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# FROM MICRONS TO NANOMETRES: EARLY LANDMARKS IN THE SCIENCE OF SCANNING ELECTRON MICROSCOPE IMAGING

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## Abstract

Optimisation of a technique such as scanning electron microscopy implies a synergism of technological advance, innovative design and user needs. Development of the scanning electron microscope (SEM) into a commercially viable and usable instrument during the 1960s and 1970s is analysed in this light. Advances in resolution, image quality, multisignal detection and processing, and specimen manipulation during this period, which contributed to the evolution of the SEM into the facility available today are examined in the context of the prevailing technological environment in and around Cambridge.

**Key Words**: Scanning electron microscope, history, Cambridge, instrument design.

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## Introduction

Those who seek a comprehensive history of the development of the SEM are referred to some excellent and authoritative reviews (Agar, 1996; Oatley, 1982). This paper asks not when? and how? but why? It is an examination of SEM development, principally during the period from its commercial introduction (1965) through its first decade, which seeks to put SEM development into the technological context of its time – in the hope that, by understanding why the SEM developed as it did, we can better select the way forward to the next generation of instruments.

This is a very personal view; I worked for the then Cambridge Instrument Company (henceforth referred to as CIC, regardless of contemporary name changes) from 1960 to 1974, a time when the Cambridge area was a veritable hot-bed of SEM-related innovation. By centering my discussion upon this small world, I mean no disrespect or slight to other researchers or manufacturers; their stories would differ, but I believe that the conclusions one may draw from them in the context of this analysis would not.

## Pre-1960

Early work on SEM-type instruments was performed in Germany (Knoll, 1935; von Ardenne, 1938a,b). It would be fair to say that at that time scientific thought was ahead of technology; it was not until the late 1940s that true instrumental development commenced. 1948-9 saw the development of the transistor at Bell Labs, the announcement of the binary arithmetic computer, and, at Cambridge University Engineering Department (CUED) the start of Ph.D. studies in scanning microscopy under Dr. Oatley. Three vital ingredients for future success were now in place.

The 1950s saw the foundations of SEM laid. Under Oatley's leadership and guidance a series of graduate students contributed many of the cornerstones of SEM theory and instrumentation (Oatley, 1966, 1972, 1982). Electron emitters and guns, lens design, double deflection scan coils, stigmators, secondary electron detectors and signal amplifiers, contrast mechanisms, power supplies and early experiments in beam writing; an unending stream of ideas and improvements poured forth.

Over this same period, Dr. Cosslett's group at the

Cavendish Laboratory had pursued a different course, not only into various branches of transmission electron microscopy (TEM), but in combining the quantitative analytical capability of Castaing's "microsonde" (Castaing, 1951) with the imaging afforded by scanning, to produce the scanning electron probe X-ray microanalyser (Cosslett and Duncumb, 1957). The potential of this was realised by Tube Investments, for whom a prototype was constructed at their Hinxton research laboratories just outside Cambridge (Duncumb and Melford, 1959).

**1960-1965**

It was this microanalyser which CIC put into commercial production in 1960 as the Microscan. This instrument fulfilled a dual role: not only was it a commercial success in its own right, but it also provided CIC development and production staff with five years valuable experience in the diverse technologies essential to an electron beam system. Over this period, as the Microscan matured through MkI, MkII and MkIIa models, advances in quantitative analytical capability were accompanied by, for example, the change from round-face, curved-screen cathode ray tubes (CRTs) to square-face, flat-screen versions and significant improvements in the phosphors used on the display screens. While, in retrospect, this may appear trivial, consider it in the context of the transfer from what was essentially an analytical instrument, the Microscan, to what was initially a purely imaging instrument, the Stereoscan. The fastest scan took one second; all viewing was done in a darkened room and even then a viewing hood was used for critical operations such as beam focusing. Recording was by 35mm camera, swung down over the CRT. Early microprobe images clearly show blurring at their extremities caused both by defocusing of the CRT itself and by recording onto a plane film from a curved screen. With the advent of the Stereoscan, a distinction was made for the first time between viewing and recording; CRTs with optimised characteristics were introduced - bright, long-persistence phosphors for viewing, high resolution, short-persistence phosphors for recording. Development of this concept by CIC's own research engineers under A.D.G. Stewart (one of Oatley's former students) allowed image quality approaching that of photographic film to be obtained for the first time. Nevertheless, it is a salutory lesson to read Everhart's recommendations on procedures for optimum recording (Everhart, 1969) which conclude with the advice: 'The CRT should be adjusted for maximum resolution by checking the CRT voltage ...and then focusing the CRT beam accurately...'. And that referred to the "new" CRTs!

So, as serious SEM development commenced in 1963-1964, what were the most significant factors restraining performance? Recall that the first integrated circuits were developed in 1958 – commercial electronics were still entirely dependent upon valve technology; while in 1963 Digital Equipment Corporation (DEC) had just introduced their PDP 8 minicomputer. SEM performance was restrained by power supply stability for both the high voltage and the electron lenses: both long-term drift and high frequency ripple posed problems in obtaining and maintaining focus. The Microscan had used a two-lens electron optical column to produce its one micrometre spot size and was fully alignable – the tungsten filament, the entire electron gun and the condenser lens were traversed mechanically on sliding O-ring seals and were aligned with the aid of a phosphor screen which could be swung into the beam path. Adjustments were frequent and owed more to a fitness-for-purpose philosophy than to true alignment.

The Stereoscan required a three-lens column to obtain the necessary demagnification of the tungsten hairpin source. Gun alignment was retained, but the lenses (still with their sliding O-ring seals) were at least factory aligned. While CIC had a longstanding reputation for precision mechanics, the introduction of computer numerically controleed (CNC) machining centres facilitated the change-over from one-off fitting to batch part interchangeability. But the total evacuated volume and the number of moving seals were enormous and posed a significant pumping problem, state-of-the-art in vacuum technology was diffusion pumps, backed by rotary pumps; the oils used permeated the system and deposited a characteristic "contamination mark" where the electron beam impinged upon the specimen surface. By the time focus and astigmatism had been corrected, high resolution detail could disappear into a murky haze!

The first SEMs were seen as purely imaging instruments, their potential for multi-signal collection and processing was to be realised later. Quality of the secondary electron image was therefore paramount. The detector itself followed Everhart's design (Everhart and Thornley, 1960), but enhancing overall signal-to-noise ratio was an unending process of optimisation of a myriad of individual parameters: efficiency of the phosphor tips which converted electrons to light and which decayed rapidly under electron bombardment; internal reflection efficiency of the perspex light guide; the careful placement of the glycerine drop which provided continuity of the light path to the photomultiplier; the quality of the photomultiplier itself, and finally the paucity of signal processing capability, all came under scrutiny. While design engineers sought eagerly for solutions to these and other shortcomings (many of which were to originate in the burgeoning field of nuclear physics), the SEM operator was left with simple alternatives: increase the beam current or the scanning time. Bear in mind that Polaroid film was both expensive and positive-only, 35 mm film was the norm, with hours or even days delay in receiving the final print and only the operators eyeball between success and failure.

These then were the innovations and limitations to

**Table 1**. Comparison of typical SEM features 1965 and 1975.

| *Feature* | *1965 - Stereoscan Mk1* | *1975 - Stereoscan S180* |
|---|---|---|
| Gun | tungsten filament<br>1 - 20 kV<br>manual alignment | tungsten or $LaB_6$ emitter<br>1 - 60 kV<br>electromagnetic alignment |
| Electron Optics | 3 demountable electromagnetic lenses<br>manual control<br>50 nm resolution | 3 prealigned electromagnetic lenses<br>kV compensated<br>7 - 10 nm resolution |
| Vacuum System | mineral oil diffusion pumps<br>manual control<br>sliding O-ring seals | synthetic oil diffusion pumps<br>automatic control<br>liquid nitrogen trapping<br>viton or metal seals |
| Specimen Stage | 12 mm diameter stub<br>X,Y,Z and 0 - 90 degree tilt | 50 mm diameter stub<br>goniometer and special function stages (heating, cooling, tensile, bending, microanalysis, transmission, microcircuit) |
| Scanning System | 1 viewing CRT<br>1000 line record CRT<br>35 mm camera<br>analogue scanning system<br>0.1 sec fastest scan<br>x200 minimum magnifiction | 2 viewing CRTs<br>2000 line record CRT<br>Polaroid camera<br>digital scanning system<br>TV-rate fast scanning<br>x10 minimum magnification<br>Pulse width modulation exposure |
| Imaging Signals | secondary electrons (SE)<br>backscattered electrons (BSE)<br>specimen current (SC) | SE, BSE, SC, STEM<br>X-rays (wavelength-dispersive and energy-dispersive)<br>cathodoluminescence (visible, ultraviolet and infrared)<br>electron channelling (selected and large area) |
| Image Processing | gamma<br>black level<br>rise time | CRT alpha-numerics and magnification, dual magnification, derivative, inversion, contours, grey levels, expanded contrast, tilt and rotate correction |

which the Stereoscan was born. Subsequent development might conveniently be classified by decades:

1965-1975 Instrumentation
1975-1985 Multisignal capability
1985-1995 Computerisation

The latter two of these were dominated by the introduction in 1970-1971 of the solid state X-ray detector and of the microprocessor, which, together with the advent of digital electronics created an extension of SEM capability unforeseen in 1965. It is with instrumental developments in the period 1965-1975, which so greatly enhanced the basic SEM performance, that we shall now be concerned.

**1965-1975**

Comparison of selected features of contemporary Stereoscans at the two extremes of this era will serve as a basis for discussion (Table 1, Figures 1 and 2).

The increase in accelerating voltage from 20kV to 60kV was not in itself technically significant (the Microscan had 50kV capability), but was an acknowledgement of the changing role of the SEM. Higher accelerating voltages were seen as desirable not only for use with the new energy-dispersive X-ray (EDX) detectors (which were inefficient at longer wavelengths due to poor transmission efficiency of

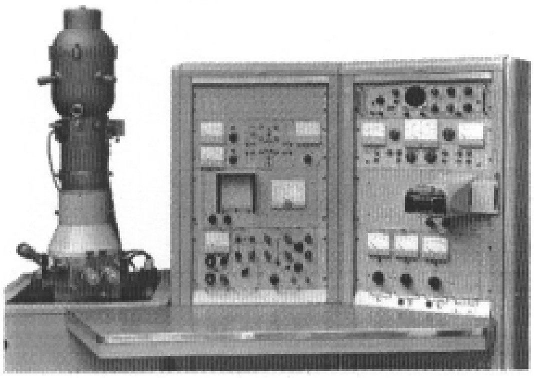

**Figure 1**. Stereoscan Mk. 1, circa 1965.

the Be window, and hence used the higher energy X-ray lines wherever possible) but also for scanning transmission imaging, electron diffraction studies and even electron beam writing, all of which had been performed experimentally in the SEM and were perceived as potential areas of application.

The changes from mechanical to coil alignment and the ability to optimise anode height for changes in kV without dismantling the system might be termed operator conveniences. But the electron emitters themselves were both a major technical advance and, from an optimisation viewpoint, a source of contention. During the late 1960s, Broers at CUED and subsequently at IBM had developed the $LaB_6$ emission system (Broers, 1969); while Crewe and then Welter in the USA had worked on field emission (Crewe *et al*., 1968). Both of these required the improved vacuum environments which had now become available. Whereas $LaB_6$ offered potential for long-term stability and a modest gain in brightness, FE systems with their small, ultra-bright source offered scope for simplified electron optics and high resolution, but proved initially to be very unstable in all but the most expert hands (Smith, 1972). Either option added substantially to the instrument cost.

The advances in vacuum technology which facilitated the use of these emitters had been gradual and less spectacular, but were no less significant. Diffusion pumps had undergone design modifications, including better baffles and liquid nitrogen cooled trapping to reduce oil backstreaming into the SEM. The pump oils themselves had changed from mineral-based to silicone; it is interesting to note that proprietary synthetics such as Santovac 5 and Fomblin were the subject of considerable discussion in the scientific press (Holland, 1972). It had also become apparent that, while electron guns demanded better and better vacuum, SEM users, particularly those investigating organic material, wished to place increasingly more vacuum-hostile specimens in the chamber; while a large diffusion pump satisfied the latter, the new ion pumps, with no boiling oil to create unwanted vibration, and which therefore could be attached directly to the gun, opened the door to alternative electron sources (Swann and Kynaston, 1973). Silicone greases, synthetic O-rings and soft metal seals contributed to the improvement in vacuum environment; while the change in electron optical design from alignable building blocks to simple stacking allowed a central liner tube to be incorporated, reducing by orders of magnitude the surface area and evacuated volume. These and many other factors contributed to the steady improvement of resolution from 500 Å (50 nm) to 50 Å (5 nm) over the decade.

The impact of the change from analogue, valve-based electronics to digital solid state was tremendous (Paden *et al*., 1973). The effect upon reliability, stability and power consumption is self-evident; other aspects require a more careful analysis. Improvements in imaging, both the scanning system and signal processing, were vital to the further development of SEM technology. By 1975, the multi-signal potential of the SEM had become apparent; the information to be derived from reflected electrons, specimen current imaging, X-ray emission, cathodoluminescence, electron diffraction (both large and selected area) and voltage contrast effects had been reported in the scientific literature; each such signal had its own frequency band, signal-to-noise ratio and fundamental resolution. Specimen stage capabilities had also proliferated; goniometric stages for stereo imaging, heating, cooling and tensile stages all required observation of dynamic events. While operator skill and a degree of compromise allowed all these imaging requirements to be met using a simple signal amplifier and a modest selection of scan speeds, the images were most certainly far from optimised. Digital electronics provided virtually total flexibility in scan speed and scan area selection; dual magnification display, micrometre marker bars, magnification and alpha-numeric labelling became the norm. Dynamic focusing of the record CRT finally disposed of the "wooly focus" at its extremities and 2000 line resolution became the industry standard, immediately improving micrograph resolution by a factor of two.

Television (TV) or pseudo-TV scan rates had resulted from advances in both scan generator and scan coil design which improved their frequency response; hence, ambient light level was no longer so critical, the SEM came out of the dark room, direct observation and video recording of dynamic events also became available. The first elements of image processing appeared, albeit hardware based; typically, amplifier response, derivative processing, contour mapping and solid state image storage were applied. Pulse width modulation, applied under recording conditions, maintained photographic exposure regardless of integration time; this, combined with availability of Polaroid positive/negative film, provided in real time, reliable photo records.

Most of these changes occurred gradually over the ten-year period. They were pioneered by many different users

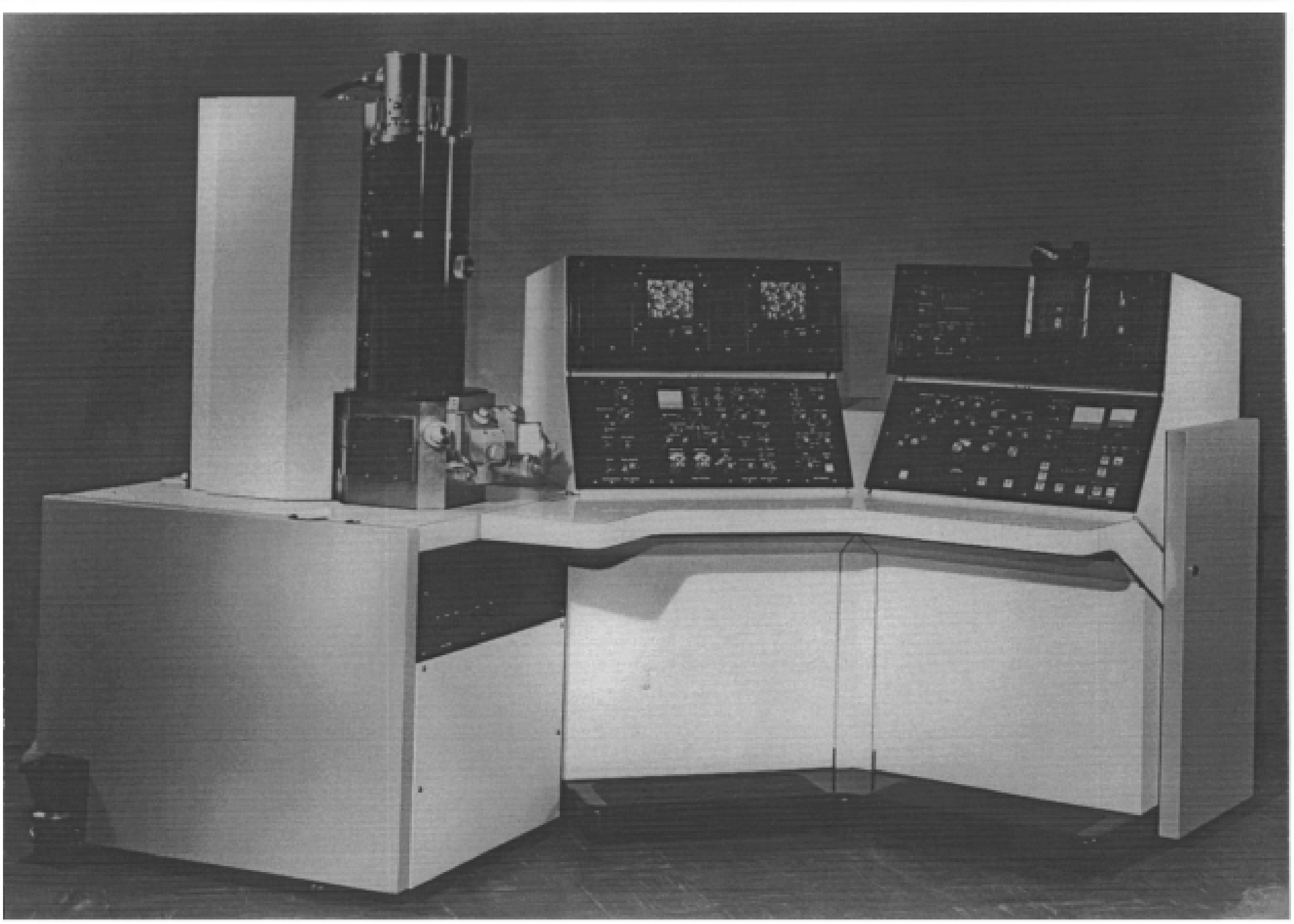

**Figure 2**. Stereoscan S180, circa 1975.

and manufacturers and by 1975 had matured to usable form and industry-wide availability.

Finally, if we step back, the concept and environment of the SEM had also undergone profound change. An early Stereoscan was both mechanically and electronically almost totally modular; partly to accommodate new developments, but also to facilitate the frequent cleaning which the electron optics demanded. It was customary to have two dedicated attendants, one scientific (operating from first principles) and one technical (with an aptitude for cleaning columns and developing films). The service engineer carried a case full of valves, resistors and capacitors, a soldering iron, an AVOmeter and a very large folder of circuit diagrams. By 1975, valve electronics had essentially disappeared and solid state devices mounted on circuit boards incorporating test points were used; vacuum systems required much less cleaning, and a single operator was more productive than two had been previously. The SEM had changed from a scientific curiosity to a powerful research tool.

Over these ten years, SEM technology benefited from considerable direct development investment, but also from apparently coincidental advances in vacuum engineering, nuclear physics, space technology, photography, radar, cryogenics and image processing among others. By 1975, more than 1000 SEMs were installed in the USA alone, in all sizes from desk-top to the sophistication of Stereoscan 180. We thought that the instrumentation had been optimised, and that its future lay in added ancillary features – we were wrong! Who will now say whether virtual reality or genetic engineering will prompt the next surprise development?

## References

Agar AW (1996) The story of European electron microscopes. Adv Imaging Electr Phys **96**: 415-584.

Broers AN (1969) A new high resolution reflection scanning electron microscope. Rev Sci Inst **40**: 1040-1045.

Castaing R (1951) Application des sondes électroniques à une methode d’analyse ponctuelle chimique et cristallographique (Application of electron probes for chemical and crystallographic point analysis). Doctoral Thesis, University of Paris.

Cosslett VE, Duncumb P (1957) A scanning microscope with either electron or X-ray recording. Proc Stockholm Conf Electron Microsc 1956. Sjöstrand FS and Rhodin J (eds). Almqvist and Wiksell, Stockholm. pp. 12-14.

Crewe AV, Wall J, Welter LM (1968) A high resolution scanning transmission electron microscope. J Appl Phys **39**: 5861-5868.

Duncumb P, Melford DA (1959) Design considerations of an X-ray scanning microanalyser used mainly for metallurgical applications. Proc Second Int Symp X-ray Microsc X-ray Microanal Stockholm 1959. Engström A, Cosslett V, Pattee H (eds). Elsevier, Amsterdam. pp. 358-364.

Everhart TE, Thornley RFM (1960) Wide band detector for micro-microampere low energy electron currents. J Sci Inst **37**: 246-248.

Everhart TE (1969) Optimum micrograph recording with the Stereoscan. Proc Second Annual Stereoscan Colloquium. Engis Equipment Co., Morton Grove, IL, USA. pp. 153-156.

Holland L (1972) Pollution free high vacuum. Physics Bull **23**: 581-585.

Knoll M (1935) Aufladepotential und Sekundaremission elektronenbestrahlter Oberflachen (Charge potential and secondary emission of electron-irradiated surfaces). Z Tech Phys **16**: 467-475.

Oatley CW (1966) The scanning electron microscope. Sci Progr (Oxford) **54**: 483-495.

Oatley CW (1972) The Scanning Electron Microscope. Cambridge University Press, Cambridge.

Oatley CW (1982) The early history of the scanning electron microscope. J Appl Phys **53**: R1-13.

Paden RS, Tillet PI, Upton JM (1973) New display techniques for the scanning electron microscope. Scanning Electron Microsc 1973: 211-216.

Smith KCA (1972) Current work on electron guns at the University Engineering Department, Cambridge, England. Proc Fifth Annual Stereoscan Colloquium. Kent Cambridge Scientific Company, Morton Grove, IL, USA. pp. 93-94.

Swann DJ, Kynaston D (1973) The development of a field emission SEM. Scanning Electron Microsc 1973: 57-64.

von Ardenne M (1938a) Das Elektronen-Raster Mikroskop (The scanning electron microscope). Z Tech Phys **19**: 407-416.

von Ardenne M (1938b) Das Elektronenmikroskop; Theoretische Grundlagen (The scanning electron microscope; theoretical fundamentals). Z Phys **109**: 553-572.

## Discussion with Reviewer

**P. Echlin**: Why didn't the author make mention of the very important work of Ken Smith and Dennis McMullan who were in Oatley's group in Cambridge?

**Author**: While this work was of paramount importance, it was prior to my involvement and it is excellently covered by Oatley (1982) and Oatley *et al.* (1985).

## Additional Reference

Oatley CW, McMullan D, Smith KCA (1985) The development of the scanning electron microscope. Adv Electronics Electr Phys Supplement **16**: 443-482.