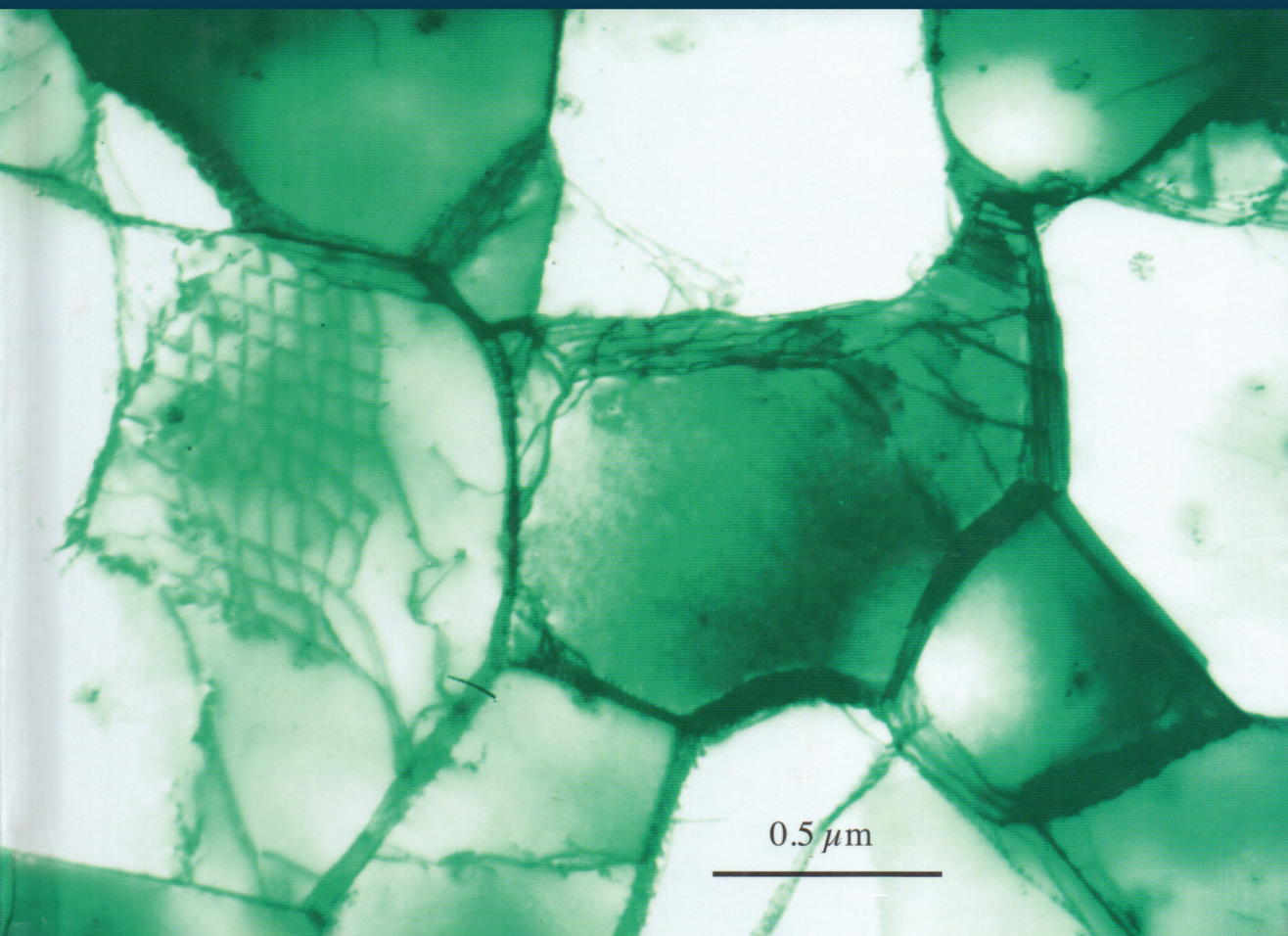
UNDERSTANDING MATERIALS

A Festschrift for Sir Peter Hirsch

EDITED BY C. J. HUMPHREYS



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Edited by
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Contents

Preface

This is a very special book for two reasons. First, it is a tribute to Professor Sir Peter Hirsch from his students, colleagues and friends. Second, it is a collection of specially written review articles by world-class scientists that take the readers from the origins of modern materials science through to the cutting edge of the subject in the twenty-first century. The book will be a valuable resource for all researchers in materials science, particularly those specialising in electron microscopy and diffraction, and in the mechanical properties of materials.

The front and back covers of this book are coloured images of historic electron micrographs depicting the first observation in the world of moving dislocations. The pictures were taken by Mike Whelan, then a research student of Peter Hirsch. The image on the front cover is before some dislocations have moved, and the back cover image is after the movement. See if you can spot the difference!

This book had its genesis in a symposium organised by Mike Goringe, John Hutchison and myself to mark the retirement of Peter Hirsch from the Isaac Wolfson Chair of Metallurgy at Oxford. This symposium brought together a large number of Peter's former students and colleagues. Some of the most distinguished of these have now written the chapters in this book. The opening chapter, by Professor Ugo Valdrè, provides a fascinating biographical sketch of Peter Hirsch from his early career in Cambridge to his retirement in Oxford. It contains many illuminating insights into the personality of Peter, both as a scientist and as a man. The next two chapters focus on the development of electron microscopy and diffraction. Professor Mike Whelan gives an eye-witness account of the seminal early work of Peter and his colleagues at Cambridge on the first observation of dislocations and their movements, using transmission electron microscopy. Professor Archie Howie extends this account to the present day, describing nanometer-scale resolution in scanning electron microscopes and atomic scale resolution in the scanning tunnelling microscope.

The following three chapters present a detailed review of three key techniques in modern electron microscopy: the weak-beam method, convergent beam electron diffraction and high resolution electron microscopy. These chapters should be required reading for all electron microscopists because they contain a wealth of information on the practice and the pitfalls of these important methods, which is not easily found elsewhere. Professor David Cockayne, the discoverer of the weak-beam technique, gives a masterful survey of its origins and evolution. Professor John Steeds describes the large angle convergent beam electron diffraction method and in particular its application to the characterisation of defects in crystals, an area in which he has made key contributions. Professor Hashimoto (Hashi to his friends) discusses the development of high resolution electron microscopy, a field in which he has made seminal contributions, giving many examples and including high resolution images formed using inelastically scattered electrons.

Not only was Peter Hirsch hugely influential in the development of electron microscopy techniques, he was above all a user of these techniques to understand and then solve real materials problems, in particular problems related to dislocations and the mechanical properties of materials. The chapter by Professor Mick Brown revisits a classic problem in the early days of dislocation theory, that of cross-slip, and presents his latest highly innovative thoughts on this topic. Professor John Spence applies quantum molecular dynamics to the long-standing problem of predicting fracture toughness and brilliantly demonstrates the power of this ab-initio method to solving complex real materials problems. These two chapters should be required reading for all those working on the strength of materials.

Dislocation sources are fundamental to plastic deformation and Professor Drahosh Vesely gives an overview of the operation of Frank–Read sources based on very careful in-situ deformation experiments in an electron microscope. Professors Duesbery, Vitek and Cserti then provide a comprehensive account of the plastic behaviour of bcc metals and alloys. Professor Terry Mitchell describes the deformation of oxide ceramics in a wide-ranging survey giving the latest results. The deformation of another ceramic, the important material silicon carbide, is then reviewed by Professor Pirouz, and Sialon ceramics are covered in detail by Professor Mike Lewis. Professor John Humphreys then leads us back to the ductility of metals and how they can be strengthened using particles.

The book ends with examples of how an understanding of materials can enable us to solve real engineering problems. First, Professor Brian Eyre describes the key role that understanding materials and their defects has placed in the development of nuclear technology, a subject in which Peter Hirsch has had a major involvement. The final chapter by myself is on designing materials and it gives two recent examples of the power of modern materials science: first, to improve substantially the efficiency of steam power plant and second, to create an entirely new form of lighting for the twenty-first century.

The book closes with the *curriculum vitae* and the publication list (to 2001) of Peter Hirsch. I write 'publication list (to 2001)' because, of course, Peter is still publishing in his retirement. This CV and list of publications, many of them seminal and brilliant, only do partial justice to the life and work of Peter Hirsch. Peter has not only fundamentally changed our understanding of materials, he also created the Metal Physics group in the Cavendish Laboratory in Cambridge University. The influence of this group can be seen from Figures 7 and 8 in the first chapter by Ugo Valdrè. Many of the people pictured here moved on to occupy leading positions in universities, research laboratories and industries throughout the world, like missionaries spreading the word they had received at Cambridge. Probably above all, Peter took the small Department of Metallurgy at Oxford University and transformed it into a lively, vigorous and world class Department of Materials, excelling over a broad range. He appointed a marvellous team of staff and students whom he led both firmly and kindly and often with a bubbling smile.

We wish him and his wonderful wife, Steve, many continued years of happy and active retirement, which they both richly deserve.

Colin Humphreys
Goldsmiths' Professor of Materials Science
University of Cambridge
Department of Materials Science and Metallurgy

Understanding Peter: A Recollection of Facts and Anecdotes about Sir P. B. Hirsch

UGO VALDRÈ

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1. THE FIRST ENCOUNTER

In 1954 I was put in charge of the Electron Microscopy Laboratory (EM Lab) of the University of Bologna, in addition to my normal research duties in nuclear physics. The EM Lab foundation dated back to 1948 and was the first EM laboratory to be established in an Italian University.[4] It was equipped with an electrostatic type of transmission electron microscope (TEM) of poor capabilities and it was, at the time, a regional facility; it provided a service, in ultramicrotomy and electron microscopy, essentially to biologists and physicians. The Director of the Istituto (now Dipartimento) de Fiscia 'A. Righi', a single chair institution at that time, was Professor G. Puppi, a high energy physicist of broad views.

About five years later, Puppi managed to get from the Ministry of Education a Siemens Elmiskop I TEM. The following year, Puppi decided to establish a research group in Solid State Physics (SSP). I was asked to apply for a NATO Fellowship, in order to be acquainted with the latest TEM techniques, in particular, those related to the study of thin crystals. The results obtained in 1956 by Hirsch, Horne and Whelan [2,3] on the observation of 'worm-like' features in thin metal specimens of Al (and in stainless steel by Bollmann [4], the filming of their movement, their interpretation as dislocation and, subsequently, jointly with Howie, the development of the dynamical theory of electron diffraction, had a decisive influence on my choice.

The excitement of the scientific community at the news of the first direct observations and interpretation of the crystal defects by TEM made such a big bang that it spread even over and south of the Alps where, usually, only the achievements of the high energy physicists seem to be the subject of attention. Since I did not know personally anybody in Cambridge, I expressed the wish, in my NATO application, to work at the Physics Department. I was therefore referred to Prof. N. F. Mott, the Cavendish Professor, once the NATO Fellowship was granted.

After some correspondence, I went to Cambridge to meet Sir Nevill Mott at the end of the 1960 International Union of Crystallography Congress, which took place in Cambridge at the Arts School and in, what is now called, the Old Cavendish Laboratory of Free School Lane (Fig. 1). He offered me two possibilities. Either I could spend my fellowship in the Crystallography Group (Headed by Dr W. H. Taylor)

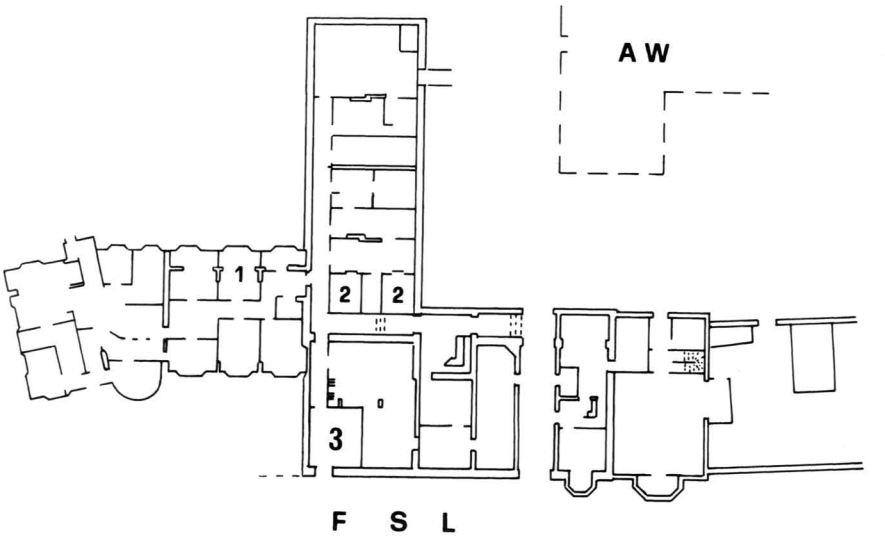

Fig. 1 Plan of Maxwell building at the Old Cavendish Laboratory (Free School Lane), adapted from *Nature*, 1874, **X**, 158. (1) Site where the Elmiskop I of the Electron Microscopy Group was installed and where the first observation of dislocations took place in 1956. (2) Location of the two Elmiskops used by the Crystallography Group first and by the Metal Physics Group later. (3) Polishing room. AW = Austin wing; FSL = Free School Lane.

under the supervision of Dr P. B. Hirsch, or at the Metallurgy Department, now the Department of Materials Science and Metallurgy. Sir Nevill introduced me to both PBH and Professor Alan Cottrell, former head of the Metallurgy Department.

I briefly described to Dr Hirsch the research project I had in mind: a comparison between the various thinning techniques of specimen preparation for TEM. PBH immediately said he was interested in it, although in modified form. Why? At that time solid state physicists and metallurgists could have been roughly divided, for the purpose of these notes, into two groups: the conservatives, who believed in the well-established replica and etch pits techniques, and the progressives, who had faith in the novel techniques of TEM. The Cambridge group obviously belonged to the latter, although conscious of the arguments made by the traditionalists on the possibility of a dislocation rearrangement during thinning. Dr Alfred Seegar of Stuttgart can be said to have been the most stubborn defender of traditionalist views. It was therefore important to assess up to which point the deductions made on TEM images of defects arrangements were valid in working out theory, particularly the work hardening theory, where Hirsch and Seegar were competing. The interest of PBH in a critical analysis of the polishing techniques was dictated, therefore, by this international scientific situation. The dispute eventually led, in the view of some colleagues, to the chair for Peter in Oxford and for Alfred in Stuttgart.

I gladly accepted Peter's proposal of using the method of the slip traces as a detector of dislocation rearrangement during and after thinning.

2. EARLIER WORK AT THE OLD CAVENDISH (FREE SCHOOL LANE)

I started my research work, in November 1960, with the understanding that I would carry on my teaching duty in Bologna by frequent visits there.

I was not accustomed to the working style of the Cavendish research students, who were practically trained between themselves, the junior by the senior, with frequent interactions with technical assistants.

After five weeks of hard work, time came to close the Cavendish Laboratory for the Christmas and New Year holidays. Before leaving for Bologna, I wanted to show and discuss with Peter the results I had obtained, so I went up to his office, where he was securely protected by the Group secretary. (He has always been very busy). I received permission to enter and began, by saying 'I am leaving tomorrow...'

Before I could reply with something like 'Hence, I came to see you to report on the work I have done so far', Peter raised his right arm to shake hands and said: 'Good-bye'. 'Oh, no', I replied, 'I came to show you my first results on the rearrangements of dislocation'. Peter fixed an appointment and in the afternoon we had a long session.

Soon after I presented the results, he said: 'Did you look at more than one specimen?' 'Thirty six', I replied. '36?' he was stunned. 'I have been working for over one month!' I explained.

At the end of the session, he concluded, 'It's a good Christmas present', with such an inflexion in his voice that I still don't know if he was really pleased by the results!

As time for the publication came, over one year later, I prepared a draft of the paper and listed the author's names in alphabetic order. When Peter saw it, he insisted my name be first, giving the reason that otherwise 'They will think I have done the work' – the implication being 'they' might have thought the results were biased. This expedient, however, did not convince Alfred and his Group, since the earliest reference to that paper was given as Hirsch and Valdrè!

However, the major outcome from my NATO Fellowship period was, by and large, the development of the first high angle double-tilting specimen stage.

At that time, a group working on transmission electron microscopy of thin crystals within the Crystallography Laboratory was formed by PBH. It consisted of PBH himself as Lecturer; Research Fellows, M. J. Whelan and A. Howie, L. M. Brown, who had just arrived from Birmingham, some other post-docs at the point of leaving, a few research students and three assistants – a photographer, an instrument maker and the technician looking after the maintenance of the two Siemens electron microscopes (models Elmiskop I and Ia). We were based in the Austin Wing, while the microscopes and the polishing room were in the Maxwell building (Fig. 1).

Events developed in the following way. During late Spring or early Summer 1961, Peter went on sabbatical leave in the USA. Since their marriage, in 1959, Peter and

Steve had been living in Steve's house in Gilbert Road. While the family was away, the house was offered to my wife and me, and we stayed there during August and September.

Very few people were around the laboratory in the mid-term, but Archie Howie was there and we had plenty of opportunities to talk. On one occasion, Archie told me about a longstanding problem: the necessity of a double-tilting specimen holder to exploit the predictions of the diffraction contrast theory of crystal defects. He also informed me of the imminent construction of a rather complicated type of goniometer. Since I had already acquired experience in the construction of multi-function specimen stages for the Bologna electron microscope and because of my initial training as a mechanical engineer, before getting my degree in physics, I thought it should be possible to produce something simpler and decided to try a different approach.

By lucky coincidence, we had the same make of electron microscope in Bologna, though a model marketed in between those of the Cavendish, so it was possible to test the double-tilting cartridge at home. I went back to Bologna, on 13th June 1961, and when I returned to Cambridge, on 8th July, I had with me a working double-tilting stage.

When Archie and I tried to install the double-tilting stage, we discovered that the old Siemens Elmiskop did not have access facilities for direct control of the specimen tilt, since the access hole available was at the level of the objective aperture, rather than at the stage level. (We did not dare to try the stage in the new Elmiskop, which was otherwise suitable to the exercise, since the rule was: it must, absolutely, be left for conventional use). The only way to operate the cartridge was by drilling a hole in the top plate of the objective lens, to provide a path between the specimen chamber and the hole at the level of the objective lens gap. Since the hole had to be rather large (about 11 mm) and close to the polepieces, there was the risk of creating an uncorrectable astigmatism. We were, therefore, very much concerned with the possible outcome.

Archie sent a letter to PBH for information and guidance, and, at the same time tried to contact Mike Whelan in vain. The answer did not come. Although a recent PhD graduate, and probably not the senior person, Archie nevertheless was to stay in the PBH group. After a reasonable time had elapsed, he took responsibility and decided to go ahead. With the assistance of our fine instrument maker, Freddy Percival Marks, the hole was drilled, the drive completed and the double-tilting cartridge successfully tested. We were, however ready to fill the hole in case things did not go right!

Peter appreciated my expertise as specimen stage designer, which filled a gap in the Crystallography Group and proposed the development of several types of stages, notably, double-tilting + heating, double-tilting + cooling at liquid helium temperatures and magnetisation stages (Fig. 2), which were constructed and widely used. The double-tilting liquid He stage was particularly successful for the wealth of original results obtained jointly with M. J. Goringe and the other research students (Fig.3). Peter drew the attention of the firm Siemens & Halske on my activity. This triggered

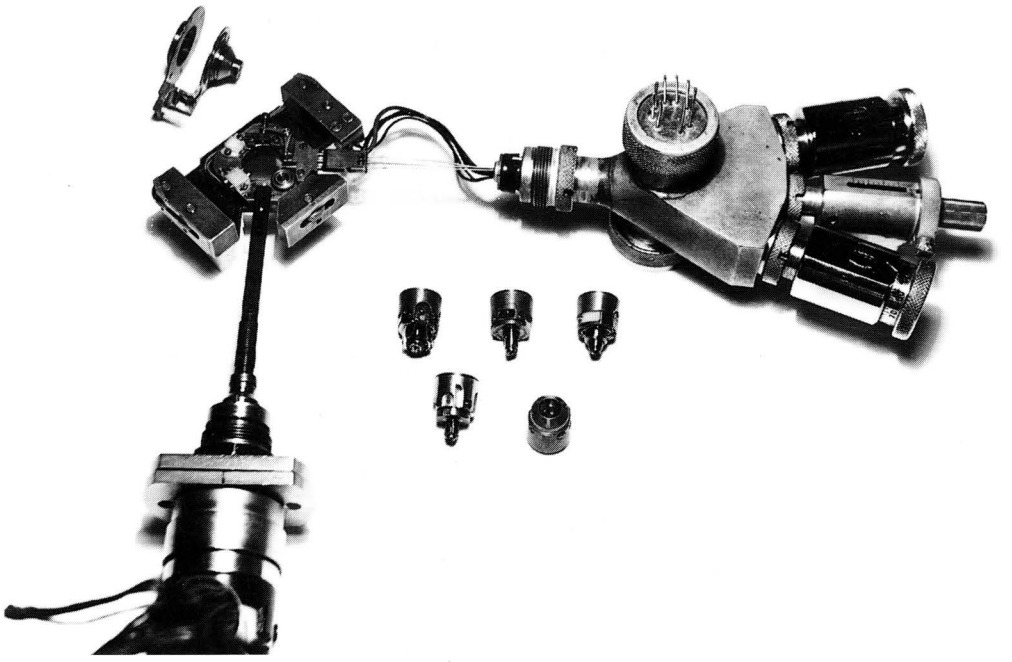

Fig. 2 Specimen stage with drives, selection of cartridges, cold finger and decontaminator.

an agreement with Siemens AG to use my patents. Similarly, through Peter, Chairman of the Users Committee of the AEI EM7 microscopes, I was invited to collaborate in the development of specimen stages for that high voltage electron microscope (IMV).

3. PETER'S NEW HOUSE

Around 1964, the builders started the construction of Peter's new house at 14a, Hills Avenue, Cambridge. This took place almost simultaneously with the construction of a house on my wife's farm, near the town of Faenza, in Italy. In our case we intended to spend weekends and holiday periods at the farm site for business reasons. The Hirsch's never believed it was for that purpose and the challenge for finishing first was so felt that in appalling weather, the whole Hirsch family (Peter, Steve, Janet and Paul) inspected the farm site during a visit to Italy and San Marino. With rain from above and mud from below, they checked the progress of the work; gladly noting my house was not going to be finished first. And so it happened.

Peter worked closely with the architect in order to shape the house according to his own taste, as I did with mine. Did these meetings sow the seeds for what later became user's meetings to discuss the design of electron microscopes? As I under-

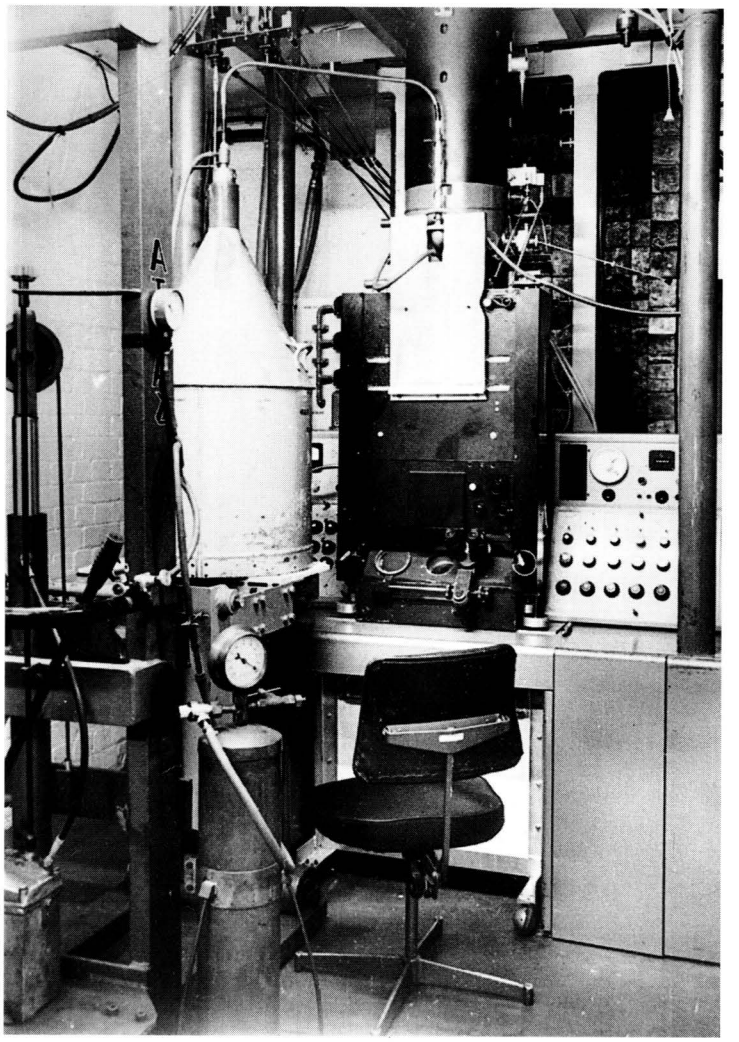

Fig. 3 The He stage mounted in the Cambridge High Voltage Electron Microscope (750 kV).

stand it, although extremely important, neither the microscope manufacturers nor the architect liked this type of approach.

I know that Peter disagrees with what I am about to write, but this is how I remember it. I recall a Peter–Steve conversation and bits of chat from other people. I might have misunderstood, of course, but I am convinced that Peter designed that part of the house which became his study. The idea was that the house should look like a battleship, with Peter's study being the prow. It is clear from Plates I and II that the front or west side of the house is wedge-shaped and the study window overlooks the entrance path. A mischievous fellow said Peter would then be alerted of the arrival of any unwanted person, - a research student, perhaps, – and could act promptly....

Plate I Hirsch's new house in Hills Avenue, Cambridge: the 'Prow'.

Plate II The 'thin column' supporting the prow and the later addition of a suitably tall plant in order to mask it.

Plate III Photograph from the front side of the new building of the former Department of Metallurgy in Parks Road, Oxford.

The column supporting the prow was allegedly designed by Peter. It is certain he checked its stability. It looks aesthetically thin, but was definitely safe 'a posteriori', since Prof. Sir John C. Kendrew, who bought it later from Peter, enjoyed it for a long time, although he masked the column behind a tall plant (Plate II).

4. SUMMER SCHOOLS

In July 1963, a Summer School was organised in Cambridge by the local physicists and metallurgists; it was the result of an enquiry made by the Institute of Physics, which pointed out such a necessity in the newly developed field of TEM of thin crystals. The school was oversubscribed and a great success. The lecturers (PBH, Archie Howie, Robin Nicholson, Don Pashley and Mike Whelan) were invited by the participants to organise the lecture notes in a coherent book. The book did come out but only in 1965; it is usually referred to as the 'bible', since it contains the basics of

Plate IV Peter and Steve.

diffraction contrast TEM and many applications to Solid State Physics. Guess who was the last author to provide his manuscript.

The same year also saw the election of PBH to Fellow of the Royal Society. This brings to my mind a connection with a journey to London to attend a speech given by Professor John W. Steeds, a research student at the time. Peter, Archie and myself decided to go in my car to save money and time. When we arrived, Peter and Archie went immediately to the lecture hall since we were late, leaving me to look for a suitable place to park my car. There wasn't one in view. I decided to park my Fiat 1500 in the courtyard of the Royal Society. To leave space for other vehicles, I left my car with two wheels over the large staircase leading to the main entrance of the Society. I then joined Peter and Archie.

At the end of the talk, we came out of the hall and Peter was horrified by my parking situation. Nearby, a custodian was trying to find out who had committed the offence. I climbed into my car; followed by Archie. Peter waited in apprehension, scrutinising the people present. When he thought it was safe, he jumped in with unsuspected agility. We left with no other problems. I suspected that Peter had thought he might not have been made an FRS if some Fellows had seen him, somehow involved in what night have seemed an insult to the learned society.

In the following years, the progress of electron microscopy towards atomic resol-

ution called for the development of instrumentation and experimental and theoretical means to deal with phase objects. I felt the necessity of another Summer School. It took place at Erice, Sicily, in April 1970, at the recently established Ettore Majorana Centre for Scientific Culture, which was the sponsoring institution, in conjunction with NATO and the Electron Microscopy Laboratory of Bologna. It was again oversubscribed.

In order to have a coherent and reliable group of lecturers; I invited my Cambridge and Oxford colleagues, in addition to a group of scientists who had been working at the forefront of phase contrast electron microscopy. Peter was asked to deliver the introductory lecture. It was on this occasion that I discovered that he could address an audience as if he were talking off the cuff, while, in fact, he was reading. When I asked him to provide a manuscript of his contribution, Peter refused. I told Peter we had recorded his speech and asked him if I could make a typescript of it for the Proceedings of the School. He then handed to me *'seduta stante'* the *'notes'* he was reading, which are those reproduced in the book. [6]

5. THE METAL PHYSICS GROUP AND THE OXFORD CHAIR

The offer to Dr Hirsch of a chair at the University of Warwick promoted, I believe, the foundation of the Metal Physics (MP) Group and gained PBH the Readership in Cambridge, in 1964. Until then Peter's Group formed part of the Crystallography Laboratory. The laboratory was the last heritage of former Cavendish Professor Sir W. L. Bragg. At Sir Lawrence's resignation, to take the position of Director of the Royal Institution in 1953, the new Cavendish Professor, N. F. Mott, suggested the activity in the structure determination by X-rays should find a more appropriate location and scope in other premises.[17] So it happened, and the achievements of that group are well documented and known. The scientific interest of Sir Nevill was the application of quantum mechanics to solid state physics; the branch of science which emerged in the nineteen twenties, to which he contributed so much, not only scientifically and culturally, but also politically. The MP Group grew fast, attracting many research students and visitors.

Then came the offer to Dr Hirsch of the Isaac Wolfson chair of Metallurgy at the University of Oxford. The news followed by a long period of consultation, bargaining and a visit to the new place. Peter would like to have moved all his Group to Oxford. About two-thirds of it eventually joined him: R.G. Booker, M. J. Goringe, P. Hazzledine, C. J. Humphreys, (the late) J. P. Jakubovics, M. J. Whelan. The ability and diplomacy of Peter was very successful. He built, in Oxford, a comprehensive laboratory, where the investigation of the structure of matter is performed with practically all modern methods; some developed by the Oxford team.

Peter asked me to join him in Oxford on a three-year grant. I did not accept because my position in Bologna was not permanent and I would have risked being cut off from it forever if I had to return later; sabbatical leave was not applicable. In addition

there were the problems of my wife's job as a teacher and those that the move would have created in a family with four young children. I did not inquire on how Peter would have seen my more distant future or the possibility of a permanent position in Oxford, since several research students were to finish soon and certainly they would have wanted to stay in research (as it happened).

To celebrate the professorship, the MP Group decided to offer Peter a silver plate. Engraved on it were symbols of PBH's scientific career (Fig. 4). References can be seen to his initial work on the microfocus X-ray technique, the column approximation, the emblems of the kinematical and dynamical theory of electron diffraction contrast and also to jog theory. To mark the occasion, Reg Brundish, the Group photographer at that time took pictures of the current members and dug out pictures of past PBH collaborators, either as research students or visitors. They were collated together mosaic-like, with the period of membership indicated (Fig. 5). Some photographs unfortunately remained unavailable. The re-edited version of Fig. 6 rejuvenates the work on dislocation.

I think Peter was moved at the presentation of 'his' Group. Some envious Americans used to call it 'That little group in Cambridge'.

An extension to the old Metallurgy building was constructed in Oxford, where Peter has his study, facing Parks road (Plate III); also a new building was erected to house the High Voltage Electron Microscope, the AEI EM7. More junior people, fresh money, more university positions and space were going to transform 'Little

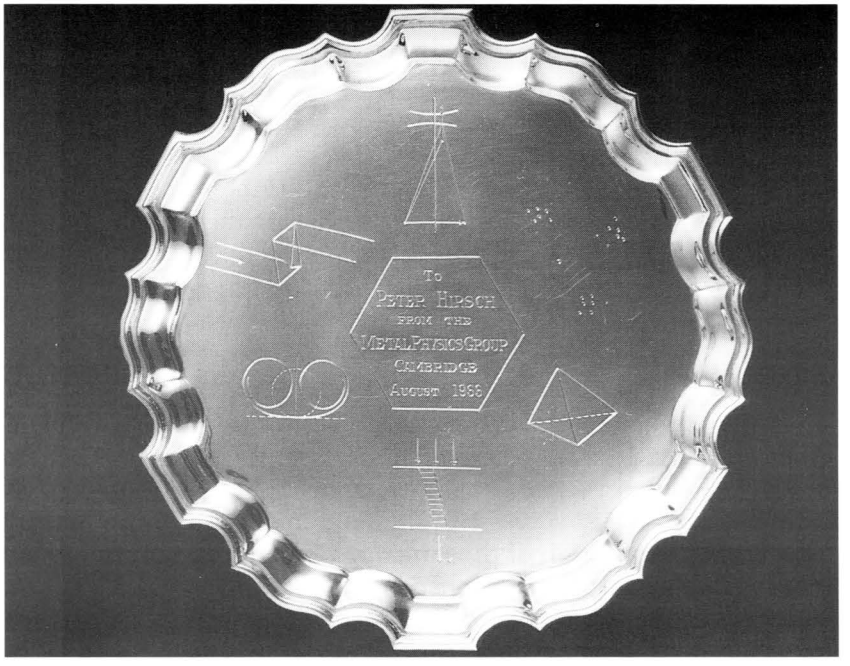

Fig. 4 Engraved silver plate with symbols of some of Sir Peter's scientific career.

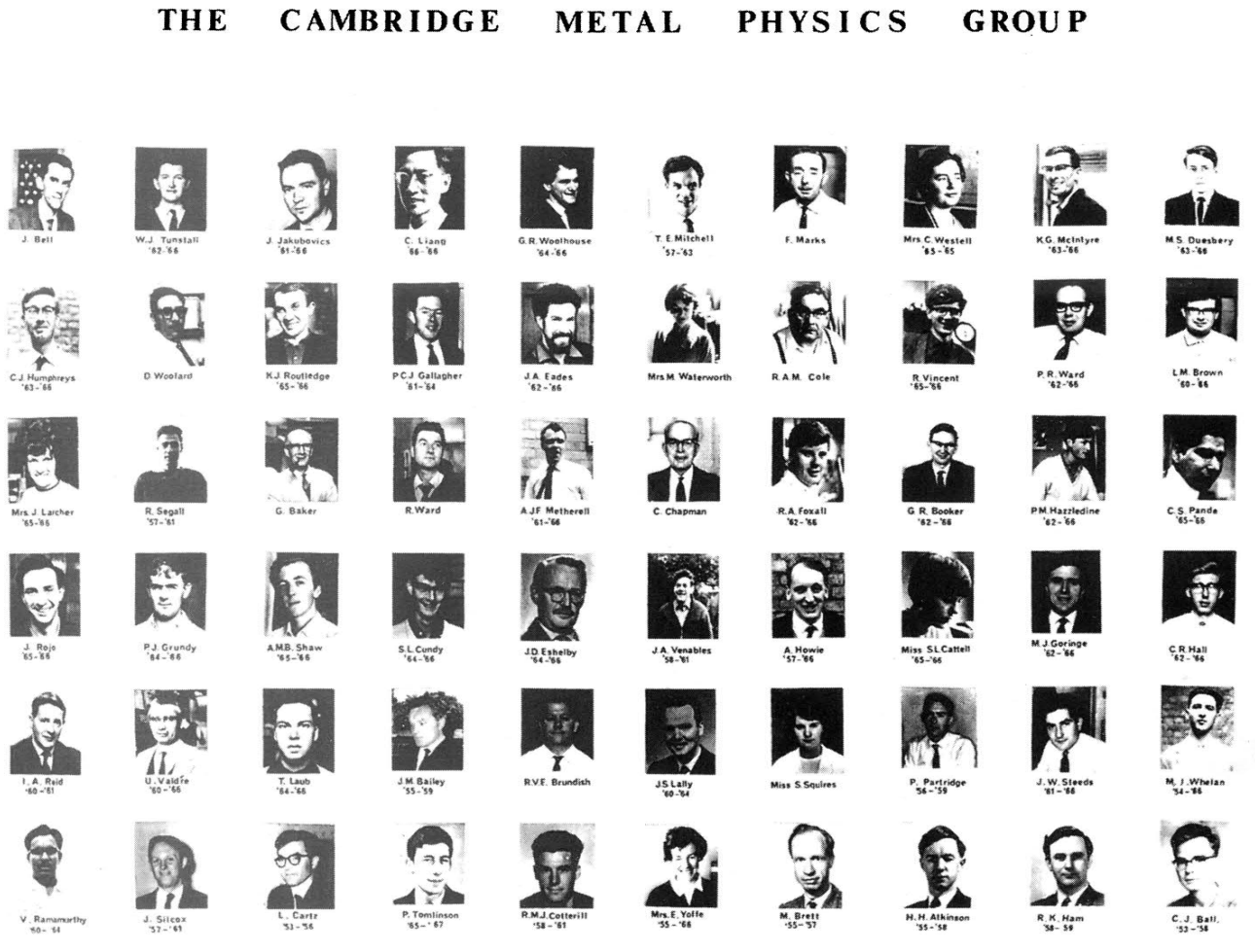

Fig. 5 Montage of MP Group members up to 1965.

Fig. 6 'Dislocation' in the MP Group 'mosaic' structure.

Cambridge MP Group' to an establishment. However, the tiny spark produced at the Cavendish at the heroic time of few resources, hard overnight work and home-made equipment had already produced a large fire all over the world.

6. FAREWELL PETER

For taking licence with Peter, Steve (Plate 4) and my readers, and for having dared enter unfamiliar not scientific fields with my limited vocabulary, I wish to present my apologies. I wish also to summarise what is scattered and somehow hidden in the above memoir about Peter's personality.

I cannot resist from noting that Peter doesn't like to write review articles and even letters, and we are poorer for it. It is unfortunate that he is always busy and that one has to fix formal appointments when more casual contacts would be enjoyable, But I must say Peter is gifted in diplomacy, management and raising funds; how could he otherwise have succeeded in building a scientific empire? He can grasp immediately the importance of new ideas or developments and is quick in taking decisions. He realises the specific talent of people, encourages and helps them; in return he takes advantage of their expertise for the benefit of all the group. Though he could be demanding within the Group, he has never, in my experience, criticised members of his Group publicly, even within the Cavendish, rather he has always emphasised their qualities and their work to the outside world. He has no prejudice against people with respect to their origin. Those make him an exceptional manager.

The days of the celebration of Professor Hirsch's retirement from the Isaac Wolfson Chair (14–15 September 1992) must have told Sir Peter how much his leadership has been fruitful and appreciated via the large participation of old students and co-workers, far more than the knighthood he received from Her Majesty the Queen in 1975.

ACKNOWLEDGEMENTS

My grateful thanks are due to Mrs Margaret Pennycook for her help in clarifying my English. I also wish to thank the various Institutions (NATO, Cavendish Lab., Istitito di Fiscia, University of Bologna, SRC, CNR, MPI, EEC, INFM. . . .) which over the years supported my joint research work with the MP Group.

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*The Early Observations of Defects in Metals by Transmission Electron Microscopy**

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ABSTRACT

An account is given of the research work carried out at the Cavendish Laboratory, University of Cambridge, which led to the observation of dislocations and their movements in thin metal foils in the mid-1950s.

1. INTRODUCTION

Contact with young research workers in our Department about contrast features arising from the interaction of a moving dislocation with an oxide coated surface (see §2B), made the author realise that a generation has grown up that have only a hazy knowledge of how the technique of transmission electron microscopy (TEM) of defects in crystals came to be developed. So it should not be out of place in this volume to start with a review of the early work and to recall some of the startling observations (for that time) which were made in Peter Hirsch's group in the Cavendish Laboratory of Cambridge University in the mid-1950s.

Electron microscopes were constructed in a number of laboratories in the 1930s and by 1939 the German company, Siemens and Halske, had begun the manufacture of an electron microscope whose successor in post-war years (the Elmiskop 1) had a large share of the world market (for review see Ruska[1]). But it was not until after World War II that serious metallurgical applications of the electron microscope could begin, and the immediate postwar use was mainly concentrated on the examination of surface features by replica techniques. There were of course exceptions to this. Following on numerous earlier TEM investigations of small crystals such as MgO smoke particles, Cr particles MoO_3 (Kossel and Möllensted,[2] Heidenreich and Sturkey[3]), Heidenreich[4,5] developed an electrolytic polishing method of thinning deformed aluminium and observed subgrains of size ~1 or 2μm, which had also been

* The author is grateful to Wiley-Liss, a division of John Wiley and Sons, Inc., for permission to reproduce material from an article on the same topic, entitled 'Reminiscenses on the Early Observations of Defects in Crystals by Electron Microscopy' by M. J. Whelan which appeared in the *Journal of Electron Microscopy Technique*, 1986, **3**, 109–129, edited by J. E. Johnson, Jr., © 1986 Alan R. Liss, Inc.

observed indirectly by X-ray microbeam diffraction (Kellar et al.,[6] Hirsch and Kellar[7]). Heidenreich[4] also applied the dynamical theory of electron diffraction to the interpretation of the thickness- and bend-extinction contours observed in the aluminium foils. This was a particularly important advance since it demonstrated the power of the TEM in revealing crystalline effects by direct observation of thin metal sections. Metallurgists at that time did not take much notice of this work, and even Heidenreich himself left the field. Furthermore, he did not report any evidence for dislocations on his micrographs. Mention should also be made of the pioneering work of Castaing,[8,9] who studied by thin foil TEM precipitation in aged Al–4%Cu alloy and reported evidence for GP zones and precipitation, but not for dislocations. Castaing[9] also developed the ion beam etching technique which was later to become widely used for thinning materials not easily thinned by other methods.

2. DEVELOPMENTS IN CAMBRIDGE DURING THE EARLY 1950S

(A) THE PERIOD UP TO 1954

As already mentioned,[6,7] Kellar and Hirsch had during the period 1946–50 developed an X-ray microbeam technique for studying metal textures. The idea was that if the X-ray beam diameter were made small enough so that only a few subgrains were exposed, the Debye–Scherrer diffraction rings would be spotty rather than continuous, and from their study estimates of subgrain size, sub-boundary misorientations and hence of dislocation densities could be made. X-ray beam diameters as small as 10μm were difficult to achieve and exposure times were several hours. The work had been suggested by Professof W. L. Bragg, Head of the Cavendish Laboratory from 1937 to 1954, and it was carried out in the Crystallographic Laboratory of the Cavendish Laboratory and was supervised by Dr W. H. Taylor. After Kellar's death in 1948, the work was continued by Peter Hirsch and other students, such as J. S. Thorp, P. Gay and A. Kelly. The technique worked well for certain metals such as aluminium, in which deformation produced well-defined substructures of size ~1μm (see Fig. 1), but for other metals such as Cu, Ni, Au the subgrain size was either too small to be detected, or subgrains were not formed at all. Kelly[10] therefore investigated first a transmission X-ray microbeam technique, and later for beaten gold foil the technique of selected area electron diffraction in the electron microscope, which was a newly provided facility for postwar instruments. An electron diffraction pattern could be obtained in a few seconds from areas ~1μm in diameter or less with exposure times of only a few seconds. However, it was necessary to use a thin foil specimen ~0.1μm thick in order to have sufficient transparency for electrons.

At that time the Cavendish Laboratory had only two rather old electron microscopes, both in the Electron Microscope Group headed by Dr V. E. Cosslett, and neither was able to do selected area diffraction. The first was a 50kV RCA EMB instrument which had been obtained through the Lease–Lend programme of World

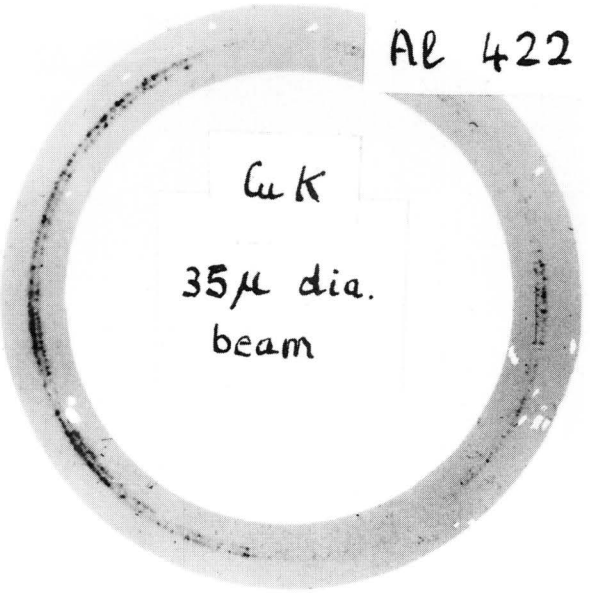

Fig. 1 Back reflection 422 Debye–Scherrer microbeam X-ray diffraction pattern of deformed Al (Hirsch and Kellar[7]). Courtesy of the *Journal of Electron Microscopy Technique.*

War II, and was about to be converted to a prototype microprobe analyser by P. Duncumb. The second was the 1939 design Siemens UM 100 (100kV) electron microscope which had arrived in the Cavendish Laboratory as war reparations from the Krupp Armament Works. For an account of the history of the Cavendish Electron Microscope group see Cosslett.[11] Since the selected area diffraction facility was not available on these instruments, collaboration developed with J. W. Menter of the Laboratory of the Physics and Chemistry of Rubbing Solids, a sub-department of the Physical Chemistry Department at Cambridge. Menter had an EM3 electron microscope manufactured by the Metropolitan Vickers Electrical Company of Manchester, which was able to perform selected area diffraction.

Although Peter Hirsch was working on X-ray studies of coal at that time, he collaborated with Kelly and Menter. A study was made of beaten gold foil and interesting streaks on electron diffraction patterns were observed indicating that the foil had a high density of stacking faults. This work, in which the emphasis was on electron diffraction effects, was published by Hirsch et al.,[12] and a successor paper was promised in which interesting contrast effects observed on the electron micrographs, as distinct from diffraction patterns, would be discussed. This paper was never completed, although some micrographs were published as a conference paper (Hirsch et al.[13]). Kelly left Cambridge for the University of Illinois, and Menter went to the

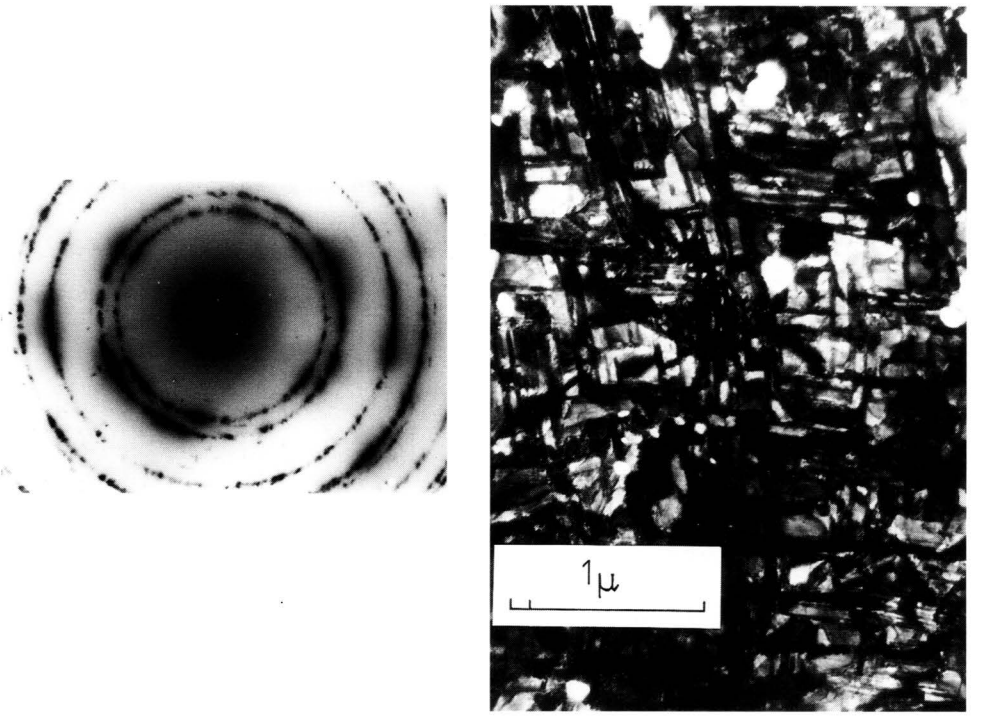

Fig. 2 (a) Selected area electron diffraction pattern of beaten gold foil from a region ~10μm diameter. Taken with Metropolitan-Vickers EM3 electron microscope at 70kV. **(b)** Transmission electron micrograph of beaten gold foil taken with the same instrument. (Hirsch, Kelly and Menter, unpublished work). Courtesy of the *Journal of Electron Microscopy Technique.*

newly established Tube Investments Research Laboratories near Cambridge, where he later made the first direct observations of dislocations by direct lattice imaging in platinum phthalocyanine.[14]

Figures 2(a) and (b) are typical of diffraction patterns and micrographs of the beaten gold foil. The foil had a preferred {100} surface orientation, and bands of dark contrast are seen in the micrograph approximately parallel to the traces of {111} planes on the foil surface. Peter Hirsch had the idea that a fault with a displacement vector **R**, cutting the foil obliquely as in Fig. 3, would cause image contrast because of the phase difference $2\pi\mathbf{g}\cdot\mathbf{R}$ between waves diffracted into the Bragg reflection above and below the fault plane, where g is the reciprocal lattice vector corresponding to the Bragg reflection. This is essentially a *phase contrast* mechanism, the phase arising from the displacement **R** of atoms from their original positions before the fault was introduced. This mechanism is common knowledge to a crystallographer (which Peter Hirsch was), but it would not have been so to most metallurgical users of the electron microscope at that time. Appreciable contrast can occur due to this mechanism even though the fault causes negligible foil thickness or density changes. This was a crucial idea, the

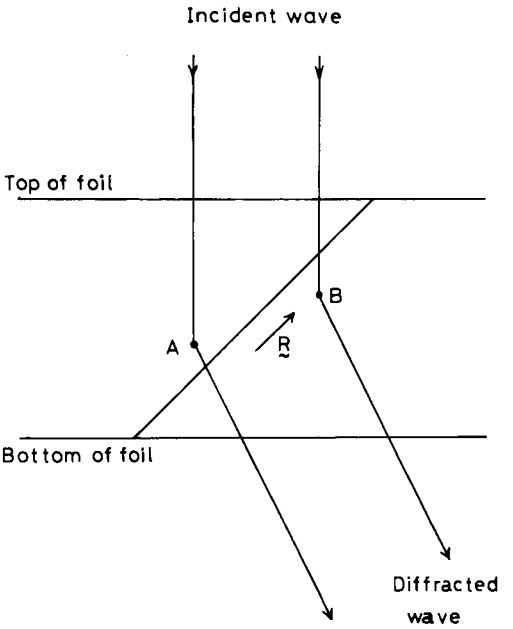

Fig. 3 Schematic diagram of an inclined stacking fault cutting across a thin foil. Waves scattered by equivalent atoms A and B on opposite sides of the fault are out of phase by $2\pi\mathbf{g}\cdot\mathbf{R}$. Courtesy of the *Journal of Electron Microscopy Technique.*

recognition of which stimulated Peter Hirsch to investigate the electron microscope method further. It is interesting to note that he and his colleagues in the Cavendish Laboratory, had not been engaged in electron microscopy until then, but were in fact X-ray crystallographers interested in the study of defects in otherwise simple metallic structures. In fact the entry into the electron microscope field was indirect, and initially the emphasis was entirely on extending the microbeam X-ray technique to electron diffraction, the interest in electron micrographs arising only subsequently as a by-product of the electron diffraction studies. It is also worth noting here that at that time metallurgical users of the electron microscope did not believe that it would be possible to see dislocations directly in the electron microscope because they associated contrast with density changes, and these would be insufficient. Furthermore electron microscopists in the Cavendish Laboratory were pessimistic about obtaining sufficient beam penetration unless the metal films were very thin (~100Å), despite the work of Heidenreich[4,5] and Castaing,[8,9] because of their experience with microcrystalline evaporated films of heavy elements used for shadowing. In fact it was recognised much later that special channelling effects operate in single crystal films, which lead to enhanced beam penetration, a situation quite different from that for shadowing films. Again it required a crystallographer's understanding to appreciate the importance of such channelling effects.

(B) THE PERIOD 1954–56

The author started work as a research student in the Cavendish Laboratory in October 1954. It was an auspicious time since the author's arrival in the Cavendish coincided with that of Professor N. F. Mott who had moved from Bristol to take up the Cavendish Chair as successor to Professor Bragg. He had an international reputation in solid state theoretical physics, including dislocation theory, and was keen to encourage the growth of metal physics in the Cavendish. Peter Hirsch was appointed as the author's supervisor, and he suggested that a fruitful field of research might be to investigate the possibility of imaging dislocations directly by TEM, following up the work[12,13] on beaten gold foil. He suggested that if a wide stacking fault produced contrast as in the beaten gold foil, then a dislocation might also be visible because, according to Heidenreich and Shockley,[15] a dislocation in the fcc structure could dissociate into two partials separated by a ribbon of stacking fault, which would be expected to produce contrast by the mechanism already recognised. The idea was just a hunch, and nothing had been worked out in detail. It was not known how to take account of the partial dislocations bounding the fault ribbon, but later it was recognised that even an undissociated dislocation gives contrast by virtue of its continuous strain field, so that the presence of a ribbon of fault is not necessary.

When the author started work no equipment for thinning specimens was available, nor did he have regular access to an electron microscope. So he set to work to construct an ion-beam thinning system from scratch, since Castaing's work[9] had seemed to suggest that this was necessary. To the present generation of research students, accustomed to an abundance of commercially made ion-beam thinning machines, it may be difficult to imagine having to make one's own. There was little money available from scarce department funds. However, there was a students' workshop at hand and a cellar full of junk electrical equipment in the stores, much of it war-surplus. With the help of the laboratory glassblower, a glass ion-beam source discharge tube, pumped by a glass mercury diffusion pump and glass U-tube liquid nitrogen trap, was constructed. The author himself machined from brass an airlock device for inserting the specimen into the ion beam. The H.T. supply for the ion source was constructed from a 5kV transformer, two high-voltage rectifier valves in a doubler circuit and some high-voltage capacitors. The valves had 4V heaters requiring a filament supply insulated from earth by several kV. Since no suitable filament transformer was available, pairs of lead-acid batteries stood on rubber sheet were used to heat the filaments. The heater circuits had to be switched on first before the Variac transformer was turned up. This crude system delivered 0–10kV at a current ~10mA, and was mounted on a 'bread board', the only concession to safety being a thick earth wire around its edge. The whole system was potentially lethal, and was made even more so by the fact that it was operated in a darkened room in order to see the glow of the ion beam, with one's head only a few inches away from the exposed H.T. electrode of the ion source. It would certainly not be permitted now by current safety regulations in force in the UK. Nevertheless the system worked well, allowing holes to be

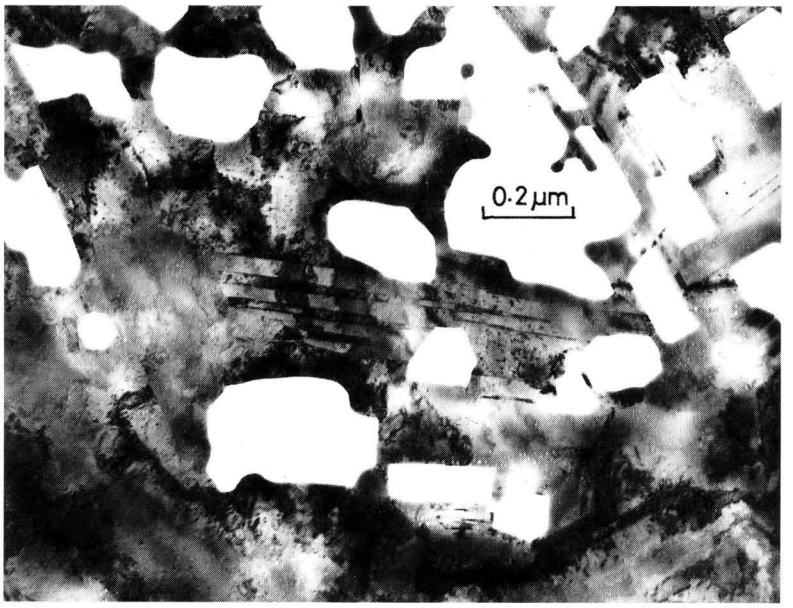

Fig. 4 Transmission electron micrograph of beaten gold foil after ion beam etching with 3kV air ions. Courtesy of the *Journal of Electron Microscopy Technique.*

etched in gold foil in a few minutes using air or argon ions at ~8keV and ~5μA beam current.

Figure 4 is a micrograph of beaten gold foil etched with 3kV air ions. In comparison with Fig. 2(b) the density of faulted regions is much reduced, suggesting some sort of annealing effect caused by the ion bombardment. Moreover, the numerous black or white spots visible were probably the first observations of ion damage in a metal. At that time such effects were not recognised, and they were usually attributed to 'dirt' or 'artefacts'. It is interesting to note that the holes shown in Fig. 4. tend to have crystallographic shapes with edges parallel to traces of {111} planes on the foil surface. Both crystallographic and non-crystallographic etching was observed. The gold foil used was the so-called 'fine' beaten foil 100 or 200nm in thickness which is used for ornamental gilding and was supplied by George M. Whiley Ltd of Ruislip. A visit was made to this company to see the ancient art of gold beating in practice, and the company kindly offered to beat several pure metals supplied by us. In particular aluminium was beaten to a thickness of 0.5μm. Further thinning of this foil by ion bombardment proved difficult because, as is well known, aluminium is difficult to sputter due to the strong surface oxide film. However, it proved possible to etch the beaten foil to a thickness of 100nm by immersion in 0·5%HF solution. Such foils were very suitable for transmission electron microscopy.

This work occupied the best part of a year from October 1954. By mid-1955 some specimens had been produced, but the next difficulty was to find an electron microscope with which to examine them. Menter's EM3 electron microscope in the PCRS

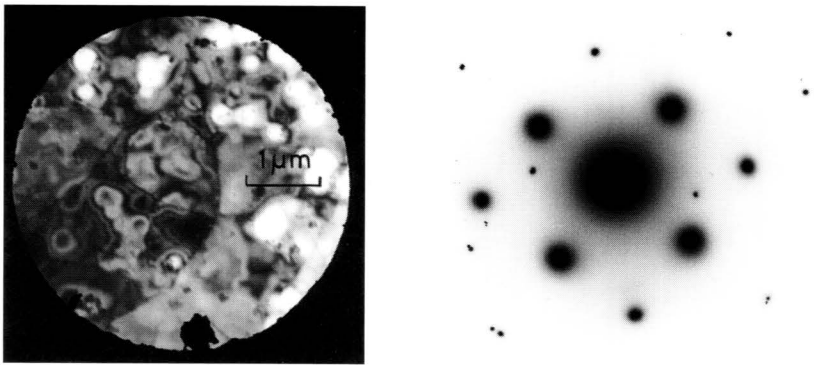

Fig. 5 (a) Transmission electron micrograph and **(b)** selected area electron diffraction pattern of beaten Al foil, annealed at ~400°C and etched in HF. Note the extinction contours and subgrain boundaries in (a) and the preferred (100) surface orientation in (b). Courtesy of the *Journal of Electron Microscopy Technique.*

Laboratory was still available, but after his departure its performance had deteriorated, and it was eventually put out of action by a fire in the electronics. Therefore a preliminary examination of ion-etched gold foil was made on a 60kV Elmiskop 2 electron microscope of Dr J. Nutting in the Department of Metallurgy. However, this instrument had relatively poor illumination and the selected area diffraction facility was not available.

At about the time the authors started research in October 1954, a new Siemens Elmiskop 1 electron microscope had been installed in the Electron Microscope Group of the Cavendish Laboratory, headed by Dr V. E. Cosslett. The instrument was in the charge of Mr R. W. (Bob) Horne. It had been exhibited at the London International Electron Microscope Conference in the summer of 1954, and was the first Elmiskop 1 to be delivered to a customer from the Siemens factory in Berlin. It was acquired with the aid of a grant from the Nuffield Foundation and was heavily used by biologists. Since it was in another group of the Cavendish Laboratory, the author was not aware of its existence until almost a year had passed. In mid-October 1955 he was able to get some time on the instrument. However, the amount of time was extremely limited. Initially only one three-hour session per fortnight was available, and it was not possible to use the instrument 'hands on'. Nevertheless, some interesting micrographs were taken, but the interpretation of some features was not clarified. In particular, the beaten aluminium foil annealed at ~400°C and etched in HF was examined, and the subgrain structure and extinction contours observed by Heidenreich[4,5] were seen (Figs 5(a), (b)).

Now, owing to a peculiarity of the design of the early Elmiskop 1, it was not possible to use the newly provided double condenser lens illumination system when doing selected area diffraction. This was because the intermediate and objective lens were normally energised in series from one current supply. For selected area electron diffraction, where a separately energised intermediate lens is required, a switching arrange-

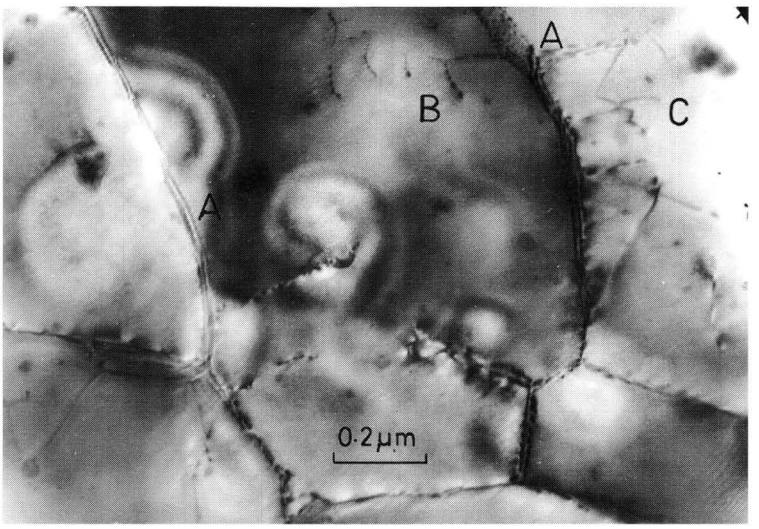

Fig. 6 Subgrain structure and contrast effects in beaten Al foil annealed at 340°C and etched in HF (Hirsch et al.[19]). Courtesy of the *Journal of Electron Microscopy Technique.*

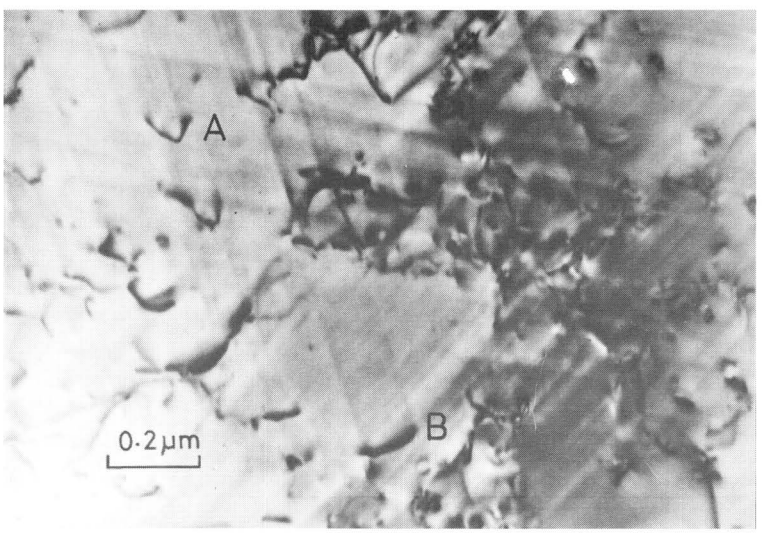

Fig. 7 A single crystal Al specimen deformed 5% and electropolished showing a bowed dislocation at A and a slip trace dislocation B. Courtesy of the *Journal of Electron Microscopy Technique.*