Pure Appl. Chem., Vol. 77, No. 7, pp. 1089–1106, 2005.
DOI: 10.1351/pac200577071089

From a Meccano set to nano meccano*

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Abstract: The hurly-burly life of a scientific nomad is traced through thick and thin from the Athens of the North to the City of Angels with brief and not so brief interludes on the edge of the Canadian Shield, in the Socialist Republic of South Yorkshire, on the Plains of Cheshire beside the Wirral, and in the Midlands in the heartland of Albion.

Keywords: catenanes; mechanical bond; molecular electronics; molecular machines; molecular recognition; noncovalent interactions; rotaxanes; self-assembly; supramolecular chemistry; template-directed synthesis.

SOURCES OF EARLY INSPIRATION

It is almost 70 years now since Linus Pauling's classic, *The Nature of the Chemical Bond,* broke upon the scientific scene, bringing much-needed rhyme and reason to chemistry. It probably did more than any other single monograph [1], published during the 20th century, to set the agenda for chemists of all persuasions. The emphasis that it placed on covalent and ionic (electrostatic) bonds has dominated the machinations and actions of chemists for the best part of six decades. As the 20th century was drawing to a close, however, the sentiment that the chemistry of the covalent bond no longer automatically commanded the intellectual high ground in chemical synthesis was already beginning to be alluded to by leaders in the field of organic chemistry, such as Dieter Seebach [2]. In his 1990 review, *Organic Synthesis—Where Now?*, the Zürich scholar comments that "we should take the risk of attacking more complicated systems, ones whose structures and properties are determined by noncovalent interactions."

Following rapidly on the heels of Charles Pedersen's discovery [3] of the crown ethers and their remarkable ability to form complexes [3–5] with organic cations, as well as hard metal cations, chemistry started to spread its wings beyond the molecule with the emergence and growth of what Don Cram [6] called "host/guest chemistry" [7] and what Jean-Marie Lehn [8] described later as "supramolecular chemistry" [9].

This essay is a personal account of how my own interest in host/guest and supramolecular chemistry gradually evolved over two decades into a fascination with the chemistry of the mechanical bond. I have chosen purposely to tell this particular story without any supporting illustrative material for two reasons. One is that the literature to which I am referring in this essay reproduces it in abundance. The other is that anyone who cares to take this essay seriously will be obliged to actually read some prose! This is an exercise which, it would seem to me, is being avoided increasingly by scientists of all ages and disciplines. Undeniably, as the old adage reminds us, a picture is worth a thousand words. But, as

Pure Appl. Chem.* **77, 1087–1296. An issue of reviews and research papers based on lectures presented at the 15th International Conference on Organic Synthesis (ICOS-15), held in Nagoya, Japan, 1–6 August 2004, on the theme of organic synthesis. This paper is based on the acceptance lecture for the Nagoya Gold Medal.
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a scientist who has relied heavily on their impact, particularly color ones, I also recognize the dangers that they can pose to the art of precise, yet measured scientific understanding, discussion, and debate, let alone to scholarship.

The only son, indeed child, of a lowland Scottish tenant farmer, I was raised during the 1940s and 1950s in a post-World War II society dominated by the rationing of food, clothes, and petrol (gas), and lacking in the conveniences of modern-day living. The farmhouse, farmstead, and cottages were not connected to the national electricity grid until my last year in high school (1959–1960). The result was that I not only had to adapt to living a very simple lifestyle, but I was also obliged to play games and identify leisure activities that I could pursue and enact solo. An insatiable appetite for doing jigsaw puzzles gradually gave way to playing with one of the most sophisticated toys of the day in the form of a Meccano™ set—or an Erector set in the United States. It was a foretaste for the development of my skills in my teenage years as an amateur farm mechanic and engineer on the first generation of tractors and the second generation of motor cars—automobiles in the United States—not to mention farm implements and other contraptions of every conceivable description for tackling a range of tasks that seemed countless at the time. The most impressive of these machines and gadgets were highly adaptable and could be modified in a relatively simple fashion to accomplish different tasks. I ascribe my early interests as a chemist in stereochemistry [10] and topology [11] to my great love of solving jigsaw puzzles. I attribute my attraction to the mechanical bond in chemistry to the influence that Meccano sets and farm machinery had on me during these formative years. A mixed arable farm provided the preparation for multitasking and a yearning to try to be a master of as many trades as possible in science and beyond. A spirit of adventure and a readiness to take risks was encouraged by the vagaries of the climate and the unexpected nature of the weather all the year round in the lowlands of Scotland. And so it was from this university of life on Edgelaw Farm and a more formal high-school education at Melville College Edinburgh that I set out to train as a chemist at Edinburgh University, starting in 1960.

MY APPRENTICESHIP IN CHEMISTRY

My earliest research experiences, first as an undergraduate student and then as a postgraduate one, were in the unraveling of the primary structures of plant gums of the *Acacia* genus [12]. This exercise left me one lasting impression, namely, that of all the many gum trees then present in the Sudan—from whence my starting materials came—they had never managed to produce between all of them any two gum molecules which were actually identical in size and constitution, one with the other! I longed for a little more precision to my chemistry, at least for a time. During my Ph.D. degree, I interacted with my professor, Sir Edmund Hirst, on no more than two occasions—once when I began my research in 1964 and finally at the defense of my thesis in late 1966 before leaving early the next year to spend almost three years in Canada as a National Research Council Research Fellow with Ken Jones at Queen's University in Kingston, Ontario. I left Edinburgh with Sir Edmund's words, "Whatever you do in research, Stoddart, make sure you work on a big problem", ringing in my ears. I was not sure what a big problem was but I was determined to heed his advice somehow to the best of my limited ability. It was so fortunate in this regard that I was visited by a miracle during my postgraduate years at King's Buildings. For it was there in 1966 that I met my wife-to-be, Norma Scholan, and that turned out, in the fullness of time, to be a blessing beyond belief until the ravages of breast cancer, radiotherapy, and chemotherapy stole her away from me in early 2004 after a struggle that consumed a lot of our time and energy for the preceding 12 years.

No sooner had I arrived at Queen's in 1967 than did Charles Pedersen's seminal paper [4] on macrocyclic polyethers appear in the *Journal of the American Chemical Society*. This paper inspired me to get involved, during my postdoctoral years, in the synthesis of chiral crown ethers from carbohydrate precursors [13,14]. It was this activity that was subsequently to dominate [15,16] my independent research activity as a lecturer in chemistry at Sheffield University from 1970 to 1978. I had returned to

the United Kingdom from Canada in 1970, supported by an Imperial Chemical Industries (ICI) research fellowship, and this piece of good fortune allowed me to establish collaborations with numerous ICI researchers under the auspices of several Cooperative Awards in Science and Engineering (CASE) from the then Science and Engineering Research Council (SERC). It was with generous support from the SERC and additional financial support and much encouragement from a number of ICI's senior managers, including Tom McKillop, Bernard Langley, and Warren Hewertson, that I joined the ICI Corporate Laboratory in Runcorn, Cheshire in 1978 on a three-year secondment.

THE RUNCORN REVOLUTION

There, I met a brilliant young chemist, Howard Colquhoun—now Head of the Chemistry Department at Reading University—who had just joined the catalysis group, and together we started looking at the ability of transition-metal ammines [17], including cisplatin [18], to form adducts with 18-crown-6 and other crown ether derivatives [19]. There were already quite a number of reports in the literature on the complexation of primary alkylammonium ions by 18-crown-6 where [N^+–H···O] hydrogen bonding was the source of a good deal of the stabilization energy between the guest ions and the neutral host [20]. When Howard alerted me to the fact that there was a treasure trove of transition-metal ammines in the basement of the laboratory that had been accrued by Joseph Chatt when he was an employee of ICI in the 1950s, I could not believe our luck. During the next few years, we were able to give a new lease on life to Werner's concept of second-sphere coordination [21–23] by working closely with X-ray crystallographer David Williams at Imperial College, London. We were to discover [24] that a dicationic platinum complex carrying a 2,2'-bipyridyl ligand, in addition to two *cis*-ammine ligands, was engulfed in an intriguing manner by dibenzo-30-crown-10 (DB30C10), both in solution and in the solid state. What happens is that the polyether loops of the crown ether, having done their best to interact with the two *cis*-ammine ligands via [N^+–H···O] hydrogen bonds, leave the two π-electron-rich catechol rings to position themselves in a π-π stacking mode with respect to the π-electron-deficient bipyridyl ligand. It did not take a lot of imagination and just a little encouragement from a former ICI research scientist, Eric Goodings, to replace the transition-metal complex as the guest by the all-organic Diquat dication where the nitrogen atoms of the 2,2'-positions of bipyridyl ligand are substituted by a bridging bismethylene unit. The outcome was just as expected. The Diquat dication was found [25] in the solid-state structure to be slotted into the U-shaped cavity of DB30C10, such that numerous [C–H···O] interactions are supplemented by π-π stacking interactions between the π-electron-deficient bipyridinium unit and the two π-electron-rich catechol rings of the crown ether host. Both the 1:1 adducts [24] with the metallo-organic dication and the 1:1 complex [25] with the Diquat dication are reasonably stable species in solution, as indicated by the presence of diagnostic charge-transfer bands that render the supramolecular entities yellow and orange, respectively.

BACK IN THE SHEFFIELD DAYS

I returned to Sheffield University in 1981 with the knowledge that ICI market Diquat in admixture with Paraquat worldwide as a wipe-out weed-killer. I was determined to find a good receptor for this particular bipyridinium herbicide as well. The Paraquat dication is also known as methyl viologen, i.e., a 4,4'-bipyridinium unit carrying methyl groups on both its nitrogen atoms. The path toward identifying a good receptor for the Paraquat dication was far from an easy one. My initial receptor designs were not only much too complicated, but they also proved to be complete failures when it came to attempting to complex the Paraquat dication. I believe it was Noel Coward who said, "The secret of success is the capacity to survive failure." And so I preserved but also changed my strategy from trying to be too clever to letting the molecules do the work for me. And indeed, the answer lay in a very much simpler crown ether that is a constitutional isomer of DB30C10 and was first synthesized for a completely different reason in the Cram group [26] at UCLA—namely, bisparaphenylene-34-crown-10 (BPP34C10). The

X-ray crystal structure of the 1:1 complex formed between BPP34C10 and the Paraquat dication revealed [27] that the guest threads its way centrosymmetrically through the former in the most suggestive of manners with respect to subsequent catenane and rotaxane formation [28,29]. The fact that the 1:1 complex is stable in solution told us that we had uncovered in these supramolecular species, which I subsequently called [30] a [2]pseudorotaxane, a template that could be employed subsequently to assemble catenanes, as well as rotaxanes.

It soon became obvious to me, as the 1980s unfolded, that we needed to understand molecular recognition and self-assembly processes involving π-donors and π-acceptors in a lot more depth. We had established [31] that we could thread a π-acceptor through a ring containing π-donating units. Next, we needed to reverse this recognition motif, making π-donors the threads and locating the π-acceptors—for example, two bipyridinium units—in a cyclophane-like macro-ring. And so we became focused on the synthesis of the tetracationic cyclophane, cyclobis(paraquat-*p*-phenylene), which Mark Reddington eventually obtained in a two-step synthesis, starting from 4,4'-bipyridine and xylylene dibromide. In the second step, which includes the all-important closure of the macro-ring to afford the cyclophane, the best that Mark could obtain yield-wise was 12 %. During the act of publishing the first two communications [31,32] in *Angewandte Chemie* on this tetracationic cyclophane, I learned that Siegfried Hünig at the University of Würzburg had also been engaged busily in the making of this and other closely related tetracationic cyclophanes [33] for quite different reasons. Since the two 1988 communications [31,32] coincided with the beginning of my use of color—red for π-donors and blue for π-acceptors—cyclobis(paraquat-*p*-phenylene) became known as the blue box and has gained considerable notoriety subsequently as a promiscuous host for π-donors of many different complexions. In recent times, we have discovered [34], using templation, how to produce it in a much more highly efficient manner.

The stage was now set for us to make our first donor/acceptor catenane [35] by simply templating the synthesis of cyclobis(paraquat-*p*-phenylene) in the presence of three equivalents of BPP34C10 as the template in acetonitrile. This template-directed synthesis proceeded in a remarkable 70 % yield at room temperature. I will never forget how excited I felt as I watched with Neil Spencer and Cristina Vincent this [2]catenane crystallize out of the side of the flask. It was obvious immediately from the initial electrochemical investigations, as well as from the dynamic ^{1}H NMR spectroscopic studies and the X-ray crystal (super)structure determinations, that we had [36] the basis, in this simple degenerate [2]catenane, for the design and construction of bistable mechanical switches.

Without putting too fine a point on it, the Sheffield years were far from easy ones—at least outside of my own research laboratory and that of Ian Sutherland with whom it was such a pleasure to collaborate [37] and compete [38]—for a shy and retiring Scot of, nonetheless, quite an independent mind and spirit, with an insatiable appetite for tackling what many clearly thought were crazy research projects, but which I believed passionately were akin to the big problem Sir Edmund Hirst had implored me to identify and work on in my capacity as an independent researcher. Any successes, however modest, only seemed to engender envy and resentment amongst some of my senior and influential colleagues who would then go to any lengths to undermine my academic activities. At times, I was so acutely discouraged that I would surely not have stayed the course without Norma's pouring scorn on my repeated threats to throw in the towel and go off and grow potatoes in some remote place with the kind of open, warm-hearted, and supportive people I had been brought up amongst in the Lothians around the Scottish capital. She would remind me firmly that, with the added responsibility of raising and educating our two dear daughters, Fiona and Alison, I really had to buckle down and get on with it, however unpleasant I found the experience to be a lot of the time.

Eventually, I stumbled across an approach to my conundrum that I found quickly worked rather well for me when it came to coping with the minefield that was academic politics within and beyond an English red-brick university. With some considerable effort on my part, I went virtually overnight from being quietly passive and resigned to my fate to being openly, visibly, and even aggressively active as my self-confidence soared. Although I know that my pronouncements and activities did not win

me many friends in high places, the fact that I was making the first moves and creating the surprises for once, left me feeling that I was finally in charge of my own professional well-being and my future. What a relief!

I wrote letters frequently to the national press condemning the wantonness and waste I witnessed all around me. I questioned the extremely high level of bureaucratic state control that accompanied the hierarchically manipulated allocation of financial and other resources to research in science and engineering in Britain. I remained during this time at a complete loss, however, trying to explain to myself why the state's monopoly in higher education was not only willingly accepted by an acquiescent academic community from vice-chancellors and principals downwards, but was also sustained through thick and thin with the connivance of scholars and students, alike. Even to this day, I wonder just how long the debilitating ritual and rigmarole that surrounds the widely respected and regarded research and teaching assessment exercises can continue before the havoc they have wreaked upon education and scholarship in British universities is recognized.

I was particularly vocal in my repeated calls for the calibration of excellence or otherwise in research against some international yardsticks. I badgered a number of complacent and reluctant administrators to put grant proposals—particularly mine—out for review in the United States and was, to some extent, successful. I had found that, while it was far from easy to be any kind of prophet in my own land, there were enthusiastic supporters—amongst them Don Cram at UCLA—overseas for the research my group was doing. I had spent a period of three months with him and his research group on sabbatical leave before going to the ICI Corporate Laboratory in Runcorn in 1978. The 12 weeks or so as a Science Research Council Visiting Fellow turned out to be an enormously uplifting experience for me. I found not only an enthusiastic supporter, but also a role model in Don, just as I had back in 1970 when Ernest Eliel, then at Notre Dame University, provided me with a wealth of sage advice and unsolicited comments on how to improve greatly the manuscript of my monograph [10] *The Stereochemistry of Carbohydrates* before it was packaged up and sent off to the publishers. Somewhat later in my career, I was to meet up at international symposia with another two chemists from the United States that I hold to this day in the highest regard. One is David Gutsche, who was at Washington University in St. Louis when I first came to know the founding father of the calixarenes [39,40], and the other is Daryle Busch, the pioneer of template-directed synthesis [41,42], from the University of Kansas in Lawrence. All four—Don, Ernest, David, and Daryle—of these great philosophers impressed me from our very first meetings, not only with their scholarship, but also with their kindness, humanity, and friendship, transmitted as it was regularly to Norma, as well as myself, through their respective very special and dear wives, Jane, Eva, Alice, and Jeri.

Aside from establishing my credentials in Sheffield as a chemist who did research beyond the mighty molecule [43], my group made another foray into the realm of exotic molecular compounds, which turned out to be particularly exhilarating at the time. A quest [44,45] to synthesize compounds, such as [12]cyclacenes [46], in the wake of the emergence [47–49] of C60 on the scene, led to the substrate-directed synthesis [50,51] of a hexaepoxyoctacosahydro-[12]cyclacene derivative [52], which I decided to christen Kohnkene, after its maker, Franz Kohnke from the University of Messina. Thereafter, we employed highly stereoselective multiple Diels–Alder reactions to synthesize [12]callarene [53], trinacrene [54], and a range of stereoregular oligomers and polymers [55–57], thanks to the fillip given to this research program by the extremely talented John Mathias. We were all set to play a game of "molecular LEGO" [58–60] and had already published two full papers [61,62] in the *Journal of the American Chemical Society* on this particular kind of "click chemistry", as Barry Sharpless [63] later called it, when the Danish toymaker made it very clear to us that our continued use of their trademark was going to end up with me in a court of law! I can only reflect now, with some amusement more than a decade on, just how random some of life's experiences can be for I have lost count of how often I have sat in an audience listening to other chemists talk about their brand of molecular LEGO. Maybe there will be refuge in numbers amongst chemists if it should ever come to a showdown with the toymaker's lawyers.

Before I departed from Sheffield in 1990 with my research group for the University of Birmingham, Pier-Lucio Anelli had assembled the degenerate [2]rotaxane counterpart of the first [2]catenane: I called it a molecular shuttle [64]. Soon thereafter, Richard Bissell, in collaboration with Angel Kaifer at the University of Miami, synthesized the first bistable [2]rotaxane [65] that could be switched electrochemically, as well as chemically. Whereas the molecular shuttle contained two hydroquinone rings within the rod section of its dumbbell component, in the bistable counterpart, one hydroquinone ring was replaced by a benzidine unit and the other one by a biphenol residue. The switching mechanism relies on the fact that the ring component, cyclobis(paraquat-*p*-phenylene), spends only four-fifths of its time on the benzidine unit and the remaining one-fifth of its time on the biphenol residue. In other words, the switch fell somewhat short of being a perfect one!

THE BIRMINGHAM ERA

In summary, molecular compounds, comprised of mechanically interlocked donor/acceptor components, can now be obtained [66–69] efficiently using template-directed protocols [70] that rely on supramolecular assistance to covalent synthesis [71]. Since the weak noncovalent bonding interactions that orchestrate the synthesis of [2]catenanes and [2]rotaxanes containing mechanical bonds live on the components inside the molecules thereafter, they can be activated such that their components move with respect to each other in a linear fashion—e.g., the ring component along the rod section of the dumbbell component in a bistable [2]rotaxane [65,72]—or in a rotary manner—e.g., one asymmetrically constituted ring in a [2]catenane circumrotating through the other symmetrically constituted ring. The best example of a bistable [2]catenane was designed and synthesized by Gunter Mattersteig and became known within my research group as the Gunter catenane [73,74]. He replaced one of the two hydroquinone rings in the BPP34C10 component of the original [2]catenane with a tetrathiafulvalene (TTF) unit and the other with a 1,5-dioxynaphthalene (DNP) ring system. Gunter based his design on an observation [75,76] made by Douglas Philp—namely, that TTF, of all the many π-donors hosted by the blue box, was by far the most tightly bound within its cavity, and certainly much more so than 1,5-dimethoxynaphthalene. The fact that the TTF unit can be easily oxidized to both its radical cation and dication in a reversible fashion allowed us, in collaboration with Vincenzo Balzani and Alberto Credi at the University of Bologna, to switch the Gunter catenane back and forth between a green ground state and a purple (electrochemically) excited state. Francisco Raymo, in collaboration with Masumi Asakawa in the Nanoarchitectonics Research Center in the National Institute of Advanced Industrial Science and Technology at Tsukuba in Japan went on to show that, if the four hexafluorophosphate counterions associated with the bistable [2]catenane were exchanged for dimyristoylphosphatidyl anions, then stable molecular monolayers of the tetracationic [2]catenane could be obtained [77] at the air–water interface using a Langmuir trough. The area occupied by each bistable [2]catenane was found to be just a little over one square nanometer. In other words, the Gunter catenane, at around a cubic nanometer in size, constitutes the smallest molecular switch synthesized to date. Our expertise in the production of Langmuir monolayers had been gained in the beginning when Jon Preece spent a period during the mid-1990s in Helmut Ringsdorf's laboratories at the University of Miami putting the original degenerate [2]catenane through its paces on a Langmuir trough. This research [78] was to have its importance fully realized later on after I moved to UCLA in 1997 and began a collaboration with Jim Heath in 1998 in the area of molecular electronics.

I left Birmingham for California in the knowledge that bistable catenanes and rotaxanes can be activated [72–74,76,79,80] by switching their recognition elements on and off between their components chemically, electrically, and optically such that they perform motions—e.g., shuttling actions or muscle-like elongations and contractions—reminiscent of the moving parts in macroscopic machines. Such motor-molecules and molecular machines hold considerable promise [81,82] for the fabrication of sensors, actuators, amplifiers, and switches at the nanoscale level. I was only too conscious of the

fact, however, that we had to take our chemistry out of solution into condensed phases and onto surfaces, otherwise it was going nowhere beyond being exotic!

I must make one final comment, however, about the Birmingham era. It was an amazingly fruitful one in respect of designing and constructing donor/acceptor catenanes and rotaxanes thanks to the combined efforts of an incredibly talented bunch of young researchers drawn to our sparkling new laboratories from all around the world. We were never less than a dozen different nationalities. The mix of cultures was sheer magic and was to serve as an exemplary model for many other academics with large research groups in chemistry departments in the United Kingdom to adopt during the 1990s. The community, however, did not receive our new mechanically interlocked molecular compounds with open arms in the beginning. Referees and reviewers were even prepared to contest the very existence of interlocked molecules, and it was in response to a ridiculously high level of skepticism and criticism that I enlisted the help of David Williams to carry out hundreds of X-ray crystal structures. It was a case of bringing structural chemistry [83] at its most elegant and convincing to the rescue. Another way in which we sought to answer our detractors at the time was to challenge ourselves to show that the template-directed protocol [42,70] could deliver higher-order catenanes relatively easily. No one was more enthusiastic about tackling this goal than David Amabilino, who worked closely with Anatoly Reder and Ju-Young Lee to make, in 1994, a [5]catenane I called Olympiadane [84,85], and ultimately in 1997, a derived branched [7]catenane [85,86]. David Williams, with no end of talent and tenacity, solved the crystal structures of both of these remarkable compounds.

He also helped Peter Glink and Douglas Philp to establish [87–89] another recognition motif where strong hydrogen bonding of the [N^+–H···O] type, supplemented more often than not by weaker [C–H···O] interactions, and sometimes [π–π] stacking interactions, aids and abets the threading of secondary dialkylammonium ions ($RCH_2NH_2^+CH_2R$) through macrocycle polyethers containing at least 24 ring atoms in total with up to eight of them heteroatoms, preferably oxygen or nitrogen, to form pseudorotaxanes from which both rotaxanes [90] and catenanes [91] can be obtained. This particular discovery in my laboratory was one that was made simultaneously and independently by Daryle Busch [92] in his research laboratory over 5000 miles away in the middle of the United States. It never ceases to amaze me how often researchers' minds think alike simply because the time is opportune for some key event to happen in science. Indeed, there are very few "firsts", and it is wise to exercise caution and avoid laying claims to "firsts". I always argue that it is best to let other scientists be the judge and the jury on that one!

The "ammonium binding" chemistry, as it became known in my group, was developed with great rapidity during our last two years in Birmingham, where Peter Glink, Matthew Fyfe, and Stuart Cantrill played major roles in the design and realization of a wide range of supermolecules [93–100] that stand to this day as monuments to their creativity and productivity. They all subsequently played their parts in summarizing [101–105] their achievement in review articles that spanned 1996 through 2000. The "new" recognition motif is one which, of course, is susceptible to switching off and on with base and acid, respectively. It fell to Mari-Victoria Martínez-Díaz and Ariana Piersanti to bring about the marriage [106,107] of the "ammonium binding" recognition motif with the donor/acceptor one and in so doing bring acid-base controllable molecular shuttles [108–110] into being with spectacular selectivities from the very beginning. The Birmingham years were golden ones also because of the excellent support Neil Spencer provided in NMR spectroscopy and the sheer brilliance that Peter Ashton brought to the use of mass spectrometry in so many ingenious ways, affording the swift characterization of mesomolecular complexes as well as high-molecular-weight compounds. Life at UCLA without Peter is something I still find difficult to come to terms with after more than seven years.

CALIFORNIA HERE WE COME

Multivalent interactions, incorporating both statistical and chelate contributions, are very important in nature and have been explored by my group at UCLA in the noncovalent synthesis [71] of various elab-

orate supermolecules [105,111,112] and molecular bundles [113,114]. Inspired by the concept of multivalency, we have discovered [115] that the strict self-assembly of a triply threaded two-component superbundle can be less than a straightforward process in certain circumstances. In particular, it transpires that a trifurcated trisbipyridinium trication and a tritopic crown ether form the thermodynamically stable triply threaded superbundle by way of a metastable doubly threaded complex obtained fleetingly from a singly threaded intermediate species. This observation begs the important question, Are there instances in nature where multivalency is expressed as a kinetically controlled process prior to an equilibrium state being reached and, if so, what are the biological consequences if any? Indeed, there are instances, as have been reported [116] by Geert-Jan Boons of the Complex Carbohydrate Research Center at the University of Georgia in Athens. We have also explored the cooperativity [117] of multivalency in synthetic supramolecular systems, as well as its exclusivity [118] in dynamic systems. Cooperativity in multivalent systems can be positive, neutral, or negative. The synthetic systems we have investigated [115,117–119] to date have all exhibited negative cooperativity. This research led quite logically to the template-directed synthesis of a molecular elevator [120], which was found, in collaboration with the Bologna group, to operate in a manner that is more reminiscent of a legged animal than it is of a passenger elevator.

Since most mechanical devices rely on solid supports—in the form of either surfaces or interfaces—for the transmission of energy or force, the advent of the molecular elevator performing elegantly in solution provided us with yet a further impetus to develop nanomechanical devices that operate at both supramolecular [121,122] and molecular [123] levels on surfaces. This particular research has been the direct result of a Nanoscale Interdisciplinary Research Team (NIRT) effort supported by the National Science Foundation at UCLA. While an operational supramolecular nanovalve [122] has been designed and fabricated in collaboration with Jeff Zink and his group in the Department of Chemistry and Biochemistry, chemical energy has been transducted into mechanical energy in a joint research effort with Chih-Ming Ho's group in the Department of Mechanical and Aerospace Engineering. Working closely with the mechanical engineers, and also with scientists at Vecco Instruments in Santa Barbara, we have found [123] recently that arrays of microcantilever beams, coated with self-assembled monolayers of palindromic, doubly bistable, redox-active [3]rotaxane molecules, undergo controllable and reversible bending when they are exposed to chemical oxidants and reductants. When the gold-covered beams are coated with a redox-active, but mechanically impotent control compound, they do not bend. A series of control experiments and rational assessments preclude the influence of heat, photothermal effects, and pH variations as potential mechanisms of beam bending. Along with a simple force calculation, our experimental observations support the hypothesis that the cumulative nanoscale movements within surface-bound molecular muscles can be harnessed to perform large-scale mechanical work.

Working closely in collaboration with the Bologna group, we had demonstrated during the Birmingham era, at both the supramolecular [124] and molecular [125] levels, photochemically driven machine-like systems based on a [2]pseudorotaxane and a [2]rotaxane, respectively, in solution. Recently, we have employed [126] a photoactive donor/chromophore/acceptor molecular triad, containing C60, as well as porphyrin and TTF units housed as a self-assembled monolayer in a photoelectrochemcial cell, to generate a photocurrent that is used subsequently to drive the original supramolecular machine, i.e., the [2]pseudorotaxane that was demonstrated [124] to have machine-like qualities in solution back in 1993. And now, more than a decade later, we are slowly bringing the essential pieces together in order to fabricate devices. It is a highly incremental process that has taken time to mature gradually from the dreams of the early days [127–129] to the realities of the present. One is constantly reminded that Rome was not built in a day.

Multivalency is most certainly a concept that is going to dominate increasingly the development [117] of supramolecular nanoscience. It was undoubtedly my long-standing interest in carbohydrates that drew us into this fascinating jungle of molecular recognition and self-assembly processes during the Birmingham era when (Jay) Jayaraman and Sergey Nepogodiev became enthusiastically involved

in the design and synthesis of glycodendrimers [130–134] that were essentially "sugar-coated balls" for the most part. Later on, Bruce Turnbull initiated an ingenious synthetic program based on oligosaccharide-based AB_2 monomers to produce all-carbohydrate dendrimers [135–136] with chemically defined sialoside scaffolds for investigating multivalent interactions with sialic acid binding proteins [137] in collaboration with Jim Paulson and his group at the Scripps Research Institute in San Diego. In our most recent attempts to display epitopes, such that they can span the typically long distances found between the binding sites in lectins, Al Nelson and Jason Belitsky came up [138] with a self-assembled pseudopolyrotaxane consisting of lactoside-displaying cyclodextrin (CD) "beads" threaded onto a linear polyviologen "string". In collaboration with Linda Baum in the Department of Pathology at UCLA, Al and Jason investigated this dynamic system for its ability to inhibit galectin-1-mediated cell agglutination. It exhibited a valency-corrected 10-fold enhancement over native lactose in an agglutination assay. This is a good outcome in comparison with a lactose-bearing glycocluster and a hyperbranched glycopolymer.

CARBOHYDRATES KEEP COMING BACK

The CDs have been a continuing source of fascination for me ever since my days as a postdoctoral fellow in Canada when I found [14] in 1969 that α-CD and β-CD could be easily transformed into 30- and 35-membered heterocyclic rings, respectively. This piece of chemistry was rediscovered [139] by the Lichtentalher group in Germany in 2000, illustrating the fact that most of what's in the primary scientific literature is largely forgotten within three decades and so becomes fair game to repeat and, of course, improve upon, using the state-of-the-art tools.

During the 1980s in Sheffield, Ric Zarzycki and David Alston employed their combined talents as experimentalists to reveal that the parent CDs could act [22,140,141] as excellent second-sphere ligands for transition-metal complexes, including the chemotherapeutic drug, carboplatin [142–144]. Paul Ellwood made our first foray into the extremely demanding practice of chemically modifying CDs by synthesizing and characterizing their per-3,6-anhydro derivatives [145,146], some of which turned out to be reasonable hosts for hard-metal ions in organic solvents. As we wished Sheffield good-bye and headed for Birmingham, I reflected on [147] A Century of Cyclodextrins in my role as the guest editor of a special issue of *Carbohydrate Research*.

In Birmingham, Dominique Armspach achieved something with CDs that had been first attempted unsuccessfully by Lüttringhaus et al. [148] in 1958—that was the catenation by synthetic macrocycles of β-CD to give both a [2]- and [3]-catenane [149–151]—while Rainer Königer was one of the first researchers anywhere to synthesize a thiolated β-CD derivative and study its interfacial binding properties [152] in collaboration with Angel Kaifer at the University of Miami. About the same time, Neil Spencer carried out [153] in-depth high-field ^{1}H NMR spectroscopic studies on some constitutionally unsymmetrical CDs where, of course, every α-1,4-linked glucoside residue is different, i.e., constitutionally heterotopic. The field of chemically modified CDs is littered with research carried out on less than pure compounds because constitutionally unsymmetrical derivatives are extremely difficult to characterize, even by high-field ^{1}H NMR spectroscopy. Neil demonstrated that, with time and care, a really good professional job can be done. More recently at UCLA, David Fulton displayed his exceptionally well-developed experimental skills in the synthesis and characterization of numerous CD-based (carbohydrate) clusters [154–157], one of which was used [157] to probe multivalency in the context of the "ammonium binding" recognition motif.

A wish to escape from the structural straitjacket of the readily available CDs, promiscuous as they are at forming complexes with almost anything and everything of the right size, prompted us to explore the synthesis in Birmingham of CD analogs, e.g., cyclic oligosaccharides [158] containing disaccharide repeating units composed of rhamnopyranosyl and mannopyranosyl units linked α-1,4, some of which are achiral [159] and others of which crystallize to form arrays of nanotubes [160]. These remarkable synthetic feats were achieved thanks largely to the very special knowledge and unique experimental

skills that Sergey Nepogodiev brought to my research group from the Zelinsky Institute at the Russian Academy of Sciences in Moscow. He was ably supported by Ph.D. student Giuseppe Gattuso from Messina University, and undergraduate researcher Stuart Cantrill. Never short on effort, Sergey put his remarkable synthetic achievements in context in a long review [161] published in a special issue of *Chemical Reviews* in 1998 devoted to CDs.

THE BEGINNINGS OF NANO MECCANO

Although the vast amount of research, carried out initially in Birmingham by Gunter Mattersteig, and then at UCLA by Jan Jeppesen and Kent Nielsen from Jan Becher's group at the University of Southern Denmark in Odense, on bistable, switchable catenanes [73,74] and rotaxanes [161–164] was performed in the solution phase, we have demonstrated during the past couple of years that the relative mechanical movements between the components of these interlocked molecules can be stimulated (i) chemically in condensed phases (e.g., Langmuir films and Langmuir–Blodgett monolayers [165]), (ii) electrochemically in a highly viscous polymer matrix [166], (iii) electrochemically in a "half device" as a self-assembled monolayer on gold [167], and (iv) electronically in a "full device" within solid-state molecular switch tunnel junctions [168–170]. Not only has reversible, electronically driven switching been observed [168,169] in devices incorporating a monolayer of a bistable [2]catenane sandwiched between a bottom polysilicon electrode and a top titanium/aluminum electrode, but a cross-point random-access memory circuit and a simple logic circuit have been fabricated [170] by the Heath group using amphiphilic, bistable [2]rotaxanes. The experiments (i) through (iv) described above provide compelling evidence that the bistable switchable catenanes and rotaxanes operate mechanically in a soft-matter environment and can even withstand quite harsh device-processing steps. In close collaboration with David Steuerman in Jim Heath's group at Caltech, and Xiang Zhang's group in the Department of Mechanical and Aerospace Engineering at UCLA, Tseng, Amar Flood, and Andrea Peters have identified, by time-dependent cyclic voltammetry in solution at low temperatures [171], as well as in the polymer matrix [166] and in the half device [167], a metastable state for the molecular switches where the cyclobis(paraquat-*p*-phenylene) ($CBPQT^{4+}$) ring resides on the DNP unit. In the full device, this state is postulated [81,168–170] to correspond to the closed or ON position (more conducting) of the switch. When the $CBPQT^{4+}$ ring encircles the TTF unit (the ground state), the switch is in the open or OFF position (less conducting). First-principles calculations of the current/voltage responses, carried out [82] by Bill Goddard's group at Caltech on model rotaxane systems of the metastable and ground states, support their association with the ON and OFF positions, respectively, of the switch—and so lend support to the proposed switching mechanism [81,168–170] in the full device. The metastable state of a switch in a full device decays back to the ground state during a period of 10–60 min. If, however, the bipyrindum units in the $CBPQT^{4+}$ ring are reduced from dications to cation radicals, molecular recognition is lost and switching becomes almost instantaneous. It transpires that the metastable to ground-state relaxation times of the bistable molecular switches in solution are much shorter than they are in the polymer matrix and in the half device by an order of magnitude. By the same token, the relaxation times in the full devices are longer than they are in the polymer matrix and in the half device by an order of magnitude. In terms of activation barriers, to get from the metastable back to the ground state, a value of around 16 kcal mol^{-1} in solution rises to around 18 kcal mol^{-1} in the polymer matrix and half device, and finally up to around 22 kcal mol^{-1} in full devices.

If *Science* is anything to go by, the field of molecular electronics goes from being [172] "Breakthrough of the Year" in 2001 to having [173] a "Mid-Life Crisis" in 2003. Only time will tell what is good and what is bad. All I can say is that my group soldiers on in determined fashion with Jim Heath's group to try to do the very best research we are capable of doing jointly, despite all the nastiness we have to endure from reviewers one would hope would know better. The science is far from easy, but we have got to where we are today in an incremental manner, stretching back over a quarter of a century in order to take molecular recognition to molecular switch tunnel junctions. The nature of these

junctions is the key to being able to observe switching by molecules—a small and delicate chemical effect that can easily be swamped by much bigger physical effects. Fishing out the chemical effect is a challenging task at this time, and only the very best experimentalists are up to the job.

The marriage between molecules and electrodes is not an easy one to perform successfully. While Jim Heath's group has found it possible to observe remnant molecular signatures for bistable molecules trapped between a polysilicon bottom electrode and a titanium/aluminum top electrode, Stan William's group sees no remnant molecular signatures when they replace the polysilicon bottom electrode in the cross-bar device with platinum [174–176]. So, since silicon and oxygen provide the opportunity to reveal remnant molecular signatures in bistable catenane and rotaxane devices, we asked ourselves, what about carbon? And indeed, when a semiconducting carbon nanotube is chosen as the bottom electrode, a remnant molecular signature is observed [177] in the appropriate devices. And so it looks as if carbon, silicon, and oxygen are all good choices when carrying out molecular electronics with mechanically interlocked molecular switches. As far as the top electrode is concerned, titanium can be considered as a gift. It works, probably because it forms titanium–carbide bonds with alkyl chains in the exposed hydrophobic parts of the molecular monolayers—dimiristoylphosphatidyl anions in the case of switchable [2]catenanes, and substituted tetraarylmethane stoppers in the case of switchable [2]rotaxanes. It would seem that our organic molecules can be seen to do their job when, and only when, the electrodes are composed of elements (C, Si, O, and Ti) that are close to those (C, N, O, Si, and S) present in the organic molecules themselves where the work functions is very similar. This conclusion is supported by differential conductance measurements made [178] just above absolute zero with single-molecule transistors where the source and drain electrodes are platinum and the gate electrodes are a degenerately doped silicon substrate. Jim Heath's group has discovered that the electronic transport properties in such devices are extremely sensitive to the chemical nature of the molecule electrode contacts, which can often completely mask the molecular signature the bistable molecule might be trying to display in the background. This observation has profound implications for molecular electronics.

Our research on molecular electronics, not surprisingly, attracted us into another challenging area of science that has been highly topical of late—that of separating, purifying, and manipulating carbon nanotubes of the single-walled variety. We have chosen a supramolecular approach, and to that end we have investigated the properties and interactions between conjugated polymers, and single-walled carbon nanotubes (SWNTs) in organic solvents as well as the ability of commoner-garden starch to solubilize [182] SWNTs in water. Most of the early work was carried out by Sasha Star who was subsequently joined by Yi Liu and Kelly Chichak. In the wake of Sasha's departure to join Nanomix in the Bay Area, a fruitful collaboration has led to the demonstration [183] that the enzymic degradation of starch can be monitored electronically using SWNTs as semiconducting probes in field effect transistors (FETs). Incubation of these devices in aqueous buffer solutions of amyloglucosidase results in the removal of the starch from both the silicon surfaces of the devices and the side walls of the SWNTs in the FETs. Presently, Kelly and Sasha are studying the influence of dynamic coordination chemistry, in conjunction with supramolecular chemistry, upon SWNTs to render them soluble in water [184].

DYNAMIC CHEMISTRY IS THE FUTURE

Dynamic covalent chemistry [185] is another research area to have grown rapidly around, particularly imine bond formation [186–190], thanks to the expertise and line of thinking that Stuart Rowan brought from Cambridge to my group in 2000. It is remarkable what a fresh mind can bring to a research group, and Stuart will always be remembered for the key role he played as an educator and teacher, as well as a researcher. His influence continues to live on several years after he left us to become an assistant professor in the Department of Macromolecular Science at Case Western Reserve University in Cleveland, Ohio. It was sufficient for Sasha Star, Al Nelson, and Sebastien Vidal to demonstrate [191] amplification of dynamic chiral crown ethers during cyclic acetal formation by threitol. Stuart's influence created the intellectual atmosphere and prepared the way in terms of the group's knowledge-base regard-

ing imine bond formation for the making of the molecular Borromean rings [192]. Stuart Rowan got Stuart Cantrill thinking about an all-in-one-step approach to this elusive piece of topology using an exquisite combination of dynamic covalent, coordination, and supramolecular chemistry. It took that all-important combination of talent to pull off the synthesis in almost quantitative yield. While Chiu did the conventional covalent synthesis to make one of the ligands, it was Kelly Chichak who brought everything together in an inspired way. This triumph in template-directed synthesis has already drawn some favorable comment [193,194]. It is amazing how something that was difficult to do in the beginning will surely become easy to do in the event of its having been done. The Borromean rings have captured our imagination [195] simply because of their sheer beauty. What will they be good for? Something for sure, and we still have the excitement of finding out what that something might be. And so the story goes on...

ACKNOWLEDGMENTS

The story related in this review is only one that I can tell because of the intimate involvement of hundreds of outstanding postgraduate students and postdoctoral scholars who have excelled in my research group over the years—and the critical support we all witnessed from being alongside Norma Stoddart. I thank them all and also the numerous collaborators the world over with whom I have had the joy of doing research at a range of levels and frequencies. Dr. Cantrill helped me put this story together. Thank you, Stuart. I have been fortunate beyond belief to have lived in places and times when many people—call them taxpayers if you so wish—support me financially and in other ways to practice my hobby every day of my life. I am indebted to countless sources for this largess and bounty. In recent times, UCLA, the NSF, the NIH, the ONR, and DARPA have been particularly generous. Thank you all.

NOTE ADDED IN PROOF

I realize on reading this manuscript again at the proof stage that there are a lot more topics and people I should have mentioned and failed to do so. I beg the forgiveness of those who would expect a mention and did not receive one. Maybe on another occasion when space—which I have already violated considerably—is not at such a premium.

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