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Andrew Keller: the nucleation and growth of polymer physics in Bristol

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Abstract

The discovery of polyethylene single crystals leading to the inference of molecular chainfolding was made by Andrew Keller with minimal resources. The background to his early research in Bristol and the development of his Group are described. His practice of combining microscopy with complementary techniques on the same well-defined system remains the most productive approach to the solution of manifold problems in polymer physics. © 2000 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Andrew Keller's unexpected death in February this year deprived us not only of a principal pioneer of our subject who injected enormous impetus into the field but also one who retained his interest and energetic activity to the end. Many of the papers in this conference, including my own, address topics to which Andrew made signal contributions. But above all he will be remembered for the introduction of molecular chainfolding to our thinking. The revolutionary dynamism this gave to the subject more than forty years ago can hardly be overestimated. It led, very quickly, to much insight into the basics of physical texture (see for example Geil's text [1] written in 1961). This success brought Bristol international attention and a stream of visitors, leading in due course to the well-equipped major international group, that Andrew headed, and with which many will be familiar. By contrast, his early contributions were made with minimal equipment from one room which he described as 'an "office" filled with fumes, sparks and scattered X-rays amidst total isolation from, in fact in ignorance of, the rest of polymer science'.

I was privileged to share that "office" with Andrew for one year, until 1959 when we acquired a second room whose upstairs enclosed-balcony then became our new shared office for the next three years. They were exciting days and nights working with our Professor, F.C. Frank, who, sadly, also died just ten months previously, in April 1998. New results came thick and fast as we explored the morphology of solution-grown crystals, the nature of the newly discovered lamellae and the radical implications for the subject. As the survivor of that pioneering time in Bristol it seems to me that it may be of some general interest to record the context in which Andrew Keller's Group nucleated and grew not least because, as with polymeric nucleation itself, the seeds of the outcome were clearly perceptible from the start.

2. To Bristol and before

Andrew Keller went to Bristol in 1955 when he was thirty, a Research Fellow with Professor Frank, some seven years after arriving in England from his native Hungary. He spent the intervening years at ICI Dyestuffs in Manchester. The story as I know it centres on Mr Warburton Hall of the then Ministry of Aviation Supply who wished to fund a fellowship in what he foresaw as important materials. He first approached Professor W.T. Astbury of the Biophysics Department of the University of Leeds as a prospective host but Astbury declined, in view of his impending retirement, and suggested Frank as an alternative host. There was no such uncertainty over the identity of the intended fellow: it was to be Andrew Keller whose work at ICI had already attracted attention.

Andrew's papers from ICI were concerned with the several crystallizable polymers then available including Terylene, poly(ethylene terephthalate) or PET, polyamides and branched polyethylene (Alkathene). He had encountered spherulites, studied their crystallization kinetics and appearance in the polarising microscope, showing that

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bands were due to periodically twisting orientation. He had isolated them by selective dissolution from their surrounding matrix and attempted to fragment them, reporting helically twisted entities in the debris. He had mastered the difficult technique of microbeam X-rays to establish local molecular orientation and used more conventional wide-angle diffraction (WAXS) to explore molecular relaxation with increasing temperature. It seems that certain of these last results, on individual fibres, aroused interest in the Company as a possible means of improving the properties of PET fibres. To this end management decreed, simplistically, that a bobbin of fibre be placed in an oven and taken through the appropriate heating cycle. Of course, the fibres became sticky, adhered to each other and could not be unwound. Andrew told me that as a consequence, he got into considerable trouble for time-wasting.

Another story from those days that I enjoyed, concerned the long isothermal experiments required for PET crystallizations carried out in a suitably-controlled oil bath. Andrew had left samples in place over the weekend with instructions that the temperature be checked periodically but that he was not to be contacted unless it varied from the set value. Indeed he was contacted, by the night watchman, to be told that the temperature had fallen substantially, which was hard to credit since there had been no previous problem. But the watchman was adamant that the temperature was much lower than it should be, and so Andrew undertook the long journey to the laboratory across Manchester by bus. When he arrived it was to find that the bath was at exactly the right temperature. Only when he asked to be shown how the low temperature had been measured was the paradox resolved. The light was poor in the fume cupboard housing the bath so the watchman had removed the thermometer, taken it to the window to see more clearly, and there read the temperature. Unfortunately, it was not a clinical thermometer.

During those years Andrew had acquired a portfolio of experimental techniques to investigate polymer morphology, a term he claimed to have invented, as well as a knowledge of structure/property problems in polymers. But he was something of a lone voice in Manchester in contrast to ICI's Welwyn laboratory where the eminent crystallographer C.W. Bunn and colleagues were based. It was another eminent crystallographer, A.F. Wells, who supported Andrew at this time for which he always expressed warm appreciation. Whatever minor differences of opinion may have existed within the company, they presumably contributed to Andrew's reputation for independent thought coupled with a vigorous and single-minded pursuit of the solution to problems, which made him the preferred candidate for Warburton Hall's fellowship.

That this should have been at Bristol was a most happy choice, it being a very strong Physics department, not least in Materials Science, with F.C. Frank, 'an intellectual firebrand' at the helm, interested in all good problems. So it was that Andrew gave a colloquium in Bristol that persuaded Frank to supervise the Fellowship. Sir Charles recalled that he then became aware of banded spherulites and thought that it would be an interesting three years in which to sort matters out.

Mr Keller, as he still was, was allocated a room to serve as office and laboratory, and a research student Tony O'Connor—officially, I believe, accredited to Frank—to share the work. That work had O'Connor recording WAXS patterns of deformed and relaxed branched polyethylene [2], with Andrew studying banded and other spherulites mostly of polyesters and polyamides [3,4]. The story goes that Andrew tended to have difficulty in finding Tony during the day although work was still being completed satisfactorily, eventually realising that Tony, already a rugby player of distinction, was spending time in day training. He played scrumhalf for Wales, later becoming an Oxford Blue (while a postdoctoral fellow with Dorothy Hodgkin) then a British Lion before taking an industrial post in the Principality.

There was very little equipment and it is interesting to note what Andrew's priorities were in this regard at the outset of his fellowship. There was a Zeiss GFL polarizing optical microscope, with a four-axis stage, for studying spherulites but which was also equipped for phase contrast imaging. Andrew had had to justify buying this last facility to Professor Frank but with the later discovery of single crystals, for which it was ideal, it was clearly an inspired and prescient choice. There was a Kofler hot bench providing an instant source of a wide range of stable temperatures, which Andrew would use to great effect. I recall his gangling, somewhat sinuous presence, attired in an ill-fitting lab coat, sleeves flapping, but unfailingly able to use the Kofler and microscope to uncover novel phenomena, often beyond or challenging current understanding, well before an afternoon was over. Inevitably, the time that Andrew was able to spend doing his own experiments declined during my time in Bristol from small to zero; but he was a most gifted experimentalist.

The optical microscope and Kofler occupied one side of the office-room. Just inside the door was a homemade, lead-shielded, X-ray generator, prone to breakdown with cable failure but capable of 30 or even 35 kV when working well. The excellent departmental workshop had made two Kratky cameras for low angle (SAXS) work which also recorded simultaneous wide angle reflections albeit for an 18 h exposure (which necessitated a seven-day operation). Andrew had learned at ICI that SAXS varied with processing conditions in ill-understood ways and had resolved to explore such matters further. This, too, was an essential tool to explore the soon-to-be-discovered single crystals.

Beyond the X-ray set were two desks, head to head with Andrew's nearer the window. Along the fourth side were crystallization baths of an unusual pedigree. All were glass double-walled refluxing condensers constructed by the Department's outstanding glassblower, J.H. Burrow. When convenient these were operated at atmospheric pressure giving, with boiling carbon tetrachloride for example, a temperature of 76°C. More generally, chosen temperatures were achieved by refluxing at a constant mercury head below atmospheric pressure, the reduction being produced by a water pump and regulated by a needle valve. When I came to use these temperamental devices for overnight crystallization, I soon found that it was a lottery whether the temperature did remain constant over the time because of the frequent fronts and associated atmospheric pressure changes, which were typical of Bristol weather. Only much later when money became available for electrical controls did the problem disappear.

3. Chainfolding

It was Warburton Hall who provided Andrew with two samples of the first linear polyethylenes. One was a gallon jar labelled Marlex 50, the other a much smaller quantity of Marlex 5 whose pellets, however, contained banded spherulites. Andrew told me that he chose to dissolve the polymer in an attempt to open up the physical structure, drawing on his previous experience of how dense and difficult it usually was to inspect polymer morphologies. Crystallization on cooling had produced dendritic crystals that were not immediately recognized for what they were and so were put on one side for some months. Concurrently Andrew had been struggling to learn to use the electron microscope with the twin aims of gaining greater magnification and observing electron diffraction patterns from selected objects. It was a struggle partly because the loss of crystallinity from radiation damage had not been appreciated and partly because the lady in charge of the Department's Philips EM100 instrument preferred to examine all samples herself, and Andrew lacked the authority to overrule her. In the event it took eighteen months, with much help from Dr Kitty Little, to learn to produce the low-beam operating conditions which allowed the fleeting diffraction patterns to be observed. As I understand it, P.H. Till, E.W. Fischer and Andrew Keller all obtained diffraction patterns of polyethylene lamellae carrying the implication of chainfolding but only Andrew published the inference [5]. He was always categoric that he would not have done it, so radical was the concept, had it not been for Professor Frank's support: the Bristol factor at work!

Matters then moved quickly. The link of SAXS periodicity to lamellar thickness was appreciated [6]. One of the then new double condenser electron microscopes, operated by Dr Alan Agar, revealed moiré patterns and distinct sectors within individual lamellae, the vital confirmation of chainfolding [7]. Other new polyolefines were examined, notably poly(4-methyl pentene-1) [8], and were found to show similar lamellar and chainfolded habits. Andrew also began to investigate what was to be a continuing concern: at what length did folding set in. He prepared crude polyethylene fractions by solvent extraction, initially from thermally degraded Marlex 50, one of which showed the remarkable four-leaf pattern of rotated terraces [9], which intrigued and confounded all visitors. With these fractions a long period was found to be more or less independent of molecular weight, [10] but when Marlex 50 was crystallized from xylene solution the long period increased with increasing temperature [11], which was tentatively ascribed to a nucleation process. This was the state of affairs described in his seminal lecture [12] at the Cooperstown conference in August 1958.

4. Bristol 1958–1962

The recognition of lamellae as a component of polymer morphology and the possibilities of using them to help resolve some of the textural puzzles posed by these inherently complex materials attracted me and many others into the rapidly developing field. I joined Andrew in October 1958 as his first de jure student. He was preoccupied with writing his PhD thesis (for which J.D. Bernal was the external examiner) on extinction effects in banded spherulites, the discovery of single crystals and chainfolding as well as tidying up observations on spherulites [4]. Single crystals were left to me and I was asked to look into why data on the increase of a long period with crystallization temperature were single-valued to 80°C but showed an additional lower spacing at 85°C and 90°C. I suggested that the former could be due to isothermal growth, the latter to crystallization on cooling. Accordingly John Burrow was asked to fabricate a hot filtration apparatus with which the two long periods were separated into residue and filtrate (and in time to remove, in proof, the two, possibly confusing, lower points from the graph in the forthcoming paper [11]).

Then, events acquired a momentum of their own. What follows may help to give a feeling of the character of research in those early days when each experiment seemed to yield new, very often intriguing, results that we endeavoured to build into a framework of understanding. Research occupied our waking hours with discussions frequently continuing over lunch and well into the evening. Mrs Eva Keller was never disconcerted when, to pursue a subject further, Andrew would take me home with him, unannounced, to Richmond Terrace where a meal would be required for three not two. Saturday morning was the highlight of the week when we would meet Professor Frank at coffee, acquaint him with the week's findings, then discuss their implications until (very often a late) lunch.

The hot-filtered crystals were single truncated lozenges with two additional faces and a characteristic triangular fold present in their centres after sedimentation [13]. We showed this to Professor Frank who, I well remember, called for a sheet of paper, a pair of scissors and cellotape. He cut a wedge from the paper, joined the cut edges to form a hollow pyramid, then creased it flat to form a triangular fold.

Hollow pyramids result from an inclination of the chain axes to lamellar normals, symmetrically different in different sectors, which is general for polyethylene. When Andrew had originally inferred that chains were perpendicular to lamellae [5] he had assumed that the diffraction patterns related to the whole lamella; in fact they are composites of those from different sectors. The actual chain inclination was measured either directly, observing crystals before collapse while floating in suspension using optical dark-field microscopy [14] or, after collapse, using dark-field electron microscopy [15]. We did the latter together in what were possibly Andrew's last personal experiments.

What was required was to tilt lamellae about a chosen crystallographic axis and record when the diffraction contrast of a sector became maximum. The Philips EM100 instrument was not specifically designed for tilting experiments but its rod specimen holder could be rotated ±45° about its length and ±60° if a stop was removed. We attached a scale to the top of this specimen holder so that its angular rotation could be measured, but as the instrument column was nearly horizontal not only was it out of reach of a seated operator but also he had to be dark adapted while the scale needed to be read with a torch: a two-man job. Andrew operated the instrument at ~1000 times magnification and with an unsaturated filament to reduce the beam intensity; he found, watched and photographed crystals in suitable orientations while I rotated the specimen holder on his instruction. To do this I had to try to avoid, not always successfully, receiving an electric shock from the exposed terminals of a colleague's experiment, which were supposed to be covered by a thin cloth. That experiment was itself a tour de force in which Dr R.G. Chambers famously demonstrated the phase shift in electron interference associated with magnetic vector potential [16], in his case the introduction of an iron whisker, confirming the recent prediction made by Aharonov and Bohm [17], theoreticians in the Department.

Photography on the EM 100 had other pitfalls: there being no exposure meter on the instrument, exposures had to be guessed. It happened occasionally that a vital negative was so black and overexposed as to be impenetrable by the light of a normal enlarger. Andrew got around that problem with the purchase of a Blumfield enlarger that could accommodate a 500 W floodlight bulb—that was how several micrographs in the literature had to be printed.

At that time our only technical support was a half-share in a photographer, a capable lady but an unremitting perfectionist. All variations of paper, with shading to bring out detail in the best way, had to be tried for every negative so that her maximum output was 13 different prints per day. To cope with the considerable number of negatives we were taking, our only course was to use the dark room when it was free, at nights and weekends, and do our own printing retaining the specialist for publications. That was also before the time of photocopiers: making a copy meant serial photography, page by page. I still have photographic prints of some of those salient early papers made in this way, at the time.

To return to polyethylene single crystals, the two {100} truncated faces implied two additional sectors of a different type from those bounded by {110} faces that were identified [13], shown to have a lower melting point [18] and claimed to be just detectably thinner, as Atomic Force Microscopy has now confirmed. The morphological changes occurring through the melting range, including the melted {100} sectors, were observed using slides placed on the Kofler hot bench.

During that period there arrived an advance copy of the first Hoffman and Lauritzen paper on the theory of fold length and crystal thickness [19]. It equated this to the thickness of the primary nucleus and derived the melting point of a lamella to be midway between that of the infinite crystal and its crystallization temperature. We endeavoured to test this relationship by observing crystals while heated in suspension in a test tube, illuminated by a desk lamp against a dark background. The intention was to measure the temperature when cloudiness disappeared but it was frustrating and far from easy to obtain consistent values. What we did learn was that the dissolution temperature changed with the heating rate implying that the crystals had reorganized as they were heated.

At the same time I was asked to investigate whether the basic assumption that the lamellar thickness was that of the primary nucleus was correct, or whether the thickness changed if the crystallization temperature was altered. When we found that the thickness decreased abruptly at the growing edge when the crystallization temperature fell [20] this information was sent to Dr Hoffman whose revised paper [19] then contained what Professor Frank later described [21] as 'two successive theories'. This was the beginning of a fruitful and long-continuing symbiosis between theory and experiment: Dr Hoffman and the Bristol Group.

By the autumn of 1959, after four years, nucleation was complete and the Keller Group started to grow, doubling its numbers and space and gaining a full-time technician. Francisco Baltá Calleja arrived from Madrid on a Ramsay Memorial Fellowship and began to study the onset of folding in oligomeric polyamides [22] and poly(ethylene oxides) [23]. Shigenobu Mitsuhashi, from Yokohama, had sent us a diffraction pattern of polyethylene lamellae showing the slight splitting of spots consequent on sectorization and the collapse of hollow pyramids, contemporary with our own observations. He was invited to come and join us as Research Fellow, funded by Gillette Research in Brussels. First, he studied the four-leaf crystals [9] then, the morphologies of increasingly concentrated solutions [24] as an approach to melt-crystallized systems whose lamellar morphologies remained mostly inaccessible for another twenty years.

Other new topics were also introduced. Andrew had concluded his Cooperstown lecture by asking why, with polyethylene crystals so similar to those of the paraffins, the study of paraffins had not been the starting point for studying polymers. He took his own advice and examined several including n-$C_{36}H_{74}$, hexatriacontane, crystals; in so doing we established an interesting parallel with polyethylene [25,26]. The first intimations of distinct sectors in polyethylene lamellae had been lines approximately along $\langle 130 \rangle$ directions that, with improved growth procedures and sedimentation on glycerine [20], had been found to be due to alternating $\{31\ell\}$ facets. Similar alternating facets were found in hexatriacontane crystals prepared near their melting point.

In the summer of 1960, Andrew spent some two months at the Bell Telephone Laboratories, Murray Hill at the invitation of Dr W.P. Slichter. There he collaborated with Dr Ronald Salovey studying the effect of radiation on polyethylene lamellar aggregates, finding that the gel point depended very markedly on the way in which the lamellar fold surfaces were in contact [27,28]. This work was taken forward by Dr Toru Kawai when he arrived in 1961 to replace Mitsuhashi as Gillette Fellow, in collaboration with Arthur Charlesby, Professor at the nearby Royal Military College at Shrivenham, whose source of γ radiation greatly facilitated the experiments. Radiation studies retained a special interest for Andrew [29] although I think he always felt that this aspect of his work had received less recognition by specialists in the field than it deserved.

With so many new things to discuss in Polymer Physics, we organized a conference on that topic [30] held in January 1961, under the auspices of The Institute of Physics and The Physical Society, the first of its kind. The breadth of coverage and interest was outstanding while the list of those attending included many of the distinguished and famous names in the field including P.H. Geil, R.St.J. Manley and R.S. Stein who are also here today. It set the seal on Bristol's arrival as a centre of innovative Polymer Physics, a reputation that Andrew maintained throughout his career.

5. To modern times

Over succeeding years, the Polymer Group continued to grow and its facilities increased, beginning with a Philips EM 200 state-of-the-art electron microscope in 1962, with my namesake G.A. Bassett, appointed to be in charge. Most important was Andrew's appointment to a Lectureship in 1963, which made him a permanent member of the academic staff. Six years later, in 1969, he rose to the rare position of Research Professor, a full-time research post that allowed him to exercise his abilities to the full.

The topics under investigation widened accordingly. Deformed morphologies—on which previous work amounted to little more than the much reproduced early photographs of drawn spherulites, samples which I remember Andrew making, pulling strips over the Kofler hot bar with his fingers—received a thorough study 'in terms of spherulites' by Ian Hay [31,32], who started his PhD research early in 1962. The whole area of mechanical properties came to the fore in 1965 when the then Dr I.M. Ward joined Andrew at Bristol for what was to be a four year stay. Much excitement was generated subsequently by the search for stiff and strong polyethylene fibres [33,34] with the elegant work of Frank, Keller and Mackley [35], initiating substantial insight into crystallization under elongational and well-defined flow conditions.

As the polymer physics community grew so did the need for a suitable forum for meeting and discussion. An initiative from Keller and Ward, supported by Professor A Charlesby, Dr G. Allen and Dr D.H. Whiffen led, in April 1964, to the first of eight Biennial Meetings at Shrivenham and the formation of the British Polymer Physics Group, with Professor Frank as the first chairman. Later, this became the Polymer Physics Group, jointly affiliated to The Institute of Physics and The Royal Society of Chemistry, whose continuing but now peripatetic Biennial Meetings remain the principal UK forum for the subject.

We can now recognize that by the mid-sixties the major components of the modern Bristol Group and UK polymer physics, to which Andrew was central, were in place. Of course, his influence was not merely national but worldwide. At the time of his formal retirement in 1991 the number of Andrew's one-time students and co-workers was well into three figures. As we have seen, from the outset these were drawn from many countries. Many of these now hold senior positions in industry and academe around the globe, not least in the UK, the Netherlands and in Japan. Andrew was also eclectic of ideas; if he saw a good problem to which he felt he could contribute, then he contributed. The list is long, too long to describe fully here, extending from collagen through liquid crystal polymers, gelation, block copolymers, neutron, infrared and Raman scattering to computer modelling, real-time SAXS and to the synthesis of monodisperse long n-alkanes that, as he rightly foresaw, are indeed able to take polymer physics significantly forward.

There are few areas of structure and deformation in crystalline polymers to which Andrew did not contribute. I consider one of his major strengths to be his use of microscopy, if possible complementing other means, on well-defined systems to establish and underpin the textural models of structure/property relationships. For, as he understood very well, the physical model needs to be right for the more popular diffraction studies to have significance. And real-space studies have an unequalled record in establishing those physical models. He was a polymer morphologist, par excellence.

Andrew always made sure he was very well informed about new ideas and where the best work was being done. Indeed, I recall how much he would complain to high authority, forty years ago, if the latest issue of the Journal of Polymer Science was delayed in reaching the University library. But there was more, much more than that. He had a rare ability to enthuse people. In discussion he would make them feel that they, and he, were engaged in important research at the centre of the subject. The twin feelings of excitement and of being at the centre of things were also conveyed by his lectures. His characteristic mannerisms, still-Hungarian English, idiosyncratic, rather breathless delivery and undoubted panache made his lectures both entertaining and always informative. Andrew was unique and in continual demand to the end. He earned and deserves an honoured place in the history of our subject.

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