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Ondrej Krivanek's early scientific research☆

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ABSTRACT

In 1806, Humphrey Davey said that "nothing promotes the advancement of science so much as a new instrument". This paper reviews some of the lesser-known achievements of Ondrej's early career, and reminds us of the level of performance of instruments in those days, in order to appreciate how great has been the progress in instrumentation, much of it due to Ondrej and his leadership, since then. Some new results in the field of EELS are described, including extraction of the time-dependence of the dielectric response (with better time resolution than an X-ray free electron laser (XFEL)) from Nion EELS data. An approximation for atomic-resolution imaging which includes multiple scattering effects is given for biological samples, for use with aberration-corrected instruments when these become needed at the higher beam energies required to preserve the projection approximation, on which the 3D merging of single-particle cryo-EM images is based. We also discuss the requirements for out-running radiation damage using pulsed electron beams, a worthy final challenge for OLK.

1. Introduction

The early nineteen-eighties were an exciting time in the physics department at Arizona State University. Continuing the excellent Cavendish and CSIRO tradition of extended "morning tea" and coffee, John Cowley presided over lengthy discussions among the postdocs, faculty and students on their various projects, covering both theory and experiment. At various times, this group might have included Ondrej Krivanek, Sumio Iijima, David Smith, Ray Carpenter, Peter Rez, Johan Tafto, Neil Long, Mike O'Keefe, Gary Hembree and myself and our students and postdocs. These were the days of punched-card mainframe computer input with one-day turn-around, at a time when the Professors used the electron microscopes, working closely with the lab manager John Wheatley to keep the machines in top condition. Iijima, especially, was forever polishing gun components with John to extract more brightness from his etched pointed filaments, producing 0.34 nm resolution TEM images from the JEOL 100B (with special C_s=0.7 mm polepiece) always just a little crisper than anyone else's. (In 1980 he had published the first HREM image of a buckyball, long before their formal discovery, whose structure was correctly interpreted then by Mike O'Keeffe in our Chemistry department). This intimate connection with experimental work was a feature of academic faculty life, now mostly lost in this age of insistent email demands, deadlines and grant-writing, with funding agency rejection rates rising from about 75% then to around 90% now. John Cowley somehow managed to maintain this lifestyle to the end, and had worked on his VG HB5 on the day he died in 2005.

Ondrej burst upon the scene at ASU (from 1981 to 1985), full of his remarkable drive and energy, committed to making EELS a user-friendly experience with higher performance than soft X-ray spectroscopy, and often working with Christian Colliex and the Cornell group from an early stage. The ASU winter schools and conferences were soon in full swing (Figs. 1–3), bringing international leaders in the field to a Hot Springs desert resort in Arizona every year, such as Albert Crewe, inventor of the modern STEM, and an inspiring figure for us all, with a lasting influence on Ondrej [28]. Ondrej had come to join the faculty from his Berkeley postdoc with Gareth Thomas, and a string of publications on interfaces in materials science, a theme he continued at ASU. His 1977 paper with Kobayashi [24] on grain-boundary structure in germanium had showed the power of the HREM method for semiconductor physics and attracted much attention, and he produced some of the first high-resolution images of the Si-SiO_2 interface, precursor to a most critical area of device physics under Moore's relentless law. This followed his Ph.D. work with Archie Howie at the Cavendish (to which he came from Leeds in 1971, where he'd arrived from Czechoslovakia in 1968) on the structure of glasses, analyzed by an ingenious new technique [23]. As a post-doc in Oxford (1973–75) I remember seeing his paper [21] on the effects on lattice images of energy spread in the beam (showing that half-period fringes were unaffected), which, with his paper on Cs measurement [25] was highly relevant to the book I was then writing on HREM. This was my first encounter with OLK.

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Fig. 1. The ASU Castle Hot Springs conference in 1981.

Fig. 2. Ondrej (right), JCHS (in hat) and son Andrew behind him with friends sailing off the coast of Yugoslavia in 1990 . (Andrew worked for Nion for period).

Fig. 3. ASU persona around 1984. JCHS, Roger Graham, Al Higgs, Ondrej, Neil Long, Amanda Petford.

Our machine-shop at ASU was kept busy building serial-ELS systems to his designs using photomultipliers, with their excellent dynamic range, and the performance of these rapidly improved due to his skill in optimizing experimental conditions, and in instrument design. This led to the publication of the famous "EELS Atlas" with Channing Ahn, an astonishing achievement aimed at publishing the inner shell edges of practically every atom in the periodic table, which was done (eg by leaking hydrogen into the JEOL 200CX) over many long nights at ASU. Parallel-detection EELS systems were in development by several groups at the time since the appearance of the first linear diode arrays and one-dimensional charge-coupled diode arrays (CCD) (Jones, Johnson, Shuman, Kruit, Egerton - see [9] for a review) and all this work led eventually to Ondrej's design of the hugely successful Gatan parallel-detection EELS (PEELS) system, which has been in continuous development ever since, and is in use today. The electron-optical design, and subsequent design of the imaging filter, based on quadrupoles, allowed many of the ideas of later STEM aberration-correction systems to be developed. Ondrej's first PEELS paper with Ahn and Keeney [22] is one of the most highly cited papers in Ultramicroscopy. But several other lesser-known projects were on-going in parallel at that time which turned out to influence his future scientific career, and a few other unreported failures which I'll mention to save somebody time repeating them.

By good fortune, I had hired as my first postdoc Johann Tafto, from the powerful group of Jon Gjonnes in Oslo. Johann arrived full of his Ph.D. work on the theory of electron channeling, which I'd been simulating in the hope of reproducing on EELS spectra some of the standing-wave effects on X-ray fluorescence which Batterman had observed using X-rays at Cornell. But the EELS case involved double-channeling, difficult to interpret, and instead Johann conducted a brilliant series of experiments on channeling effects on EDX spectra from polyatomic crystals containing dopants. (Since multiple scattering affects host and dopant atoms equally, the effect cancels). This lead to our Alchemi technique for locating foreign atoms in crystals, used recently in the study of turbine-blade alloys [20] and ceramics [10]. But in a brilliant collaboration with Ondrej and his new spectrometer, they were able to combine the standing-wave effect with the sensitivity of near-edge structure (ELNES) to chemical valence for site-specific valence determination in iron oxide [47]. Johann went on to use the channeling effect to study localization in EELS by a clever experimental arrangement [43], solved the double-channelling problem (through reciprocity and a large beam divergence) and established the usefulness of ELNES with a famous series comparing spectra from octahedral and tetrahedrally coordinated Mg and K in oxides, showing how these could be distinguished [46]. All this provided an important market for future applications of Ondrej's wonderful spectrometers.

A second parallel development related to Ondrej's future involved my first Ph.D. student, Mark Disko, who took on a summer project with John Cowley in 1980 on Rochigrams, which eventually became crucial to the success of the Nion STEM. I'd become