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Invited Comment

My life and the world of crystals

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Abstract

This is an account of my life and my contributions to crystallography which have led to my receiving the 2015 Aminoff Prize. Periods discussed in this article are childhood influences, formal training at Harvard, life as an independent researcher at Bell Labs, starting the academic routine at Illinois and then London. Three major discoveries are presented in the form of anecdotes, on the silicon 7×7 structure, on crystal truncation rods and coherent x-ray diffraction. Much of my work has centered on the need for developing the instrumentation behind the intellectual steps, such as beamlines at the Brookhaven, Argonne, and Diamond synchrotron radiation facilities. This trend continues with the emergence of new possibilities for crystallography using x-ray free-electron lasers.

Keywords: beamline, surface, coherence, crystal, structure

(Some figures may appear in colour only in the online journal)

1. In Reading: kings, rivers, and Oscar Wilde

I was born in Reading, England, a town drawn like a leaf or a butterfly on UK maps and sandwiched between the rivers Kennet and Thames. I like rivers. In my youth, my parents used to take my siblings and me to many places by the Thames, and in some of our trips I would see dozens of locks, notice how easy it was to turn the cranks which so interested me that I even dreamed of becoming a lock keeper when I grew up. On our free days, my friends and I would go to the Thames, sometimes swim there, or row our canoes checked out from the local club.

I was fascinated by my town's history especially by the kings who walked its grounds. On some weekends, I would roam the ruins of Reading Abbey, adjacent to the rivers, founded by Henry I; when he died, he was buried in front of the abbey's altar. Among the ruins, it was easy to imagine the glory of those days when other kings would come to the abbey, kings like Henry II who met the patriarch of Jerusalem there. Henry III was also a frequent visitor, and it was there that Edward IV was married. But if the Abbey was built by King Henry I, another king, King Henry VIII would destroy it when he dissolved all the monasteries. Today, the ruined abbey still stands at the center of Reading, near the prison house where Oscar Wilde, the poet who made my town immortal in his poem 'The Ballad of Reading Gaol', was once jailed.

My parents are scientists themselves. My late father, Keith, was a physicist and a crystallographer; my mother, Mary, is a botanist. He worked at the Cavendish Laboratory at the same time as Crick and Watson, and when my parents got married, Francis Crick presented them with a tea table, a joint wedding gift from him and from many of those who worked at the Cavendish. Today, it is still in my mother's house in Reading; we fondly call it the (Nobel) table.

2. In my youth: my first crystals

When I was three years old, my father gave me a ball and stick model of atoms in a crystal. My mother said it became my favorite toy. Essentially, it was my first contact with crystals, but then I was just a toddler interested only in the model as a toy. I may even have played with the crystals,

Figure 1. Natural octahedral diamond of 84 carats. 'The Harvard Diamond Crystal' (Wikipedia).

banging them or biting them, or kicking them imagining them as balls. Life really is a marvel. How was I to know at that time, that one day I would spend a great part of my life studying crystals?

One lunch time when I was about eight, my father came home bringing a diamond sample which he borrowed from his colleague Henry Dyer, a large natural octahedron (figure 1), probably of several carats. I clearly remembered him showing it to me, but I did not only look at it; my father allowed me to hold that precious crystal, and holding it, looking at it, I was mesmerized! It was awesome. It filled half of my small hand; it felt rather heavy, and it reflected the light of the Sun. Truly, the look and feel of that Henry Dyer's South African diamond had made a lasting impression on me. My father took it back with him to work, so it was the first and last time I saw it, but for a long time, I would remember its almost perfect octahedral shape with natural facets that were still rough but were also so transparent that they glistened. That was my second encounter with crystals as a child. Later, in school, I would make models of 'polyhedra' from folded paper templates that I designed myself. The most difficult was a 'Mobius band' that was a continuous solid ring with a triangular cross section, but with only one surface, which took drafting skills to construct.

I was privileged to attend Reading School, in the days when it was a traditional 'grammar' school. It was very strict with full-suit uniforms, frequent detentions and mandatory haircuts, but the science teachers were excellent motivators of interest. Ed Bicknell was the mathematics teacher of the upper years. His style was very informal and insightful. His monthly 'maths projects' were so wonderfully creative that I can still remember half of them to this day: measuring food-container dimensions to learn calculus, length of daylight calculations to learn three-dimentional (3D) geometry, and working out the facet angles of the Platonic solids. The other great moment was a school lecture by Art Schawlow, who much later won the 1981 Nobel prize, demonstrating the first ruby lasers by shooting balloons from the San Francisco Zoo.

3. In Cambridge: a new life

In 1972, after completing the Cambridge entrance exams, I spent the last eight months in Reading working at ICL Dataskil, a branch of International Computers where work was rather easy, giving me plenty of time to read the first year textbooks ahead of my actual placement at Churchill College. I particularly enjoyed the 'Feynman Lectures' [1] and learned the story of DNA and the 'Central Dogma' from the physiology text [2]. In September 1973, I went to Churchill College on a scholarship, and in Cambridge, my new life began.

It was a big change … moving away from home, leaving my family, leaving my school friends, but it was also exciting, meeting new friends. The summer after my first year in Cambridge, I went to the USA on a sponsored working visa and had a chance to visit the Massachussetts Institute of Technology and Harvard University. I decided right then to take up graduate studies in Harvard, despite strong local advice that it would be a 'waste of time'. Back in Cambridge, that school year I took extra courses in Engineering. When summer came, I went to the Risø National Laboratory in Denmark to do a 10-week summer research project working with Jørgen Kjems on the design of a position sensitive neutron detector system, which may have been the last experiment ever conducted at the DR2 reactor. Later, I would write my undergraduate dissertation based on the result of that research. Then, I went to ILL in Grenoble, France, spending six more weeks doing research with John White on intercalation compounds using neutron powder diffraction. My local contact person was Christian Riekel, whom I would continue to know later at ERSF.

But Harvard was still in my mind, and so I applied there, took the GRE, and had an offer and a phone call acceptance. Taking biophysics was a big decision. At that time, I thought that physics was finished and that brain research was the future of science. The summer of 1976 was spent at CERN in Geneva, Switzerland, where I divided my time going to the library reading about biophysics, learning about particle detectors from my supervisor, and going on trips to the Swiss mountains. My final school year at Cambridge was quite intensive, with vacations spent studying the Part II Tripos with a 3 week revision period in isolation before 3 days of final exams. Burnt out after 2 days (4 exams), I could not concentrate on the last one but still got a 'first class' degree. Life was good.

4. Crystals in Harvard

The biophysics curriculum at Harvard, which still exists today, included three 3 month 'rotation' projects, which was an effective introduction to research. Other universities are only now starting to realize the value of this form of graduate training. Because it was in my original plan, I did the first rotation in neurophysiology, but immediately found out how hard the experiments were. I did a rotation in population biology and found out that theoretical ideas were still very

primitive within biology. Finally I did crystallography where, in a three month rotation project with Don Wiley, I studied hemaglutinin crystals, freshly grown in the laboratory, and took some of the first x-ray diffraction patterns. It was my third contact with crystals and my first time seeing them under a microscope. Don Wiley and Ian Wilson, a postdoc, later solved the structure, which has had a lasting impact on our understanding of influenza infection and its epidemics.

In the summer of 1977, I went back to Paris where I met Steve Harrison and his group doing the crystallographic phasing calculations of the Tomato Bushy Stunt Virus (TBSV) at the CECAM computation center there in Orsay. I decided to undertake my PhD with him as my adviser. My summer project was at CEA Saclay with Tom Ypsilantis (Berkeley) designing a particle detector, completing the work I started with him previously at CERN the year before.

I discovered that doing research is rather hard and very different from taking courses. So many things could go wrong without explanation. For three years, I experienced a general feeling of despair at the slow pace of discovery, but finally overcame it when I realized that I had learned a lot of things and that I could help others by sharing my accumulated knowledge with them. In 1978–80, I taught physics courses such as 'Introductory Electromagnetism' and 'Electronics' (the famous Physics 123) first as a TA (teaching assistant) and later as instructor. Physics 123 was the pride and joy of Paul Horowitz who developed it while he was writing *The Art of Electronics* [3], still a best-seller today. I redesigned the digital part of their electronics laboratory and introduced the first exercises to build a computer in class. Our lab notes were written up for publication as *Lab Manual for the Art of Electronics* in 1981 by Cambridge University Press [4].

Summer vacations were mostly taken in USA: Colorado, Utah, California. I bought my first car (a VW Beetle) in 1978, a major American right of passage, which enabled me to do ski trips to New Hampshire, often on Wednesdays for the $3 special at Mount Sunapee.

In 1979, at 23 and at about the half-way point to my PhD, I took the Qualifier Exam by writing a research proposal on something different from my PhD topic (a Förster resonance energy transfer study of protein folding of bovine pancreatic trypsin inhibitor). It was examined by Martin Karplus, who couldn't have had any inkling that in 2014, 36 years later, he would win the Nobel Prize. At 26, I had my PhD defense of my dissertation on the structure of the expanded state of the TBSV, with Harrison, Wiley and Bill Lipscomb (Nobel 1976) as internal examiners, and with Don Caspar (Brandeis, external). All of these people had a strong influence over my thinking, and I was proud to have learned from them. The crystallography community in the Boston area in those days was tightly bonded among the various universities and, by the end of my PhD, I felt like I was part of it.

5. From Harvard to Bell Labs

I met David Moncton before my PhD defense in late 1981. As member of Bell Labs Technical Staff, he went as a 'recruiter' to Harvard seeking graduating students for staff positions. Talking to him for about an hour, I convinced him that I was worth inviting for interview at Murray Hill. In preparation, I attended Moncton's own seminar, read his papers, then got ready for the Murray Hill interview. I also learned how one should dress for this type of interview.

At the interview, I remember that Steve Davey, who is still today part of the x-ray community, ran the movie projector for me to show Art Olson's movie about the structure, assembly, and phase changes of TBSV. Lots of questions were asked by Phil Platzman and Bill Brinkman about whether crystallography was even possible on proteins. In a heartbeat, I said yes: I had done it in Harvard for years, so it never occurred to me that it couldn't be done! That was my first introduction to the 'Bell Labs Seminar' style. The discussions went well enough for me to get a job offer on 15 October as a Member of Technical Staff (MTS) at an attractive $34 000 annual salary. Rick Freeman would be my first department head, a laser physicist, a pioneer in free-electron lasers, also new to the management job. Steve Chu (Nobel 1997 and future Secretary of Energy) and Phil Bucksbaum were in the same department. I accepted the offer but delayed my start date so I could take a vacation in Ecuador (Galapagos) and Peru (Machu Pichu), where it is a custom to drink coca tea because of altitude. When I returned from my vacation, I failed the medical test at Bell Labs, because the cocaine from the coca tea was still in my blood. Three weeks later, I was tested again and I passed. On Monday 1 December 1981, I started working at Bell Labs in Murray Hill, but the buildings were like a maze. I had no lunch for three days because I could not find the canteen. Ok, stupid not to ask!

On Friday 5 December 1981, it was arranged for me to fly to Stanford for three weeks of synchrotron experiments. The first one was with David Moncton, Robert Fleming and John Axe looking at diffraction from disordered Hg in 1D lattices within crystals. The second was with Paul Citrin doing surface extended x-ray absorption fine structure (EXAFS) studies with Fabio Comin, his new postdoc. The third was a surface x-ray diffraction (SXRD) experiment with Paul Fuoss and Sean Brennan using the vacuum chamber of Peter Eisenberger, shown in figure 2, taken over by Fuoss when Eisenberger left, and which he brought to Stanford. It was there that I was introduced to the low energy electron diffraction (LEED) pattern of silicon 7×7. All this took place before Christmas in my first month on the new job.

In January 1982, David Moncton left Bell Labs to take over the x-ray physics group at Brookhaven (BNL). Peter Eisenberger had left just before and they had all already agreed to take 2 ports (5 beam lines) at the new Brookhaven synchrotron, National Synchrotron Light Source (NSLS). Bell Labs now needed new people to pick up this direction and Freeman asked me to build a beamline at Brookhaven. In March, Fuoss left for Holmdel and Freeman gave me the old Eisenberger vacuum chamber (figure 2) to start my own SXRD experiments.

Moncton promptly hired two new postdocs at BNL, Doon Gibbs and Kevin D'Amico, to start surface diffraction there. They came for a day trip visit at Bell Labs, and they were impressed that I was completely open about sharing my

ideas and plans for surface diffraction. Since they too were supposed to build something similar, they were potential competitors. Looking back, I think that meeting might have helped start a precedent of openness in the synchrotron radiation (SR) community, which seemed natural to me at the time. Later on in 1984, when the beamline construction started seriously, I became one of the subtenants in Doon's rented house in Center Moriches. I had a bedroom overlooking the Great South Bay and would watch one of the neighbors take off by seaplane every morning to go to Wall Street.

6. Silicon 7 × 7 surface: the holy grail of surface science

For years, surface scientists had attempted to study the structure of the silicon (111) crystal surface with its 7 × 7 'reconstruction' but failed largely because they were using LEED, which uses electrons to create a diffraction pattern. Though the method results in great surface sensitivity, it could not solve the structure of silicon 7 × 7 surfaces because the necessary dynamical diffraction calculations were too difficult. Finding a solution for this, then, became the holy grail of surface science. Surely, I thought, there must be a method. At that time I first learned about the 7 × 7, SXRD was in its infancy. But I knew that if x-ray diffraction could be made sufficiently surface sensitive, the accurate kinematic structure factor measurements it could provide would allow the powerful methods of crystallography to solve the structure.

So, after seeing the elegant LEED pattern of the Si(111) 7 × 7 surface during my first visit to Stanford Synchrotron Radiation Laboratory (SSRL), I set the solution of its structure as a personal goal. Every three weeks at Bell Labs we held a seminar devoted to surface science. In those seminars, the 7 × 7, kept coming up with its 'milkstool' models and many more variations, and everyone talked excitedly about it; everyone in the room, it seemed, was applying their own personal techniques to solve its structure. It became clear to me then, why the structure of the 7 × 7 was the 'holy grail' of surface science, and I decided to set my sights on it with x-ray diffraction. I was quite dedicated to this goal and even decorated our bathroom in New York with a 7 × 7 design, as shown in figure 3.

By 1985, we were already building the X16A beamline at Brookhaven, but the x-ray ring of the NSLS was badly delayed in coming on line and had entered another long shutdown. Being at Brookhaven gave me the opportunity to collaborate with Peter Bennett, who was a postdoc working with Jack Rowe at Bell Labs. Peter also spent a lot of time at Brookhaven on the UV ring and I got to know him quite well there. During his PhD, Peter had studied the 7 × 7 with Barnie Webb (Wisconsin) and was similarly keen to solve the structure. Peter had already identified one of the key features of the 7 × 7, the 'stacking fault' that lies between the reconstructed layer and the rest of the silicon crystal over half of its 7 × 7 unit cell [6].

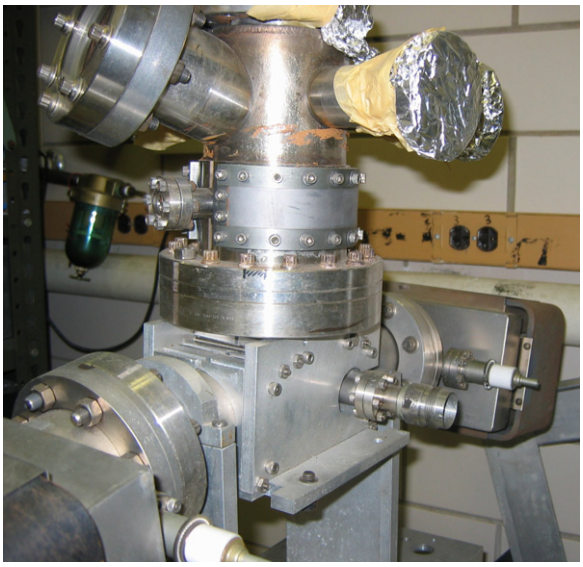

Figure 2. The vacuum chamber used for the first ever Surface x-ray Diffraction (SXRD) experiments carried out by Eisenberger and Marra just before I came to Bell Labs [5]. The x-ray-transparent Beryllium window can be seen around the waist of the chamber.

Figure 3. Sally Robinson with the 7 × 7 mosaic tile decoration of our apartment in New York.

Peter also knew that Kunio Takayanagi had built an ultra-high vacuum (UHV) electron microscope in Yokohama in the early 1980s and was developing the transmission electron diffraction (TED) technique that could be performed in such an electron microscope [7]. For thin enough samples, TED had the same kinematical diffraction data providing analogous information about the structure as x-ray diffraction. However, Peter and I believed that x-ray diffraction would be fundamentally more accurate at measuring the structure factors.

The other major revolution that swept through the surface science community at that time was the invention of the scanning tunneling microscope (STM) [8]. Because of the importance of the Si(111) 7 × 7 surface [8], it was the second sample ever looked at by Binnig and Rohrer with STM, and they saw a symmetric array of 12 bumps in the unit cell, which were later identified as 'adatoms'. These were the second of the three key elements of the final structure.

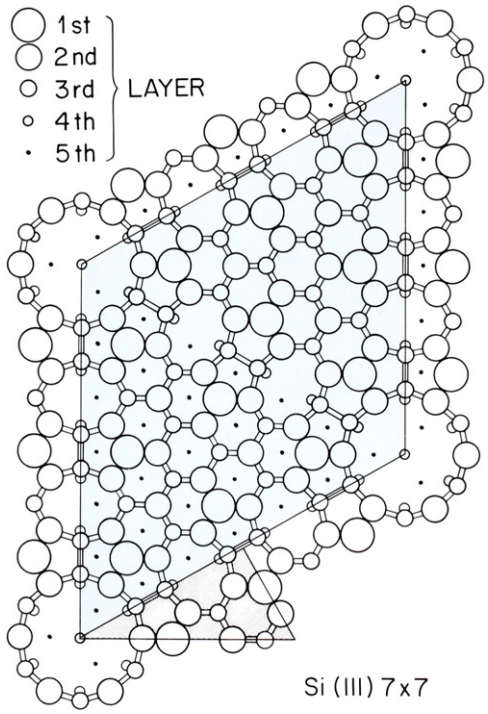

Figure 4. Full unit cell of the DAS model of the silicon (111) 7×7 structure, comprising 49 repeats of the underlying bulk crystal. Silicon atom heights are coded by the diameter of the circles. The crystallographic asymmetric repeating unit is the lower triangle. [11, 12, 14, 15]. Reproduced with permission from I K Robinson, J Vacuum Science Technology A6 1966 (1988). Copyright 1988, American Vacuum Society.

Meanwhile, I started working on the preparation method for the 7×7 with my Bell Labs colleagues and discovered it could not only be made on Germanium surfaces [9], it could also be preserved underneath an amorphous Si layer that conveniently avoided the need for UHV and lent itself to easier x-ray diffraction measurements [10]. I could immediately see the mirror symmetry of the buried 7×7 structure that was understood to be due to its stacking fault [10]. While we were waiting for the Brookhaven facilities to be ready, we designed and built a small portable UHV chamber for use at SSRL in Stanford, in order to develop the experimental techniques and the methods to be used in interpreting the results. This was in parallel with designing the permanent UHV chamber and beamline to do the routine SXRD measurements at Brookhaven.

However, while we were performing the Stanford experiments on silicon, the structure had already been solved by Takayanagi, using his TED methods [11]. As we were all hoping, it turned out to be a highly beautiful and extremely elegant structure, shown in figure 4, which he called the dimer-adatom-stacking-fault (DAS) model. The new feature that completed the model was the introduction of 'dimers': the other features were already known. These were identified in a classical crystallographic analysis of the Patterson function [12], which showed new interatomic peaks at short distances, pointing along new directions where bulk silicon has no bonds. The result caused great excitement in the surface science community and the paper has been cited over 1000 times [11].

We completed our Stanford experiments and confirmed the 'adatom' features of the 7×7 [13]. Then in 1988 we completed a full crystallographic dataset of the structure as the debut experiment for the full UHV SXRD setup at beamline X16A at Brookhaven, once the storage ring and beamline were fully debugged [14]. These structure factor data were substantially more accurate than the TED measurements because our data measurement was not affected by dynamical effects. This allowed us to complete a full crystallographic refinement of the atomic positions. There were clear distortions from the ideal bulk positions of the silicon atoms that yielded a systematic picture of strains in the structure [14]. Since then, there have been further advances in the structure refinement both by TED and SXRD, going to 3D data with high enough resolution to see bonding charges [15].

7. SXRD methods move out into the world

X-ray diffraction was my first interest, but at Bell Labs, I further developed it by combining it with SR and surfaces; thus I can say that SXRD was born in Bell Labs. As a method, SXRD has become the definitive technique for the determination of the atomic positions at surfaces and interfaces, completely displacing LEED This surface method is still used today at the major SR facilities, NSLS (Brookhaven), ESRF (Grenoble), APS (Chicago), Diamond (Harwell), Soleil (Orsay), DESY (Hamburg), Spring8 (Harima), and SLS (Villigen).

For developing SXRD, I was awarded two prizes for the surface structure work: the Warren Prize in 2000 and the Surface Structure Prize in 2011.

The years 1982–1990 then, were exciting times for me, years when I started several things simultaneously. At Murray Hill, I did my first SXRD experiment, studying Au(110), using the Eisenberger chamber, shown in figure 2, with the 60 kW Rigaku rotating anode generator, with the result published in *Physical Review Letters* in 1983 as my first solo work [16]. I collaborated with Paul Fuoss in building the first permanent SXRD system for use at Brookhaven, testing it over a period of two years at Holmdel, before moving it to Brookhaven [17]. I started designing pieces of the X16A beamline at Brookhaven, slowly working towards the structure of the Si(111) 7×7 surface. I helped design and build up SXRD at LURE in Paris and then at ESRF in Grenoble. Last but not least, I started doing experiments with the Danish group at HASYLAB and there discovered the crystal truncation rod (CTR).

The first sources of SR were parasitic on high-energy physics experiments, SSRL on the SPEAR ring at Stanford and HASYLAB on the DORIS ring at DESY. Both of these gave me opportunities for methods development in collaborative experiments. Between 1982 and 1987, I was allocated several periods of beam time at SSRL in collaboration with a number of scientists on separate occasions, including Gabriel Aeppli and Jakob Bohr on nitrogen on graphite;

Robert Feidenhans'l on silicon interfaces; Peter Bennett, and Peder Estrup, Bob Birgeneau, Mike Altman and with Warren Waskiewicz as technician on Tungsten. The portable vacuum system taken to SSRL was also used by Mike Altman to do his doctoral dissertation.

The NSLS in Brookhaven was the second of the 'second generation' storage ring facilities designed specifically for producing x-rays and Bell Labs was keen to capitalize on the research potential offered by a dedicated source. Things came together when beamline X16A and the permanent SXRD chamber started operating in 1988, as described below.

In 1987, Michèle Sauvage, a scientist from CNRS, invited me to help her set up her own SXRD chamber at the LURE synchrotron in Paris. Taking advantage of the NSLS shutdown, I went to Paris on a sabbatical leave for 8 months, staying at Place d'Italie and commuting to Orsay by RER to help Michèle set up her SXRD. I contributed a version of the SUPER control program [18], originally written by Robert Fleming which was ready to install at X16A. This survived for many years in both places. We obtained the first SXRD data on GaAs(100) surface structure using her new beamline. An important paper was published from that experiment in *Physical Review Letters* and is widely cited [19].

Then in the winters of 1990–93, I spent a 3 month block each year in Grenoble on a Chaire Municipale award to help design the SXRD instrumentation for ESRF during its build-up phase over 1989–94. I worked with Salvador Ferrer in designing the ID03 UHV surface diffractometer system, played with components for a fast Kappa diffractometer and interacted with the key team of scientists who would go on to lead the ESRF project.

8. At HASYLAB: the CTR

In the scientific world, new phenomena are always on the horizon. Sometimes they remain unimportant, unappreciated, until they are discovered. With discerning eyes, we are able to discover how they work, give names to them, and use them. Such is the case of the CTR. Considered an unimportant curiosity until I explained it, it has since sprung to life because of its many applications. In 1985, before NSLS turned on, I was collaborating on experiments at HASYLAB (DESY, Hamburg) with the Danish group composed of Jakob Bohr, Robert Feidenhans'l, Mourits Nielsen, Francois Grey, and Robert Johnson. The 'baby chamber' method of surface diffraction was developed by this group to look at indium antimonide (InSb) surfaces.

One night during my shift, while measuring InSb surfaces, the data ran off the page requiring me to extend the logarithmic graph paper with an extra sheet (on which the intensities were plotted); then it ran off again, and so, I started a third sheet. The intensities, as I noticed, just kept getting stronger and stronger as I got closer to the Bragg peak. This is now understood as a smooth crossing over of the CTR from the surface contribution to the bulk. But that night turned out to be an exciting one for me because I then realized that it is the cutting of the crystal which caused the rods in the diffraction. The quantitative agreement came a little later, by the inclusion of surface roughness in the theoretical description. I named that phenomenon, which my colleagues at that time called 'integer-order reflections', CTRs; expounded the idea behind it; and carried it forward.

In the morning, Robert Feidenhans'l came in for his shift and told me (facetiously) that I had just wasted the night measuring integer-order reflections when I could have measured the surface-specific fractional orders. I explained to him what I discovered, then tried to make him understand the significance of what I saw. We discussed it again over the next few days of beamtime. Buoyed by my discovery, I later wrote the ideas into a solo paper 'CTRs and surface roughness' which was published in 1986 in *Physical Review* B [20]. It was in this article where the name 'crystal truncation rod' was first mentioned; it is now often called by its acronym, CTR.

9. Beamlines X16A and X16C at Brookhaven

It was a great relief when the x-ray ring at Brookhaven finally completed its upgrade in 1988, allowing us to start working there full-time. The upgrade installed wigglers into the straight sections to provide higher power beams. Since 1983 I had been working on the design and construction of X16A, which was to be dedicated to SXRD. I designed many of the electronic control systems myself because there was nothing commercially available. The control computer was a PC running a Microsoft C version of SUPER [18], which we also maintained ourselves. The construction was a Bell Labs team effort with contributions from Paul Fuoss, Alastair MacDowell, Ed Melczer, Rick Levesque, Mike Altman, Warren Waskiewicz and Steve Davey.

We had completed building X16A at the very end of 1986. The NSLS's planned wiggler shutdown was delayed for a few months and that gave us the opportunity to test whether X16A would work. This was when Michèle Sauvage, who had already started building her own SXRD beamline at LURE in Paris, first came to see how the beamline worked. Further collaborations with Francesco Sette, Alastair MacDowell and Robert Feidenhans'l allowed us to complete our first test experiments by February 1987. The X16A beamline worked!

From 1988–1992, my regular routine was to spend 3 day blocks of time at BNL, twice per week. Shutdowns were usually on Tuesdays. Travel back to Bell Labs was once or twice per month, so I became a bit out of touch with the coworkers there. Walter Brown, my new department head, was always very supportive of this routine, but other managers wanted me to spend more time at Bell Labs.

Those early years 1988–1992 saw a high production of SXRD results from Brookhaven involving my first Bell Labs postdoc, Elias Vlieg. Many collaborators came to use the new beam line including Klaus Kern, Randy Headrick, Len Feldman, Ronan McGrath, Salvador Ferrer, Hartmut Zabel, Ed Conrad and Roberto Felici. We solved many crystal surface structures and started exploring phase transitions and

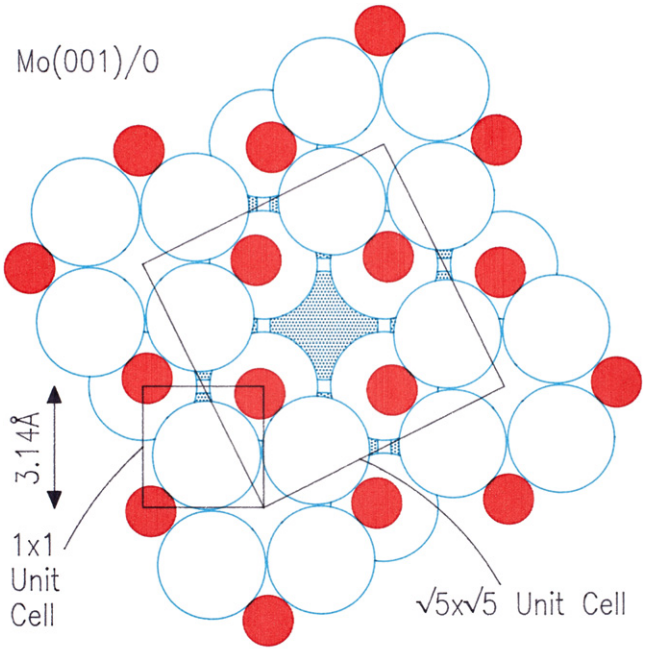

Figure 5. Structure of the Mo(001)/O $\sqrt{5}x\sqrt{5}$ reconstructed surface measured at X16A. Oxidation of the Mo surface causes the formation of clusters of Mo surrounded by oxygen. One Mo atom is lost per $\sqrt{5}x\sqrt{5}$ unit cell leaving a vacancy behind [25]. © IOP Publishing. Reproduced by permission of IOP Publishing. All rights reserved.

surface defects. One highlight was the study of the role of steps in the Pt(110) surface that explained how its missing row structure evolved from a 1×2 to a 1×3 reconstruction and were coupled to its phase transitions [21].

In 1992, Peter Eng became my second postdoc, starting first at Bell Labs and then overlapping with the Illinois period. Collaborators then at X16A were Detlef Smilgies (for one year on fellowship in 1992–93), Rolf Schuster (also for a one year fellowship, 1994–95), Peter Bennett, Holger Meyerheim, Harald Reichert, Helmut Dosch, and Ulrich Pietsch. Don Walko was my student who did his thesis work at X16A from 1996 to 2000. Then from 2002 to 2005, Sanjit Ghose became my third postdoc. Sanjit worked with Bob Averback and Peter Bennett on their regular visits to keep using X16A. Peter Eng and Detlef Smilgies undertook studies of the Mo(001) surface which was very challenging for the UHV system of X16A, which achieved its peak performance during this period. With the help of these collaborators, I studied such crystallographic curiosities as adsorbate-induced faceting [22], anisotropic surface vibrations [23] and surface ordering of alloys [24]. An example of one of the many surface structures determined during these years is the Mo(001)/O $\sqrt{5}\times\sqrt{5}$ reconstructed surface shown in figure 5 [25].

We also built up X16C in 1992, a second beamline at NSLS for EXAFS and general diffraction experiments. This was organized by Alastair MacDowell who used a new monochromator designed by Paul Fuoss [26]. While I was in Grenoble, in 1990, I started designing a kappa-geometry diffractometer, then called the 'Goniomètre a Grande Vitesse'

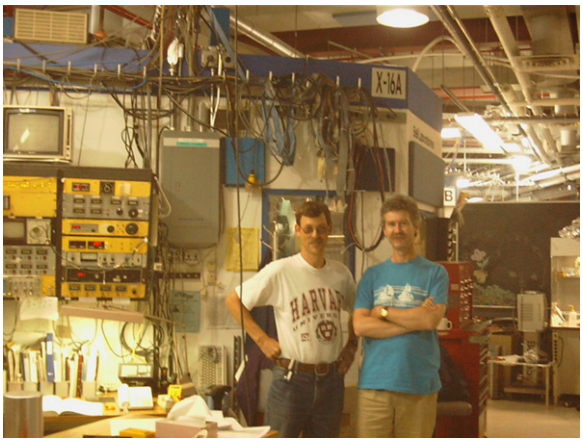

Figure 6. Peter Bennett and myself standing in front of the X16A control station around 2004.

(GGV) for use at X16C. Its high speed came from the first use of direct-drive servo motors [27]. This was fabricated in the Illinois machine shops in 1993 and delivered to Brookhaven and tested in 1994 [27]. It served well for training students and developing SXRD ideas for surfaces and interfaces outside vacuum. Arunabha Ghosh helped assemble the instrument and devised optical methods of aligning it. Yong Chu developed x-ray diffraction for electrochemical interfaces and completed his PhD studying these [28]. Dave Fanning studied ferroelectric crystals which he grew using strategic doping and helped develop the diffraction anomolous fine structure (DAFS) technique for studying them [29]. Chinkyo Kim examined strain transfer effects between thin films of electronic materials [30]. Sébastien Boutet observed the first CTRs from protein crystals [31]. X16C was a self-organized project and was amicably shared with Dave Adler (1993–8), Anatoly Frenkel (1998–2003), Matthew Marcus and Alastair MacDowell (1985–99).

A fact of life is that good things too can end. Around 2006, X16C was taken over by Stony Brook for use as a powder diffraction beamline. X16A was taken over by a BNL group from NSLS-2 sometime in 2008 and the SXRD activity there ended. Finally, the whole NSLS facility closed permanently in September 2014. One of the last pictures of X16A while it was still active is shown in figure 6.

10. Switzerland or USA?

It was in 1987 that I met my future wife, Sally Calong-Robinson, whose doctoral degree is in Literature. I talked to her about a Japanese novelist and asked her if she wanted to see a Japanese play, and that secured a yes answer from her. We got married in 1989.

Meanwhile, the long-term funding at Bell Labs was threatened by the 1984 divestiture of the 'Baby Bells' from the parent company, AT&T, which lost its virtual monopoly of the telephone network. I decided it was time to look for a university position as I had sufficient success to be able to expect to go straight into a tenured teaching position.

In 1992, I got two offers: one from Switzerland and another from Illinois. After careful consideration, weighing the pros and the cons for each, we decided to accept the offer from Illinois, and we moved to Urbana, a university town, in August of the same year. In New York City, we lived in a one bedroom, two-bathroom apartment, as seen in figure 3; in Urbana we bought a huge 400 m^2 house for a very reasonable price.

After moving to Illinois in 1992, Sally and I would go to Brookhaven every summer, driving East in May and West in August. We would usually take an apartment on the BNL site for 1–2 months. The UHV SXRD diffractometer [17] on X16A would be exchanged with a different one operated by Ken Evans-Lutterodt once per year and this equipment could be swapped over quickly within a single day. On X16C we would install the kappa diffractometer [27] for each experiment, usually 2–3 weeks at a time. Aligning this was a good routine for the students.

At the University of Illinois, I became active in learning to use the high degree of coherence produced by the new 'third generation' light sources, ESRF in Grenoble starting in 1994, APS at Argonne in 1995, and Spring8 in Harima. These third generation sources use magnetic undulators to generate x-ray beams that are thousands of times brighter than the second generation, such as Brookhaven. New ideas were needed to harness the coherence that follows from this brightness and apply it to the study of everyday things like materials or biology. An early breakthrough, still using a second generation source, was the observation of x-ray 'speckle', analogous to its optical equivalent seen with laser beams [32].

ESRF (Grenoble) came on line in 1994 and I joined with a group of colleagues including Jens Als-Nielsen, Ron Pindak, Robert Fleming and Steve Dierker to try its new capabilities. We were ready with test samples and motorized pinholes to look at the 'speckle' they would generate, but on the first occasion we were given just a single day of beamtime on ID10. In spite of this, already starting with disordered multilayer samples, I was able to understand their behavior by modeling the disorder to obtain crude images from that single measurement [33]. This was the first example of coherent imaging with x-rays, published in 1995.

Moving steadily forwards, my Illinois group started to develop the slit hardware needed to control the small beams needed to achieve coherence and to invent the analysis methods needed to obtain images. Jeff Libbert was the first postdoc and John Pitney the first student to try this. Ivan Vartanyants came to visit us in Illinois from Moscow for 3 months in 1997, then every year for 9 months from 1998 to 2004. We came to Troika beam line (ID10) of ESRF to image etched silicon surfaces, looking at their surface morphology after treatments that change their structures on the nanoscale [34]. We learned how to invert the coherent diffraction patterns by reading the papers of Jim Fienup from the optics literature, which showed that they could be inverted, i.e. solving the phase problem, simply because they were oversampled. These ideas had also been discussed by David Sayre back in 1952, immediately after the publication of the Shannon theorem [35].

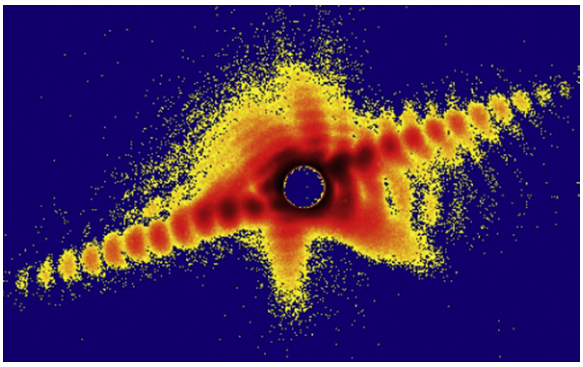

Figure 7. Coherent x-ray Diffraction pattern taken from a small gold nanocrystal at the UNICAT 33ID beamline of APS. The dark circle in the center is the shadow of a beamstop used to protect the CCD camera [36].

Another breakthrough moment took place in the free-flowing discussions at one of our group meetings in the Materials Research Laboratory in Urbana. I knew that slits can diffract a laser beam into a fringed pattern and started to wonder what the coherent x-ray diffraction (CXD) pattern of a small crystal would look like; I concluded it would look the same and clearly remember sketching this on the blackboard. We decided to test this idea using the new 33-ID beamline run by the University of Illinois at APS. My students Garth Williams and Mark Pfeifer heated films of gold until they broke up into small crystals, and when these were placed in the coherent beam they gave beautiful fringed diffraction patterns, like the one shown in figure 7. These patterns could be inverted into credible images of the crystals, but the quality was limited by beam stability and imperfect coherence [36]. The CXD method was then born.

Ivan Vartanyants and I had many discussions about the potential and limitations of CXD. We tried to understand how the partial coherence of the beam would affect the result and how dynamical effects would appear in the images. We thought about new phasing algorithms. Above all, we discussed the sensitivity of CXD to crystal strains [37]. As a result of those discussions, I started campaigning for funding to build a dedicated CXD beamline at APS, in order to continue developing methods of using the very high coherence for direct 3D imaging of structure. The potential application was its ability to examine strain distributions inside complex materials on the nanometer length scale.

One valuable opportunity offered by the University of Illinois was the chance to take regular sabbatical leaves, typically for 6 months every three years. As Urbana was a little isolated, this offered me the chance to get back into active research environments in other places. In 1996, I spent 3 months with Wolfgang Moritz in Munich, playing with an interesting idea to use CTRs as a reference to solve the crystallographic phase problem for the surface layer. This worked well enough eventually to lead to a publication [38]. In 2004, I divided my sabbatical between the Nagoya

University Venture Business Laboratory (VBL) and the Max-Planck-Institut für Metalforschung, Stuttgart where I was awarded a Humboldt Foundation Senior Research Fellowship. The VBL work with Masao Tabuchi also continued the CTR-based phasing work and led to another publication [39]. In Stuttgart, I looked at oxidation of Pt(111) surfaces with Andreas Stierle.

11. The 34-ID beamline at the advanced photon source

My grant to build sector 34-ID at Argonne was announced in 1999 by the National Science Foundation. Howard Birnbaum as MRL director arranged the matching funding and the project was incorporated into the UNICAT consortium headed by Haydn Chen. Curtis Kenney-Benson was hired to help me design the beamline, order the components and coordinate the writing of its 'Preliminary Design Report' and 'Technical Design Report' required by Argonne. My students Garth Williams, Mark Pfeifer, Sébastien Boutet, and Tommy Angelini spent the summer of 2002 actually building the beamline. As at Brookhaven before, we bolted all the components together ourselves and wired up the 'Equipment Protection System', under the guidance of Pete Jemian. Paul Zschack, as the manager of UNICAT, interfaced our project with the existing sector 33 beamlines and with the Argonne management.

Construction completed, the operation started in 2003. The first experiments were conducted by Sébastien Boutet used the pink beam to look at damage to protein crystals, for small-angle scattering by Gerard Wong and Tommy Angelini, and speckle from thin films in the grazing incidence small-angle x-ray scattering (GISAXS) geometry by Wei Zhang. A monochromator was later added using an economic water-cooled design, which was not in our original budget [26]. We started using diamond monochromator crystals, but found they were not good at preserving the beam coherence; later we switched to silicon. Though these developments, we ended up with the only water-cooled silicon monochomator at APS, a component which is still operating successfully today.

Because of its potential application to the study of surfaces using CXD, we then designed and built a UHV chamber directly into the beamline. We also had to include very delicate UHV roller-blade slits, fabricated by Alex Nozko, to select the coherence just in front of the sample. This led up to the first UHV CXD experiment in 2003 by Mark Pfeifer and Garth Williams in which they grew lead nanocrystals *in situ* in the beamline. This experiment was the first to image strains, illustrated in figure 8, a major breakthrough that was published in Nature in 2006 [40]. This defined the beamline and established the Bragg coherent diffraction imaging (BCDI) method.

Initially, the operation of the 34IDC beamline was paid by the University of Illinois through UNICAT, but transferred to the Department of Energy in 2006. Garth Williams, Mark Pfeifer, Sébastien Boutet and later student Mengning Liang all continued on in the CXD field. Sébastien Boutet, Garth Williams and Mengning Liang became founders of the new x-ray free-electron Laser (XFEL) field (below). My postdoc at that time, Ross Harder (2004-7), continued into the UCL funding period then transferred onto the Argonne staff in 2007.

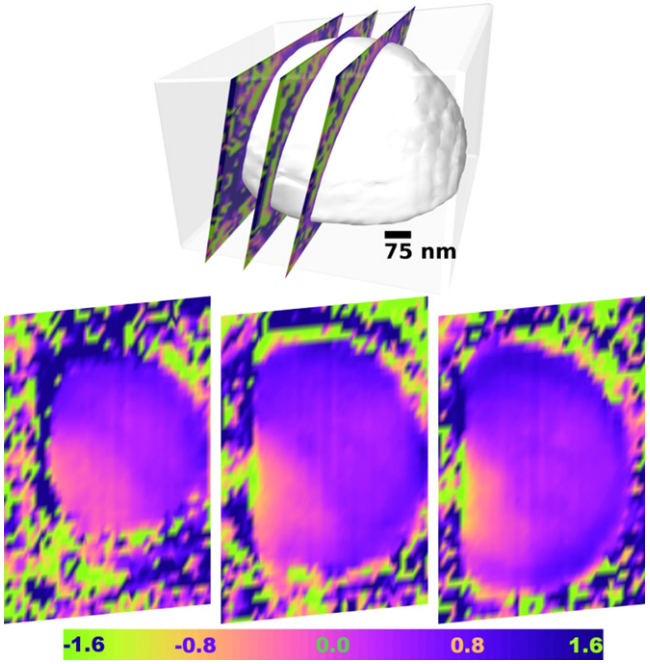

Figure 8. Images of strains inside a 700 nm hemispherical lead nanocrystal obtained at sector 34-ID of the Advanced Photon Source (APS). The strain projection (color) is associated with contact with the silicon substrate upon which the crystals were grown [40].

This story illustrates the convergence of theoretical ideas, methods development, the involvement of large facilities and, above all, teamwork required to bring about revolutions in science in the modern era. BCDI was pioneering work aimed at developing new SR-based techniques by using the simple fact that the light waves are in phase with each other. I originally named this CXD, but the name evolved into BCDI to incorporate the role of imaging and the crystallographic concept of Bragg diffraction from crystals. Over an entire century, starting with the Braggs, diffraction based methods have succeeded in three-dimensional imaging of materials, under the title of crystallography. My contribution to this story was to demonstrate how it is possible to obtain three-dimensional representations of deformations and defects in nanomaterials.

12. Nanotechnology in London

In the summer of 2002, Gabriel Aeppli recruited me to move to London where he was already Director of the London Center for Nanotechnology (LCN). He was my friend and colleague from Bell Labs, where he had started as MTS at the same time as me. Sally and I were strolling down Third Avenue after dinner one evening that summer and spotted Gabriel in another restaurant. This chance encounter was to

change my life, when he told me he was moving to London and would have some new faculty positions opening. I applied and interviewed in April 2003. The position was originally listed as an opening for neutron diffraction but this was broadened to include x-ray diffraction as well. In the end two professorial appointments were made, one for myself, the other for Des McMorrow. My appointment was at University College London in both the LCN and the Physics and Astronomy Department as Chair of Physics. A 5 year half-time visiting appointment at the Diamond Light Source (DLS) as a 'Diamond Fellow' was arranged by Gerd Materlik, its director and CEO. 2003 was an interesting and exciting year, a time to believe that everything is possible!

It was in London where the CXD methods developed. New opportunities lay with the DLS a third generation synchrotron located at Rutherford Appleton Laboratory (RAL) near Oxford. My research at UCL has been largely tied to this development, although we continue to do experiments at ESRF, SLS and APS during the buildup period. Following the DLS appointment, I became a founding 'Diamond Fellow' of the Research Complex at Harwell (RCaH), also located at RAL. This is a meeting place for scientists interested in the transfer of methodologies from the physical to the life sciences. Materials and biological imaging are the main directions under development there. Through these connections, I have been developing methods of using the very high coherence of the latest SR sources to enable direct 3D imaging of structure.

Three major grants supported the work of my group, divided between the UCL and RCaH centers. The first, entitled 'nanosculpture', looks at strains induced in nanometer-sized crystals either synthesized from atoms in a'bottom up' procedure, or else carved by lithography from bulk materials in a 'top down' approach. The second is to study the structure of the human chromosome by coherent x-ray imaging methods. The third is to develop new x-ray imaging methods based on deliberate modulation of the phase by suitably developed x-ray optics.

My first UCL project was to perform BCDI on nanowires. Steven Leake, Ross Harder and Marcus Newton looked at ZnO nanowire structures. Steven's most important result was to find that the apparent coherence was dramatically different from one Bragg reflection to another, explained as an effect of the rarer longitudinal coherence of the beam [41]. The 'nanosculpture' grant was coordinated with a partner user proposal (PUP) running from 2009 to 2011 with Argonne. We introduced a new confocal microscope to the beamline to select and align known nanostructures in the beam. In this way, we developed the methods of using multiple Bragg peaks from the same nanocrystal. There were important results at 34-ID-C from Moyu Watari, Marcus Newton, Ross Harder, and later Jesse Clark and Gang Xiong.

Building on this success, we renewed the PUP to start developing x-ray ptychography [42] during 2011–14. We provided new precision piezo scanning stages to 34-ID-C and supported Xiaojing Huang's postdoc at Argonne. Many more papers emerged in the course of developing the method, accounting for the results of the experiments done by Felisa Berenguer, Jesse Clark, Gang Xiong as postdocs, Richard Bean, Laura Shemilt, Xiaowen Shi, Nicolas Burdet, Marianne Monteforte, Maria Civita, Ana Estandate and Chris Lynch as students. Like before at Brookhaven, we had the first beamline dedicated to a promising new technique which attracted prominent visiting scientists during this development period: Franz Pfeiffer (2003), Virginie Chamard (2004), Lorenz Stadler (2004), Roberto Felici (2005) and Hyunjung Kim (2008-pres), all coming to see what we were up to on the new beamline and take home ideas. Throughout both PUPs, Ross Harder was fully involved with both the science and the technical improvements of the 34-ID-C beamline.

Meanwhile back at Diamond, I had prepared Beamline Proposal 048 'A CXD and XPCS Beamline for the DLS' for presentation to the Science Advisory Committee in November 2004. This was accepted as part of its Phase II construction. Christoph Rau was hired as the Principal Beamline Scientist and construction began in 2007. Under his direction, the beamline evolved into the longest beamline in Europe with two parallel branches, one for tomographic imaging and one for coherent diffractive imaging (CDI). BCDI was catered for by the use of giant robots to carry the area detectors needed to reach the Bragg peaks with enough distance from the sample to allow oversampling. My current group consists of postdocs Fucai Zhang, Jörg Schwenke, Bo Chen, Mohammed Yusuf and Graeme Morrison. They have used the CDI branch on a number of occasions for looking at chromosomes, with fresh samples prepared in the RCaH laboratory next door, and for developing new modulation-based imaging methods.

Last, but not least, my first XFEL BCDI experiment was done at Stanford's new Linac Coherent Light Source (LCLS) facility in November 2011 with Jesse Clark, Ross Harder and a large group of other scientists and Stanford staff. 60 h of XFEL beam led to two important results [43]. Using the extremely short x-ray pulses from the LCLS, we were also able to show how one can excite motion (phonons) of the atoms in individual nanoparticles and follow how these movements propagate in the particles. These new directions, opened up by XFELs, point the way to the future for me: time domain crystallography for the discovery of new materials.

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