Ondrej Krivanek’s contribution to microscopy: Memories of an adventure!

Recollections by Peter Rez, written in 2017 and revised in 2018

I first met Ondrej in 1975, when I was a research student at Oxford. I had gone to visit Richard Leapman and Jean Pierre Chevalier, undergraduate contemporaries of mine in the Cavendish. They had stayed in Cambridge for their PhDs and were sharing an apartment. At the time of my visit Ondrej was also staying there. Shortly afterwards he moved out to a shared house in Grantchester, the place he stayed at until he left Cambridge for his post-doc in Japan.

Richard was doing his research on Energy Loss spectroscopy, under the supervision of the legendary VE Cosslett, using a spectrometer built by Dave Wittry. In those days Energy Loss was considered a somewhat esoteric dead end, and getting results was challenging, hence Richard’s saying “Energy Loss is a Dead Loss”. Ondrej was doing his PhD with Archie Howie. I think it was the first iteration of finding fringes in the images of amorphous materials and wondering whether they indicated the presence of microcrystallites[1]. Ondrej concluded that looking at amorphous materials was invaluable for characterizing the microscope and its aberrations [2] and also that in “amorphous” carbon, high resolution electron microscopy did reveal microcrystallites, while in other amorphous materials it didn’t tell one much [3]. The finding of “microcrystallites” in amorphous images came back from time with a period of about 8-10 years, the matter only being settled with the development of fluctuation microscopy by Mike Treacy and Murray Gibson[4].

We next met at the European Microscopy conference in Jerusalem in 1976. The plan was that Richard, Jean-Pierre, Ondrej and I would all stay with Richard’s relatives in Katamon. When we got there it was clear that this wasn’t going to work, so Ondrej and I split off and found a hotel (I knew the city and could speak some Hebrew). Ondrej did not stay long: after presenting his conference posters, in his adventurous way he hitched to the Dead Sea and Eilat on the Gulf of Aqaba, and came back for the end of the conference. In view of his desire for interesting new experiences, he didn’t come back to the hotel where I stayed, but instead went into a hotel on the Arab side in the old city. I don’t think any of us knew it while we were having fun in Israel, but the image of a grain boundary in Ge that showed the promise of phase contrast high-resolution microscopy[5], was in his baggage.

Ondrej had just got back from a stay at Kobayashi’s lab in Japan where they had a unique high voltage electron microscope with a low Cs. The challenge was seeing the image well enough to stigmate and focus. At that time high-resolution microscopy was looked on as an art-form. Certain individuals had the gift of the “right touch” and “magic eyes”, and the rest of us could never achieve their level of skill. Ondrej saw high-resolution microscopy as a science. He used his knowledge of measuring microscope performance on amorphous materials to develop procedures to iteratively focus and stigmate from images recorded on plates[6]. This image set the standard for more than a decade and justified the development of high voltage, low Cs instruments.

After the Israel conference he went on to a 1 year postdoc at Bell labs. With no access to a high resolution microscope he travelled to Cornell to use their Siemens 102. It was on one of these visits that he produced the classic image of the silicon/silica interface in cross section [7, 8]. At that time there was considerable debate on the nature of this technologically important interface, was it rough or smooth, were there sub-oxide interface species present? Surface science techniques gave ambiguous results. As Ondrej said at the time, the simplest thing to do was to just look at it. The high-resolution image showed an atomically flat interface between the crystalline silicon and the amorphous silica.

We met again as postdocs in Berkeley in Gareth Thomas’s group. The emphasis was on two things, lattice imaging and whether there was an intergranular phase in silicon nitride ceramics. Ondrej elegantly showed the presence of an amorphous phase between Si3N4 grains using dark field imaging with an aperture displaced from the Bragg spots from the crystalline grains [9]. One of the EMSA abstracts that he produced on contrast transfer functions for tilted illumination imaging was well ahead of its time. He assembled this tableau of diffractograms for different x,y tilt [10]. Under computer control many years later this became the technique for aligning the multiple elements in aberration corrected microscopes. If I remember correctly it was part of a competition to see who could produce the most EMSA abstracts that year (with Gareth’s strong disapproval!). In Berkeley he told me that only 4 things mattered in high-resolution microscopy, the source, the specimen, the objective and the detector! While others would spend a lot of time carefully aligning the microscope, Ondrej would come to the lab in late morning, we’d have a nice lunch and dinner, plenty to drink (not quite the microscopissed song!) and he’d then work late in the evening when vibrations had died down. He never bothered with superfluous alignments and often he’d go home to sleep before developing the recorded plates in the dark room the next day.

While Gareth’s dream was “seeing atoms”, Ondrej and I were more excited about the new developments in analytical microscopy. Roy Geiss at IBM had just hung a Kevex EDX detector on a Philips 400 microscope, and was performing elemental analyses at spatial resolutions far superior to what was being achieved in SEMS and microprobes. Originally Ondrej planned to turn the lab’s Philips 300 into an analytical microscope but then the opportunity came to exchange it for a more modern Philips 400. In true Ondrej style he worried about how we could keep both microscopes later: he went around with a “collection hat" for saving the older Philips, one of the department’s workhorse microscopes, several groups chipped in, the workhorse was saved and we gained a new microscope to do developments on. Ondrej thus got exactly what he needed for his planned EELS developments - a microscope with a clean vacuum and good probe-forming capabilities, without having to go through the slow process of raising the full sum for it.

Richard Leapman’s comments notwithstanding, it was also apparent that EELS would be a valuable complementary technique to EDXS, especially for the light elements. David Joy, Ray Egerton, Henry Shuman and Nestor Zaluzec were all building EELS spectrometers at that time. Ondrej went to the 1978 Analytical EM workshop in Cornell and came back inspired to do EELS, and persuaded Gareth that it would be useful for determining the oxygen content of the Si3N4 grain boundary phase. Since there were no commercial spectrometers available, he decided to build one himself. Originally the plan was to copy Ray Egerton’s design, but on one of our weekend hikes (I think it was in Tilden Park), Ondrej announced, “I can do it better”. From the outset Ondrej thought that the spectrometer should be easily fitted to an electron microscope with minimal alteration. This clearly made the instrument more appealing and marketable.

A spectrometer is an electron-optical element, it forms an image (dispersed in energy) of an object. The other designs used the image or diffraction pattern on the viewing screen as the object for the spectrometer. Ondrej realized that it made more sense for the object to be the tiny final projector crossover. To keep the image small the object should also be small! Incidentally the spectrometer that Richard Leapman used in Cambridge, built by Dave Wittry, was also matched to the projector crossover. Fitting the spectrometer into the space under the camera chamber meant that the spectrometer had to be asymmetric, the image distance had to be shorter than the object distance. This further demagnified the image of the object, and the asymmetry later turned out to give extra degrees of freedom for spectrometer aberration correction. Another advantage was that all microscopes had roughly the same distance from the projector cross over to the viewing screen and camera chamber, a dimension constrained by human factors! Ironically this even proved true for the VG where Christian Colliex pointed out that the post specimen objective field moved the effective object point to a distance from the mounting flange comparable to the equivalent distance in a TEM. Most other designs at that time were symmetric, and Ondrej’s design represented a radical departure from the conventional wisdom. Berkeley was a good place to build the spectrometer, there were workshops with relevant expertise from the days when LBL was a particle Physics lab and at that time they did not have much to do.

By then I was working at Kevex. I adapted the company’s EDXS software to run serial electron spectrometers, and Ondrej and I used this software for the new spectrometer. I also traveled around the country and got to see what other people were doing. On a visit to Illinois I met Peggy Mochel, a charming lady who ran the VG STEM. Instead of directly measuring the output current from the photomultiplier tube on the VG spectrometer, she passed it through a discriminator and did single electron counting. This clearly had advantages for the inner shell edges with low cross sections, but the detection circuitry was saturated by the intense zero loss peak (ZLP). Ondrej’s design used current measuring, which was able to handle strong signals, for the ZLP, and pulse counting for the higher losses. Another innovative feature was an energy-selecting slit based on Ondrej’s experience with making cross-sectional TEM specimens. This was superseded by a better design from Peter Swann after Gatan started producing the spectrometer.

On my travels around the country I could see that Ondrej’s design was far superior. Ondrej always liked to get spectacular results for the EMSA conference. I remember we left it acquiring data overnight, to record a silicon K-edge at 1850 eV, which was considered an impossibly “high” energy at that time. We had our result the next morning, but there was oil from the overworked high voltage tank all over the floor! In those days spectra were acquired with an image on the viewing screen. Later on Ondrej used a diffraction pattern on the screen and a small camera length, and got much higher count rates. The area analysed is then defined by the probe on the specimen, as formed by the combination of condenser and objective prefield.

I “sold” a copy of the Berkeley design to Mike Stobbs in Cambridge. However at that time Ondrej had no interest in running a company and so he looked for an appropriate commercial partner. Many of the obvious candidates had already reached agreements to produce one of the other designs. Peggy Mochel told him at a conference in Washington DC, where Ondrej had a poster about Berkeley EELS results, that he should call up Peter Swann of Gatan. Ondrej did this and Peter told him: I saw your EMSA abstract about spectrometer design, and I was thinking about calling you up about it.

A contract was duly drawn up, and since Ondrej was new to the commercial world, my Oxford room-mate (who was a lawyer in San Francisco) vetted it over dinner in a restaurant in Pacific Heights and gave it his blessing. The partnership between Ondrej and Peter Swann was one of those “partnerships made in heaven”. They had the same dedication to perfection and technical excellence. Their skills and interests were complementary and while Ondrej concentrated on spectrometers and CCD cameras[11], Peter continued to design many different types of sample holders and specimen preparation equipment.

The original spectrometer had no aberration correction beyond the first order. It was therefore limited in terms of the angles it could accept, and Ondrej saw that a mark II design was needed. Incorporating adjustable quadrupoles, plus sextupoles and curved faces of the magnetic prism could fix aberrations up to 2nd order. Ondrej worked with Mike Scheinfein and Mike Isaacson at Cornell to come up with the optimum prism shape, Joe Lebiedzik on the electronics and Peter Swann on the mechanical design. The result was the Gatan 607 spectrometer, which soon became the standard serial spectrometer, establishing Gatan as the major player in the EELS field [12]. As usual, the spectrometer started working correctly a day or so before EMSA.

By then Ondrej had moved from Berkeley to a faculty position at ASU. He didn’t like the heat and missed being close to good skiing, and went back to California after 5 years at ASU. His official ASU job duties consisted of doing research and running the ASU winter schools and workshops. However, he volunteered for teaching, and the electronics course he taught used the Art of Electronics as the textbook. It was based on the idea that physicists should be able to build useful electronics, and it was a major change from the somewhat outdated course based around simple transistor circuits.

Ondrej’s use of strong quadrupoles for magnifying spectra and later for imaging applications, implemented when he joined Gatan as its Director of Research, marked a turning point. Till then electron optics in electron microscopy, with a few exceptions, consisted of round lenses, deflection coils and weak quadrupoles used as stigmators. Use of non-cylindrical elements like quadrupoles, sextupoles (hexapoles), and octopoles is standard in accelerator design. Ondrej brought them into the mainstream of electron microscope design with the Gatan parallel EELS, in which strong quadrupoles were used to magnify spectra, and weak sextupoles to fine-tune second-order aberration properties [13]. The next stage was to use more quadrupoles and sextupoles (and later on also octopoles) to produce an imaging spectrometer, the Gatan Imaging Filter [14].

Although the use of quadrupoles and octopoles for aberration correction had been discussed from a theoretical point of view and tried in Cambridge, Chicago and Darmstadt, practical implementation typically floundered on the difficulty of adjusting the strength of all those elements so as to null “parasitic” aberrations. After Peter Swan retired, Ondrej found it difficult to continue with Gatan under the new management. In 1995 he left to go back to the Cavendish and develop an aberration corrector for the STEM. Mick Brown provided a hospitable environment and the Royal Society funded the development[15]. The result was the quadrupole-octopole Cambridge corrector, which demonstrated improved spatial resolution on the VG HB 501 in the Cavendish [16]. The computer code to make alignment possible was written by Andrew Spence, son of John Spence, and the procedure was based on a method outlined in Ondrej’s and Gary Fan’s 1992 paper on autotuning [17].

The Cambridge corrector was a proof-of-principle instrument that was not going to break any resolution records. The best way to transform it into a truly useful machine was to start a company, and Ondrej with his partner Niklas Dellby founded Nion (originally called Colibris) in the Seattle area, where his wife Angela came from. Ondrej liked the cool climate and the proximity to skiing in the Cascades, and sailing and kayaking on Puget Sound. He and Andrew Bleloch climbed Mt Rainier for his 50th birthday (after an aerial reconnaissance in my airplane!). Orders for aberration correctors came first from Phil Batson and soon after from Daresbury SuperSTEM and Steve Pennycook. It then became clear that aberration correction needed a whole new ultra-stable electron microscope, and that Nion should build it. The aberration correctors and the GIF already had as many (if not more) electron optical elements as most TEMs, so it was the obvious next step. Ondrej secured initial funding for it by calling up John Silcox and saying: “John, I think you should order an electron microscope from Nion,” whereupon John replied: “I was thinking the same thing.” The next step was a successful grant application to NSF, which provided $995k for the development, and the result was the Nion UltraSTEM.

Ondrej always saw the purpose of aberration correction as increasing current density and bringing down detection limits. A collaboration between Nion and David Muller’s group spectacularly demonstrated the power of the new instrument by atomic resolution elemental mapping of oxide interfaces [18]. The Daresbury group have gone further and shown differences in fine structure for single Si atoms with different coordination [19].

Since its earliest days analytical electron microscopy has been limited by energy resolution from the combination of spectrometer and electron source. Using a cold FEG gave a brighter source with a narrower energy distribution, but getting resolutions better than 0.3 eV was challenging. The Mook monochromator [20] in the gun developed in Delft was modified and commercialized by FEI and became widely available. With an energy resolution of 0.05-0.2 eV, it was more than sufficient for all inner shell fine structure studies (like the multiplets in transition elements and rare earths). It was still the case that EELS was limited to the UV (single electron excitations, plasmons) and soft X-ray regions (inner shells). Interesting features in the optical region (apart from plasmon resonances from nanostructures) such as optical emission from defects were undetectable due to the large background from the zero loss peak.

The problem with monochromation was high voltage (HV) instabilities as well as monochromator and spectrometer prism current wobbles, which is why some monochromators were tied to the HV supply. Ondrej introduced a new approach to monochromation: a ground-potential in-column filter that stabilized the HV by adjusting it so that the energy-dispersed beam would remain centered on the monochromator slits, and which also tied the spectrometer prism current in series with the current in the monochromator prisms [21,22]. Energy resolutions improved by almost an order of magnitude, and after Nion introduced an energy loss spectrometer also designed for ultra-high resolution, FWHM of the ZLP of under 3 meV has been achieved. The real benefit is not necessarily in the reduction of the width of the zero loss peak as much as in the reduction of the tails. It became possible to detect the signatures from isolated defects in the optical region such as NV centers, or color centers. Most importantly it opened up significant parts of the Infra Red spectrum to analytical microscopy [23], and made it possible to directly detect hydrogen bonded to other elements from the vibrational signature [24].

Where do we go from here? Nanometer scale vibrational spectroscopy opens up new possibilities in the study of polymers and biological materials, especially since it can be performed “damage free” at reduced spatial resolution [24].

Every user of a modern transmission microscope has benefited from Ondrej’s developments of technique and instrumentation. In an environment where the emphasis is on getting “results” rather than developing new instruments and techniques, Ondrej is one of the true pioneers who have shown that the results don’t come unless there are radical advances in instrumentation. I think despite his initial reluctance to form a company back in the late 1970’s, later on it became apparent that a small innovative company was the best way to create an environment where new machines could be dreamed up, designed and built. With a freedom that would not be possible in academia saddled with the constraints of the grant application process, and unlikely in established industry, which tends to frown on pioneering instruments with an uncertain future.

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