work. Electronic recording of events soon followed, as did computer control of the process [13]. By the early 1970s, home-built atom probes were recording one-dimensional data streams of atoms at a rate of 10^3 atoms per hour. An overnight run would yield 10^4 atoms. Panitz soon developed the Imaging Atom Probe (IAP, originally called the 10-cm Atom Probe) in 1973 ([14], [15]), which is the precursor of today's 3-Dimensional Atom Probes (3DAP).

Panitz did not have access to any technology that could record the position and identity of arriving atoms on a detector. We believe that it was just a matter of time, perhaps one development at a time, before someone built on what had been established in the atom probe and imaging atom probe and took it to atomic-scale tomography (the position and identity recorded for every atom in a large volume) and Atomic-Scale Analytical Tomography (AST with analytical information added).

In 1989, the first fully three-dimensional atom probe, the Position-Sensitive Atom Probe (PoSAP), was designed by Alfred Cerezo, Terrance Godfrey, and George Smith at the University of Oxford [16]. For the first time, the position and identity of different atoms in real space in three dimensions could be studied. The field soon adopted the term Atom Probe Tomography (APT) for this type of work. Though the invention of the PoSAP was a seminal moment for us and the world and was a spectacular breakthrough, we also came to understand its limitations. The technique was constrained by speed (1 atom/second) and field of view (15 nm diameter by 50 nm deep). One million atoms took over a week to collect. For real-world materials studies, 1,000,000 atoms really are not a lot of matter. And so, the stage was set for the next development.

One of the authors (TFK), upon learning about the PoSAP, immediately started thinking of ways to speed up the technique and increase the field of view. The technique needed a concomitant increase in field of view with increase in data collection rate; otherwise, it would be limited to datasets from long, skinny volumes. He sought a means to break APT out of its limited niche and make it a broadly applied technique. Experts in the field were skeptical at the time that atom probe specimens could be evaporated at the rates proposed. However, the Local Electrode Atom Probe (LEAP) [17]–[19] succeeded in this mission. Finally, at this point it was possible to collect data for a three-dimensional image at a high rate, 10^6 atoms/minute, which meant we could routinely create 100,000,000-atom (10^8) images, and 1,000,000,000-atom (10^9) images were within reach. When the LEAP became commercially available in about 2003, a 1,000,000,000-atom image was considered very large, maybe unachievable. In the present day, though it is not common, 1,000,000,000-atom images have been recorded from single specimens. In 1967, this achievement would have been unthinkable.

The state of development of atom probe tomography is such that we now seek, not merely to increase speed and field of view, but to fix its two main limitations: (1) less than 100% of the atoms in the specimen are detected, and (2) aberrations in the image reconstruction process cause distortions and limit spatial resolution. In this book, we spell out the problem in detail, look at the history of how we got to this juncture, and

consider what it will take to overcome these two limitations. We are optimistic that these limitations will be conquered. Indeed, currently there are experimental programs in progress around the world to address them. For instance, Gorman's group at the Colorado School of Mines has several funded programs focused on building an APT in the objective lens of a TEM with the goal of correlating atom positions from the TEM to the atomic masses measured using the APT. We expect these programs to come to fruition in the next few years.

The largest effort is a recently (2019) funded program at the Ernst Ruska Center of the Forschungszentrum Jülich in Germany called Project Tomo. This project will integrate an atom probe (LEAP™ CAMECA Instruments, Inc.) into the objective lens pole piece of an aberration-corrected Transmission Electron Microscope (TEM, ThermoFisher Scientific) such that the field-evaporated ions fly toward a Superconducting Delay-Line (SCDL™, Steam Instruments, Inc.) detector. Note that, at press time, the SCDL detector is not funded on the project. The TEM will be used to determine the specimen apex shape to enable precise computation of ion trajectories in a reconstruction. The SCDL detector is expected to deliver 100% detection efficiency for the ions. With these two limitations addressed, the stage is set for achieving atomic-scale tomography. We recognize that this combination of TEM and APT also offers crucial information about the electric fields around the specimen and analytical information, hence the more general name, atomic-scale analytical tomography.

So, assuming the ASAT projects are successful, are we finished? Is atomic-scale analytical tomography complete? One question we ponder is, will 1,000,000,000-atom images be enough? Even if they aren't, is this figure a sweet spot for data size anyway? In terms of the microstructure of a material, 1,000,000,000 atoms still does not represent a large amount of matter. After all, a typical field of view in a transmission electron microscope image at 10^5 times magnification is the averaged product of the scattering from 10^{10} atoms. In this book, we will speak to the possibility of acquiring images from atom probe tomography where the particular (not the average) atomic position and neighborhood information are available from trillion-atom datasets. We envision that it will be possible in the next decade or two to approach this result. You are probably wondering, do we need a trillion atoms? For that matter, will the specimens withstand evaporation at the rates needed (1,000,000,000 atoms per minute) to reach a trillion atoms in a modest amount of time (one day)? What will we do with the information if we get it? In Chapter 10 we discuss the potential for data mining to be a major tool for microstructural analysis if data are available. Data mining will need trillion-atom images if it is to deliver new information. And how will we store that much information? We note simply that Google today has exabytes (10^{19} bytes) of storage. A trillion atoms at 23 bytes/atom would require about 23 terabytes ($\sim 23 \times 10^{13}$ bytes) of storage. In 2020, one can build a local storage capacity of 1 petabyte (10^{15} bytes) for about \$50,000. Clearly, it is a bit expensive; but by the time this kind of storage capacity is a real need, expense likely will not be a problem. We hope to chuckle at this question as we head toward 2050.

Is this the stuff of science fiction? Hardly. It is quite the opposite. Larger datasets will deliver more information of relevance to the real world. Admittedly, visualization systems and atomistic simulations are not going to be able to handle these large numbers of atoms any time soon. New strategies for working in large datasets will be needed. For example, analysts already work with sub-volumes of an image. Tools can be built to search for and find relevant sub-volumes for a particular question. Machine learning has already been implemented to find and analyze microstructural features in TEM images, for example. As mentioned above, data mining may well be the key to unlocking the large amount of information contained in such datasets.

In the history of humanity, a constant theme is that better tools lead to the creation of even better tools. Electron microscopes help to create better semiconductors, which help to create better electron microscopes. This virtuous cycle expands so rapidly that in a mere fifty years we have gone from field evaporation of a few atoms of tungsten to a program to build an atomic-scale tomograph that can capture 1,000,000,000-atom images of nearly every atom in a specimen with atomic-scale precision. Could John Panitz and Erwin Müller have envisioned this eventuality? Yes, we think so. Then imagine, if you can, what the next fifty years will bring.

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