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# Microprobe Design in the 1950s: Some Examples in Europe

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**Abstract:** The early days of the electron microprobe were characterized by the variety of designs emerging from different laboratories in Europe, the United States, and the USSR. Examples from Europe illustrate well the diverging trends in the evolutionary process at that time. Later, commercial pressures and a better understanding of user needs forced a rationalization on both sides of the Atlantic, to the point where only few variants have survived. These were memorable days, with scope for healthy rivalry and vigorous debate.

**Key words:** electron microprobe, magnetic lens design, X-ray spectrometers, microprobe analysis, X-ray microanalysis, instrument design

## Introduction

In this commemoration of 50 years of microprobe analysis, I shall look back to the first decade, before the technique became commercialized in the 1960s. Papers in this issue by Jean Philibert, Kurt Heinrich, and others introduce the broad dimension, so I can concentrate on a relatively narrow aspect of the history: the evolution of instrument design in Europe during the 1950s. There are two reasons for choosing this topic, and I hope it will interest those who came later to the subject, as well as those who were active at the time:

1. To try to convey something of the flavor and freshness of the subject in its early years. By the end of the decade there were still fewer than 20 instruments in existence or under construction around the world. All were home built; all were different; most of the designers knew each other, and there was ample fuel for healthy debate. Nearly all were represented at a memorable meeting in 1958 in Washington, DC, organized by LaVerne Birks of the Naval Research Laboratory, who was himself one of the early pioneers in the US.
2. To show how the design of these instruments was influenced by the particular applications for which they were intended, and by the interests and background of the designer. Later the pressures of commercialization led to a convergence of design into fewer variants, but it was these early instruments which explored for the first time the trade-offs needed to reconcile the many conflicting design requirements.

Before looking at some of the early probe instruments in detail, we travel back a further 50 years to survey the foundations upon which the subject was built (Fig. 1). With the discovery of X-rays and electrons at around the turn of the 20th century, the basic physics was soon understood, but it required the invention of the curved crystal spectrometer and the magnetic electron lens to provide the practical components of the microprobe. Although the possibility of microprobe analysis was recognized by Hillier, he preferred to concentrate on electron energy loss spectroscopy, and it

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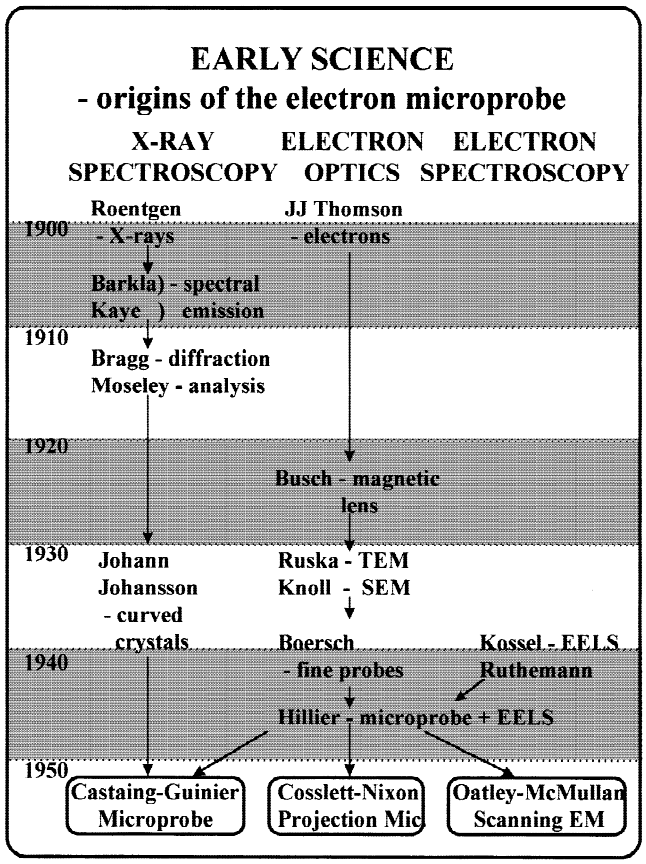


**Figure 1.** Streams of discovery and invention leading to three forms of electron probe instruments under development in the early 1950s. TEM, transmission electron microscope; SEM, scanning electron microscope; EELS, electron energy loss spectroscopy; EM, electron microscope.

was left to Castaing and Guinier to build the first working microprobe. Hillier's work is also acknowledged by Cosslett and Nixon in the invention of the X-ray projection microscope, and by Oatley and McMullan who pioneered the scanning electron microscope. In turn, these three instruments provided a firm basis for the microprobes to be built in the 1950s.

## Trade-offs in Microprobe Design

It was never difficult to write a wish list of the features a microprobe analyzer should provide—even one which was superficially in accordance with the laws of physics. The difficulty came in putting these wishes into engineering practice, given the limited space around the sample and the practical efficiency of the electron-optical column and X-ray detectors. Figure 2 shows a schematic diagram of a

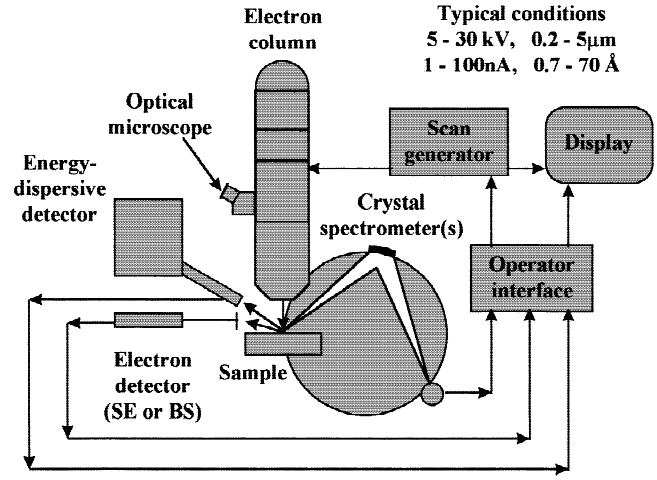


**Figure 2.** Schematic diagram of an electron probe microanalyzer, showing the electron column, X-ray spectrometer(s), detector for secondary (SE) or backscattered (BS) electrons, energy dispersive X-ray detector, and scanning system.

generic microprobe. This has remained relatively unchanged over the years except for the form of the energy-dispersive detector, which in the 1950s was a gas proportional counter.

It is convenient to list the design compromises under three headings:

1. *X-ray spectrometers:* Should these be fully focusing to give high spectral resolution, energy-dispersive to give a high collection efficiency, or semi-focusing to provide a compromise? How many spectrometers should there be?
2. *Electron-optics:* Should the gun be operated at high potential, such as 20–30 kV, in order to give high intensity and well-resolved characteristic radiation, or below, say, 10 kV to give good spatial resolution and low sample absorption? Should the lens be designed with a long working distance to allow a high take-off angle to the spectrometer or with a short focal length to improve the current density in the probe?
3. *Sample disposition:* Should the sample be viewed optically or by secondary electrons, by electron backscatter, or by all three signals? How many samples should be accommodated, how big should they be and how easily interchanged? Should space be allowed for a cold finger or air jet for decontamination, or for devices for in situ experimentation?

These were just a few of the problems facing designers in the

1950s and, with no body of experience to draw upon, it is not surprising that some very different solutions emerged.

## Castaing and His Two Microprobes

Castaing's thesis (Castaing, 1951) is one of the most widely referenced works in microprobe literature, and was the first real test for my schoolboy French. Later, it was translated into English by David Wittry, and has just been reproduced. In it, Castaing describes the modification of an electron microscope made by the Compagnie Française de Télégraphie Sans Fil to prove the principle of microprobe analysis. This was an electrostatic instrument which did not easily lend itself to the delivery of a high intensity electron probe, nor to the inclusion of an optical microscope for viewing the sample. Initial estimates of the count rates obtained with normal X-ray tubes were discouraging until Castaing realized that, in the microprobe, the X-rays would all be coming from within the spectrometer focus, thus greatly improving the detection efficiency. For this reason, no thought was expressed in his thesis of scanning the electron beam over the sample, as this would cause an unaceptable drop in efficiency—an effect which indeed is real, but which is capable of being offset by degrading spectrometer resolution. Thus, the first instrument produced a static probe, capable of delivering only a few nanoamps into a micron-sized probe. The sample was viewed by a mirror under the lens and the X-rays detected with a fully-focused Johannson spectrometer in a direction almost at right-angles to the electron-optical axis. To give a reasonable take-off angle, the sample was tilted a few degrees towards the spectrometer.

Soon after his thesis was complete, Castaing began the design of a new instrument at the ONERA Laboratories, intended for metallurgical applications. The probe-forming lens was totally redesigned as a magnetic lens, with greatly improved spherical aberration, and had the objective of an optical microscope built coaxially into the lens bore. The lens configuration is shown in Figure 3a and was first described at the International Conference on Electron Microscopy in London in 1954 (Castaing, 1956), where, with awe as a new research student, I met Raymond Castaing for the first time. In his second instrument, there were two spectrometers—both fully focusing—carefully aligned to the point on the sample under the cross-hairs of the microscope, to which the probe itself was also directed. The contamination spot that appeared on the sample after prolonged exposure to the beam provided an important confirmation of the alignment and served to show up any error due to the presence of a magnetic sample. A further instrument of this type was used by Philibert and others in the mid-1950s for pioneering investigations into problems in ferrous metallurgy (Castaing et al., 1957), and became the prototype of the first CAMECA microprobe.

## To Scan or Not to Scan?

In the meantime, my activity in the Cavendish Laboratory in Cambridge was devoted to building an instrument for scanning the electron probe over the sample, in order to image the distribution of selected elements by means of their characteristic radiation. This was inspired by Ellis Cosslett, my supervisor, and greatly helped by Bill Nixon who had developed the X-ray projection microscope (for a full account, see Cosslett and Nixon, 1960). We were joined in 1954 by Jim Long, who brought with him a knowledge of experimental techniques, including how to make gas proportional counters. I owe much to them all. We could also visit our colleagues in the Engineering Department nearby who were developing the scanning electron microscope under Charles Oatley, Dennis McMullan, and Ken Smith (McMullan, 1953). We learned a lot from them—particularly in the matter of scanning—and in return were able to steer them towards the use of magnetic lenses. There was a clear demarcation between us: Engineers were not interested in X-rays, and we had no use for probes of very low intensity. Consequently, relations were very harmonious, and I even survived a grilling oral examination from Oatley when my dissertation was presented.

Early on, Cosslett had realized that the only way to produce high intensity probes was to use magnetic lenses, with their generally lower spherical aberration. This was already well demonstrated in Nixon's projection microscope. Cosslett was also well attuned to the work at the Associated Electrical Industries Laboratories (AEI) on electron guns (Haine and Einstein, 1952) and on electron lenses (Liebmann and Grad, 1951). Both provided invaluable information for the microprobe designer and do to this day. So it is not surprising that the first scanning microprobe used a strong immersion lens with a small sample in the bore of the lens itself and a pathway out for the X-rays through the pole-piece gap. This is illustrated in Figure 3b. Like Castaing's first instrument, it was modeled around a converted electron microscope—in this case a type EMB by the Radio Corporation of America that came over from the

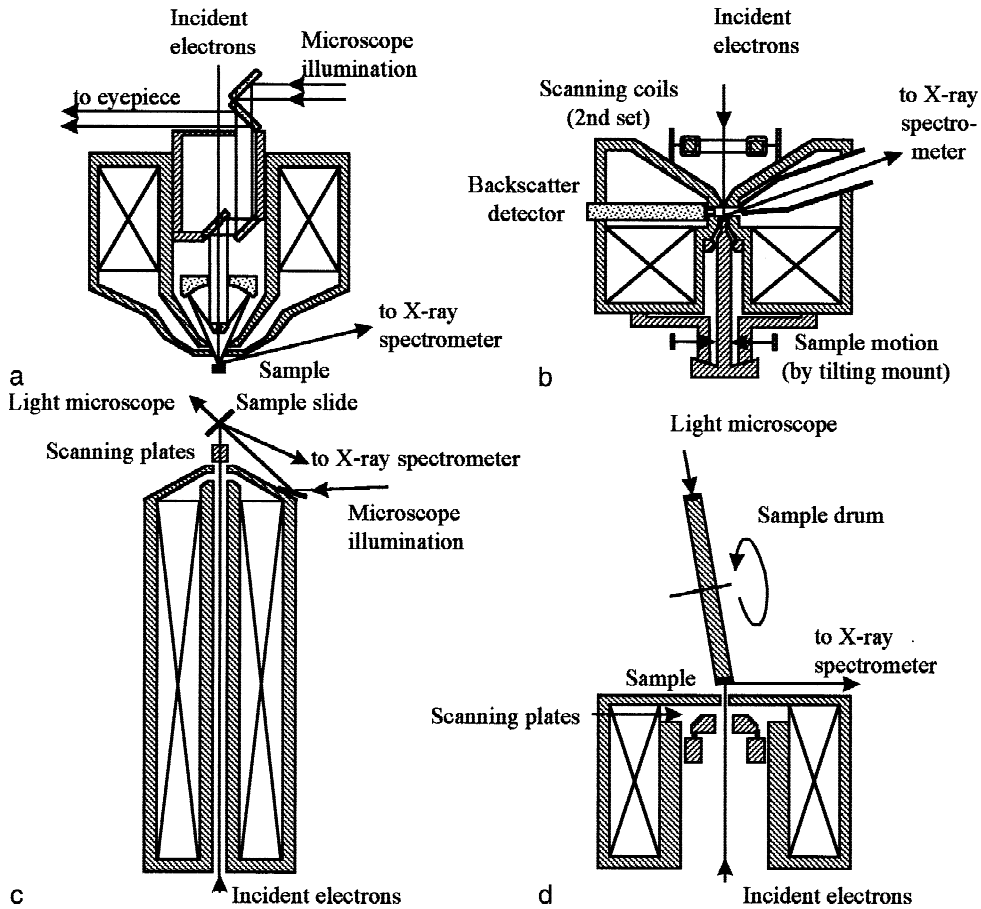


**Figure 3.** Some contrasting designs of magnetic probe-forming lenses in the UK and France, 1954–1958. **a:** Castaing's 1954 lens for CAMECA, leading to the first commercial microprobe. Features included a built-in light microscope, static electron probe, and a fully focusing spectrometer. Lens a was designed for the metallurgist user and had a built-in microscope for positioning the sample. **b:** Duncumb's 1956 lens for probe scanning, a forerunner of the Cambridge Microscan. Features included a high intensity probe, double-deflection scanning coils, and a semi-focusing spectrometer. In lens b, the focal length and lens aberration were made as small as possible to improve quality in the X-ray scanning image. **c:** Long's 1958 lens for mineral samples, a forerunner of the Cambridge Geoscan. Features included transmission light optics, scanning plates, and a semi-focusing spectrometer. Lens c was designed to allow a slide-mounted rock sample to be viewed in transmitted light under the electron probe. **d:** Mulvey's 1958 wide-bore lens, leading to the Associated Electrical Industries Laboratories (AEI) microprobe. Features included a sample rotated on drum for viewing, scanning plates inside lens, and a fully focusing spectrometer. Mulvey's lens combined a high efficiency lens with a fully focusing spectrometer.

US after the war under the generous Lease-Lend scheme. The EMB formed the basis of at least two other microprobes in the US and provided an excellent "Meccano set" for a research student. I fitted ours with a semi-focusing spectrometer (with help from Jim Long) from which the crystal could readily be swung out of the way to allow X-rays to enter the proportional counter directly.

The first results were shown at the "Symposium on X-ray Microscopy and Microradiography" in Cambridge in 1956 [the first of the triennial International Congress on X-ray Optics and Microscopy (ICXOM) series which continues to this day], including a color map of crossed silver and copper grids made by superimposing the element distributions as color separations (Duncumb and Cosslett, 1957). Later, a resolution of 0.3 μm was achieved by reducing the beam voltage to 6 kV, using an end-window proportional counter inside the lens to improve collection efficiency (Duncumb, 1960). There was no shortage of spherically tipped anodes for the counter; they were obtainable from any high-street record shop ("soft playing" recommended!). It seemed a good idea at the time, but of course was superseded entirely by the semiconductor detector.

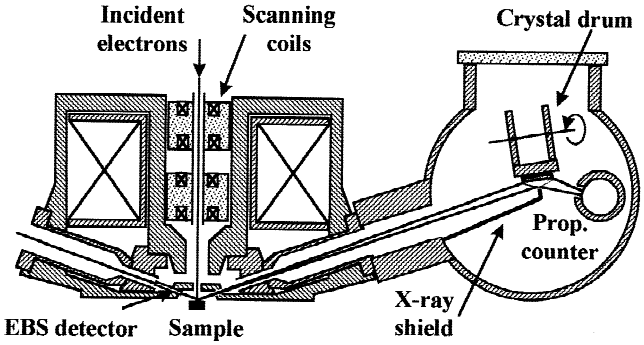


**Figure 4.** Duncumb and Melford's 1959 lens and semi-focusing spectrometer for metallurgical applications. The sample was mounted on a turntable clear of the lens and could be swung under a reflecting microscope for optical viewing. Electrons backscattered (EBS) as the incident beam was scanned over the sample were recorded by the EBS detector, while X-rays were detected by the proportional (Prop.) counter after diffraction by the crystal.

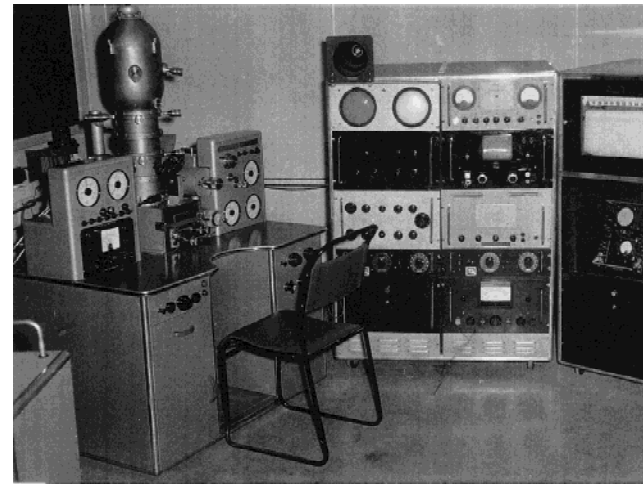

**Figure 5.** Scanning electron probe microanalyzer for metallurgical use, built at Tube Investments Research Laboratories (TI) in 1959, showing the column (left) and scanning displays (center).

## Pretty Pictures, But Are They Useful?

While showing the capabilities of the instrument, scanning pictures of grid samples did not really advance the boundaries of knowledge in materials science, and it was not until 1957 that "useful" applications emerged. Notable among these was a problem posed by David Melford of Tube Investments Research Laboratories (TI) concerning the segregation of trace elements in steel, which was shown to cause surface cracking in the finished tubes (Melford, 1960). We managed to solve this problem in one of those rare days when everything went right. The consequence was that TI decided to build an instrument of their own which was subsequently licensed to the Cambridge Instrument Company (now LEO Electron Microscopy) as the Cambridge Microscan. It also led to my own employment at TI for 28 years—ample reward for a day's work!

As a physicist, I had to concede that metallurgists did need to examine rather larger samples than was possible in the Cavendish instrument—only 3 mm across. Consequently, we designed the Microscan to focus the probe outside the bulk of the lens, so that the sample could be easily interchanged and examined under an optical microscope (Duncumb and Melford, 1960). The lens geometry is shown in Figure 4 complete with the semi-focusing spectrometer that was attached. This was later supplemented by a second spectrometer, both being provided with four crystals of different radii and with a blank position for straight-through transmission to the gas proportional counter. Notwithstanding the poor energy resolution, this last facility was surprisingly useful for the initial exploration of an unknown sample. This combination seemed to appeal to material scientists, and some 87 Microscans were sold worldwide during the 1960s. The prototype of this instrument is shown in Figure 5.

Before I moved to TI, we were joined in the Cavendish by a research student, Ray Dolby, who took on the task of detecting the light elements—a goal of major importance to metallurgists and biologists alike. No one had counted individual X-ray quanta from carbon radiation at 44A before, and I have a vivid memory of Dolby demonstrating this one coffee time in 1959 to the assembled group, using a proportional counter (the only method available at that time). The next question was how to separate overlapping pulse-height distributions from other elements in this region of the periodic table. Dolby solved this, too, with his "matrix" system—a type of analogue computer—described at the 2nd ICXOM in Stockholm (Dolby and Cosslett, 1960) and 3rd ICXOM in Stanford (Dolby, 1963). In the later paper, he went even further to show separated images of beryllium, carbon, and oxygen from one sample. A copy of his equipment was supplied with the first Microscan to be built commercially in 1960. Dolby went on to do other things, making his first impact on the audio field later in the 1960s.

## Other UK Instruments and Their Descendants

As I moved from the Cavendish in 1959, Jim Long and Bill Nixon moved too—Long to the Department of Mineralogy

and Petrology, and Nixon to Engineering. Long had formed an interest in the application of the microprobe to mineralogy and had built an instrument for the purpose which embodied a light microscope for viewing samples in transmission and reflection, necessitating the design of a new form of lens (Agrell and Long, 1960). This took the form of an elongated cylinder that could be inserted into the spectrometer chamber, leaving room for the light optics as well as a semi-focusing X-ray spectrometer. Although it was less efficient electron-optically, the system could accept normal slide-mounted rock samples, which in any case could not withstand high current densities in the probe. The lens is illustrated in Figure 3c. The high take-off angle of 75° made it practicable to study rough samples, and results were so encouraging that Long created a new design having the same take-off angle but with a much improved lens configuration and two fully focusing spectrometers. This was later made by Cambridge Instruments as the Geoscan.

If the Microscan and the Geoscan were two descendants of the Cavendish work, a third was the combined microscope and microanalyzer (EMMA), which we built at TI in the early 1960s (Duncumb, 1966). Its successor was commercialized by AEI under the name EMMA4 in 1967 and was the forerunner of the modern analytical electron microscope. This instrument was applied not only to thin metal samples but also to sections of biological material, and proved useful in the study of various types of asbestosis. It is interesting to note that it had been the possible application to lung tissue that had stimulated Cosslett's interest in microprobe analysis some 15 years earlier.

As the only British manufacturer of the transmission electron microscope during the period, AEI played a major part in the field as a whole, starting with the fundamental electron-optical work referred to above. Quick to apply this to the microprobe was Tom Mulvey, who designed the AEI microprobe described at the 2nd ICXOM (Mulvey, 1960). With the strong background in electron optics in his laboratory, the probe-forming lens was a low-aberration pinhole (shown in Fig. 3d). The sample was mounted on a drum that allowed it to be rotated accurately under a normal light microscope. The spectrometer was set horizontally which required the sample to be tilted to allow the X-rays to enter it, but this gave ample space for fully focusing geometry. Scanning plates were added and the instrument was sold as the AEI microprobe. Moving to Aston University, Mulvey went on to explore a whole range of novel lenses, including the "pancake" and "mini-lens," which created widespread interest in the electron-optical community and beyond.

In terms of sheer numbers, evolution has favored the scanning electron microscope over the true microprobe. The first commercial instrument was the Stereoscan, launched in 1965 by the Cambridge Instrument Company (CIC). This derived mainly from the work in Oatley's Laboratory, which was transferred to CIC by Gary Stewart, who eventually became Technical Director there. With their experience in making the Microscan, CIC were well placed to take this on—indeed some of the same components were employed—and sales exceeded all expectations. This was partly due to the eye-catching high-resolution display, combined with an excellent standard of engineering. Later, the introduction of the silicon detector gave the scanning electron microscope (SEM) some of the capabilities of a microprobe at a much lower price, and this compromise proved popular for many users.

## Rest of the World

I intended at the start to confine this contribution to Europe in the 1950s, but it has proved unrealistic to do so entirely. The meeting in snowy Washington in February 1958 was a key event on the international scene, and we regretted only the absence of Igor Borovskii from Moscow, who had been working in parallel with Castaing, unreported in the West. Little was known about his early work. I had received a copy of David Wittry's Ph.D. thesis in 1957—the same year as my own—and was delighted to meet him in person in Washington. I also met Bob Fisher at US Steel and Bob Ogilvie from MIT, both having a strong interest in the application of the microprobe to materials science. It was not until the following year that I met Jean Philibert, but we continued the relationship into the 1960s by exchanging our staff (and later our children). These were some of the contacts that lasted for many years afterwards. By 1960, Raymond Castaing had developed other interests, though he was strongly in evidence at the 3rd and 4th ICXOM meetings. His last major review of the technique appeared in *Advances in Electronics and Electron Physics* in 1960 (Castaing, 1960) and marked the "end of the beginning" in microprobe analysis.

Growth was exponential and, at the "Zeroth" meeting of the Microbeam Analysis Society in 1964, there were nearly 50 papers on instrumentation, theory, and applications. These included important contributions by Kurt Heinrich and by Ryuichi Shimizu, and I shall leave it to them and other authors to build a picture of the world

scene. Before doing so, however, I should like to return to the subject of tradeoffs in design, as seen from 50 years on.

## 50 Years of Evolution

The microprobe analyzer is alive and well. It has come to be an instrument with multiple spectrometers under computer control with analysis conditions that can be set up before an experiment or adapted as the experiment proceeds. Accuracy and stability have improved, as has ease of operation. Results are processed automatically by much improved correction procedures, and the determination of elements down to boron in the periodic table is commonplace. How was this achieved and what remains to be done?

Of course, the advent of cheap computer power has revolutionized the operation of the instrument. It has also greatly improved the ability to optimize lens design and indeed spectrometer geometry. Thus, it has been possible to model design performance with far greater accuracy without having to build an actual instrument, thereby greatly speeding the evolutionary process.

However this is only part of the equation for producing a successful instrument. Equally important is the recognition of user needs—the segmentation of the potential market to match the techniques available. Here we have seen the SEM plus energy-dispersive spectrometry (EDS) displace the microprobe where high accuracy is not needed or there is not a large number of elements to be resolved from one another. The visual and almost three-dimensional appeal of the SEM image was recognized in naming the Stereoscan, and much compositional information can be gained from the use of modern EDS. The introduction of the silicon detector in the late 1960s gave a huge impetus to the technique. New types of energy-dispersive detectors now promise even better performance.

Where many elements are present with overlapping lines, such as in minerals containing rare earths, there is currently no practical alternative to the crystal spectrometer. The same is also true for accurate analysis below about 1 keV in energy, where the resolution of the silicon detector is often not adequate. For practical purposes, not just one or two spectrometers are required, but as many as five may be needed for simultaneous operation. Control has to be by computer, in order to program an overnight run which may contain a degree of adaptive control. We look to the day when this degree of intelligence equates to that of an "expert," able to advise the non-specialist user on experimental setup and run the experiment entirely. However, this expert intelligence is unlikely to extend as far as the design process itself and, as new techniques appear, there will always be a challenge—and indeed an art—in resolving the same compromises faced by the designers of the 1950s.

## Acknowledgments

I dedicate this short history to Raimond Castaing and to the many others who shared their experiences in microprobe analysis in those early days.

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