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Walter Eric Spear. 20 January 1921 — 21 February 2008

Alfred Adams

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Elected FRS 1980

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Walter Spear was a multi-talented individual who decided to make a career in science in the UK. After his studies in London he joined the academic staff of the Physics Department in Leicester in 1953. In 1968 he was appointed to the Harris Chair of Physics at the University of Dundee in Scotland, where he stayed until retirement. Although his work was primarily driven by a desire to understand the basic physics of electrical conduction in solids, his discoveries had considerable impact on the electronics industry. For example, he will be well remembered for his research on amorphous semiconductors, which resulted in a variety of applications within the optoelectronics industry, including solar cells and the liquid crystal displays (LCDs) used in televisions, computer monitors and a wide range of hand-held devices. His work was characterized throughout by high experimental skill, felicitous choice of materials and full theoretical understanding. Walter won many international prizes during his career, including the Royal Society Rumford Medal in 1990.

FAMILY BACKGROUND AND EARLY LIFE

Walter Spear was born on 20 January 1921 in Frankfurt on the Main, at that time one of Germany's most attractive historical cities. His father came from an old-established Jewish family who lived in the Odenwald, not far from Heidelberg. He was a graphic artist, although later in life he turned towards photography and became one of the pioneers of colour photography and processing. Walter's mother, the daughter of a Lutheran pastor, was a professional violinist, a well-known soloist and teacher in Frankfurt.

During the economically difficult years after World War I, Walter and his younger sister, Marianne, grew up in a somewhat crowded family apartment, occupied by their parents and maternal grandparents. It was a happy childhood and, as a young boy, Walter developed

close ties with his grandparents. His grandmother, a lively and gifted lady from Neuchâtel in Switzerland, taught him French from an early age, which proved to be a great asset in later life. Because of his mother's professional commitments, Walter grew up in an atmosphere of musical activity, which led to his lifelong enthusiasm and love for chamber music. At the age of eight years he began violoncello lessons on a half-sized instrument. A few years later he was fortunate enough to inherit a beautiful seventeenth-century Italian cello, which he played and cherished all his life.

After primary school he was accepted in 1931 to the 'Musterschule', a grammar school that stressed modern languages and the sciences rather than the traditional classical education. The teaching was enlightened and throughout the years Walter made good progress, particularly in mathematics and physics. In spite of the growing Nazi menace, which made his schooldays increasingly difficult, he was able to complete his final examinations (Abitur), achieving an overall grade of 'very good'. By then, however, it was 1938 and the Nazi persecution of Jewish and partly Jewish persons made life virtually impossible for the family. Through the generous efforts of friends and relatives in Britain, his father was able to emigrate to Britain and thus escaped imminent arrest and deportation. The rest of the family followed separately.

STUDY AND WORK AT LONDON UNIVERSITY

Walter arrived in London with his suitcase and his cello. He was determined to follow a scientific career and, as a first step, he attended evening classes to work for the entrance examination of the University of London. He attained his goal, but when war broke out all members of the family were temporarily interned on the Isle of Man. Like many refugees of his generation, Walter joined the Pioneer Corps in 1940 and later transferred to the Royal Artillery. His military career was not particularly distinguished and he was demobilized in 1946 with the rank of bombardier. He returned to London with a modest further education grant. His father had died during the war, and Walter lived with his mother and sister in a small flat near Finsbury Park. In view of the financial difficulties, he felt it was essential to obtain a professional qualification as soon as possible. He enrolled for an external London University degree and joined a small class of physics honours students at the Regent Street Polytechnic. The excellent lectures and often personal tuition enabled him, after just one year of concentrated study, to obtain an external 2.1 special degree in physics.

In 1947 he was accepted by Professor J. D. Bernal FRS of Birkbeck College, University of London, to work for a PhD degree in the newly established Crystallography Research Laboratory, housed in a bomb-damaged building in Torrington Square. His supervisor was Werner Ehrenberg, an outstanding and ingenious solid state physicist who later succeeded Bernal to the chair at Birkbeck College. It was a challenging time in Walter's career. He joined four enthusiastic colleagues, all attempting to assemble and construct basic apparatus for their research, largely from former Ministry of Defence stock and from captured German equipment. His project was in the field of electron-optics, aimed at investigating and developing a compact electrostatic focusing system to produce an intense, fine electron focus. If successful, the idea was to incorporate such a system in a highly loaded, fine-focus X-ray tube, which could open up new possibilities in crystallographic studies on complex organic molecules and virus structures. It took many months to build and set up a demountable pulsed electron gun and a vacuum system, incorporating a home-made diffusion pump. Research students were

expected to construct most of their own equipment; in this way they learned many workshop skills, a valuable experience for their later careers.

By 1949 a small but very effective electron-optical system had been developed and optimized, which produced an electron focus $40\text{ }\mu\text{m}$ in diameter at a remarkable loading of 11 kW mm^{-2} . Walter designed an elegant demountable all-metal X-ray generator incorporating the system and built two prototypes in the laboratory workshop for assessment (1)*. The tube had an outer diameter of 3.2 cm, so that the intense X-ray source could be approached within 1.6 cm from the outside. A small-diameter clip-on X-ray camera was constructed, which led to a remarkable decrease in exposure times without a loss of resolution.

The development played an interesting role in the discovery of the DNA structure. In 1950 Bernal gave one of Walter's tubes to Maurice Wilkins (FRS 1959) of King's College, London, for work on the structure of DNA. After designing a suitable camera, Rosalind Franklin and Ray Gosling discovered the A→B transformation in DNA; further, diffraction pictures from single DNA fibres were obtained that provided important experimental evidence for the eventual interpretation of the double helix.

It was a curious turn of events that Herbert Wilson, one of Wilkins's collaborators, brought that particular fine-focus tube to Dundee University in the early 1960s, when he was appointed to a lectureship there. Eventually he returned it to Walter, who, after retirement, restored it to its original form and presented it to the Science Museum in London, where it has been shown in the Bioscience section among the DNA exhibits.

Walter was awarded his PhD in 1950. He obtained a college fellowship and supplemented his income by part-time lecturing and laboratory demonstrating at Birkbeck College and the Regent Street Polytechnic. The old-established scientific firm Hilger & Watts bought the patents to the Ehrenberg–Spear fine-focus tube and Walter became a consultant, assisting in the commercial development of the new micro-focus X-ray system. His scientific interests now began to move towards solid state and electron physics and he continued some of the earlier work of the laboratory on electron-bombardment-induced conductivity in dielectric films.

In 1952 he married Hilda King, who at that time was doing postgraduate work in English literature at Birkbeck College. It was a happy marriage. Both enjoyed academic life and they had many common interests and complementary gifts. Hilda followed an active academic career until her retirement from the University of Dundee as Senior Lecturer in English.

THE LEICESTER YEARS

In 1953 Walter was appointed to a lectureship in physics at the then University College of Leicester, which became an independent university a few years later. The young couple settled in Oadby, south of Leicester. It was a friendly physics department, run in a relaxed manner by Professor E. A. Stewardson. The new lecturer was asked to take charge of a 'home-made' 1 MeV Van de Graaff generator, constructed by a former member of staff for experimental work on electron scattering from thin metal foils. Walter struggled with the numerous mechanical, electronic and vacuum problems, attempting to improve the stability and safety of the machine. After about a year, however, he abandoned this 'white elephant' and managed to

* Numbers in this form refer to the bibliography at the end of the text.

gather enough spare equipment to continue the work he had started in London after his PhD.

In 1955 he published a paper dealing with the effects of electron bombardment (up to 50 keV) on thin dielectric layers fitted with evaporated metal electrodes on opposite sides. In addition to the transmission characteristics of the primary electrons, he studied the distribution of secondary electrons within the specimen. The work established that with both top and bottom electrodes at ground potential, equal but opposite secondary electron currents begin to flow to these electrodes when the primary beam has penetrated half the specimen thickness. This effect, which seems to be a general feature of dielectrics under electron bombardment, arises from the symmetry of the internal space charge field.

Probably the most interesting results at this stage were obtained from experiments on thin amorphous (or vitreous) selenium films, which are deposited by vacuum evaporation as glossy, dark red layers. Walter recognized that this material represented an electronic system with two mobile carriers and remarkably long lifetimes of excess electrons and holes. This gave him the idea of extending the investigation into the time-resolved domain, an approach that proved to be extremely fruitful for much of his subsequent work on transport properties. In retrospect, amorphous selenium (a-Se) was a fortunate choice on which to begin this development. Small specimens, about 5 μm thick, were fitted with overlapping top and bottom gold electrodes. Fast-rising electron beam pulses, from 10 ns to 300 ns in duration, were produced by a delay line technique. At an energy of about 8 keV the electrons passed through the top electrode and generated secondary electron–hole pairs close to it. Shortly before the arrival of the excitation pulse, a synchronized electric field pulse was applied across the electrodes and, depending on its polarity, either generated electrons or holes were drawn across the specimen. The transit times were measured on a sensitive wide-band oscilloscope by a charge integration technique and determined from the linear rising edge of the displacement signal during transit. Typical transit times were in the microsecond range, much shorter than the dielectric relaxation time of the highly insulating material; in this respect the approach differed fundamentally from the well-known Shockley–Haynes experiment in crystalline semiconductors.

Walter published the first paper on this work in 1957 (2). Room-temperature drift mobilities in a-Se ($0.15\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ for holes and $5 \times 10^{-3}\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ for electrons) were extremely low in comparison with crystalline semiconductor mobilities. The interesting feature of the results, however, was the temperature dependence of the drift mobilities, which showed a strictly activated behaviour, with the mobility increasing with rising temperature. This excluded the traditional interpretation of carrier interaction with the thermal vibrations of the structure. Following the ideas of Albert Rose, Walter interpreted the results in terms of a multi-trapping transport: the excess carriers drift in the extended states at the band edges but interact with a reasonably well-defined level of shallow localized states through trapping and rapid thermal release back into the extended states. The latter process is the rate-limiting step, leading to the activated temperature dependence.

This interpretation, which proved to be essentially correct, was received with considerable doubt at the time. Walter recalled a visit by Sir Nevill Mott FRS in 1960 during which he was able to convince him of the elegant directness of the drift mobility approach to transport studies and the interpretation of the temperature dependence.

Important supporting information came from experiments on monoclinic selenium crystals (4), a structure of Se_8 rings. Walter grew the small platelet crystals by a somewhat hazardous procedure in a fume cupboard. They showed a room-temperature electron mobility of $2\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$, considerably larger than for the amorphous form. Below room temperature the

results led to the previously observed activated behaviour but, with increasing temperature, the mobility curve turned over into a $T^{-3/2}$ dependence, expected from the interaction of the conduction-band electrons with the lattice vibrations, which now had become the determining process.

Walter had met Professor Karl Lark-Horovitz of Purdue University, Lafayette, Indiana, who invited him to spend a year in his department as a visiting professor. Lark-Horovitz, one of the pioneers of crystalline semiconductor research, had made the Purdue Physics Department into an active centre for this work. In 1957 Hilda and Walter left for the USA; it was an interesting and enjoyable year and led to many lasting friendships. Hilda worked as an assistant in the English Department and Walter joined a lively group studying the effects of high-energy electron irradiation on n-type and p-type germanium crystals. He began to investigate the creation of surface states by low-temperature irradiation using field-effect photoconductivity and Hall-effect techniques. The new research gave him valuable experience of semiconductor physics and also brought him into contact with leading figures in the field. The work progressed well and at the end of the year Walter published a paper on it in *Physical Review* (3). Before returning home, the couple set out on an unforgettable three-month trip by car and tent through the west of the USA, California and Canada.

Back at Leicester University, with new ideas, Walter began to establish a research group specifically for the study of low-mobility solids, both amorphous and crystalline. Although worldwide research interest at the time was focused on crystalline semiconductors, Ge and Si, he felt that the transport, optical and related properties of the large group of low-mobility and wide-energy-gap materials were largely unexplored and that a concentrated research effort in this field would be fruitful. During the early 1960s he succeeded in obtaining more research space in the department and also attracted an increasing number of capable research students. His carefully planned solid state lecture courses and tutorials to undergraduate physics students made him a popular choice as a PhD supervisor. Support for his research came from various sources. American Army contracts for the study of recombination and space-charge-limited current flow in a-Se films allowed him to buy the latest Tektronix wideband oscilloscope, extending transit time measurements into the nanosecond range. A spectrometer and other optical equipment came from a Science Research Council grant. Throughout this period, the Xerox Corporation in Rochester, New York, was greatly interested in Walter's work and gave him generous financial support for research projects of common interest and for student grants. Amorphous selenium had become the preferred photoreceptor in the first generation of xerographic photocopiers and Walter was a frequent visitor and consultant at the Xerox Research Laboratories in Webster, near Rochester. He also became a consultant to the EMI laboratories in Hayes, Middlesex, and was involved in the development of the Vidicon image tube.

During the 1960s Walter embarked on several successful new research projects, which enhanced the reputation of his Leicester research group. The first of these concerned the transport and related optical behaviour of CdS crystals. These are highly sensitive photoconductors, and small platelet crystals of the material were grown from the vapour phase in several British industrial laboratories. Joe Mort, one of Walter's first research students, who later became a Senior Research Manager at the Xerox Corporation, conducted interesting work on the electron and hole transport in these crystals, which was subsequently related to the controversial mechanism of the optical band edge emission excited by ultraviolet irradiation at low temperatures. The discovery of ultrasonic amplification in CdS crystals stimulated considerable

interest in the interaction of the drifting carriers with the piezoelectric modes of lattice vibration. With the fast time-resolved techniques available in the group, Peter Le Comber (FRS 1992) conducted a PhD project on the transient interaction of the generated excess electrons (or holes) with the acoustic phonons in CdS and ZnS crystals along different piezoelectrically active axes. The results (5) were intriguing: a plot of the drift velocity v against the applied field E gave initially the expected linear rise but at a well-defined critical value of v a sharp decrease in dv/dE to about half its original value was observed. It was shown that the discontinuity always occurred when the drift velocity of the carrier equalled the velocity of sound along the particular piezoelectric axis of the crystal. The phenomenon is therefore associated with the interaction of the piezoelectric modes of lattice vibration with the carriers, strongly enhanced when both travel across the crystal with the same velocity. This strong interaction developed within a surprisingly short time, less than 20 ns, far shorter than could be explained on the basis of current theories. In a subsequent research project the crystal was mounted on a silica buffer coupled to a quartz transducer, which made it possible to study directly the stress wave generated in the CdS crystal as the critical electron velocity was approached.

The second major project initiated in the Leicester laboratory was the detailed study of orthorhombic sulphur crystals, a structure of puckered S_8 rings. When grown from solution under carefully controlled conditions, large double pyramids with sharp edges or thin platelet crystals could be obtained. The present author was the first PhD student to embark on this research, later joined by David Gibbons from the EMI Laboratories. They found that the mobile carriers generated by fast electron or light pulses in those highly insulating crystals were characteristic of two fundamentally different transport mechanisms (6). Excess holes possessed a room-temperature mobility of $10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ controlled by lattice interactions, but at lower temperature this reverted to a multi-trapping transport, a behaviour pattern similar to that mentioned above for Se_8 . In contrast, excess electrons drifted with a room-temperature mobility of $6 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, showing a strictly activated temperature dependence and surprisingly long lifetimes. All experimental evidence pointed towards an intermolecular electron-hopping mechanism between the weakly bonded array of S_8 rings (7, 8). On the covalently bonded rings themselves the electron is temporarily localized through strong interaction with vibrational modes of the ring structure, forming what Holstein called a 'small polaron'. The electron results were fitted to Holstein's small-polaron transport theory with encouraging results; in fact, orthorhombic sulphur is now regarded as one of the best examples of this transport mechanism. Attempts were made by the group to obtain further relevant information on the localization mechanism by a molecular orbital analysis of the S_8 molecule and a study of the vacuum ultraviolet spectrum of S_8 . A high-pressure system was set up by Fred Dolezalek, who had joined the group from Germany. He found a remarkable increase of the electron mobility in S_8 with pressure, mainly associated with the critical pressure-dependence of the overlap between polaron states on neighbouring molecules.

An aspect of solid state physics that Walter found particularly interesting was the fundamental relation between transport and band structure. He thought that simple solids such as the rare-gas crystals Ar, Kr and Xe should be ideal model materials for an investigation of this topic. Their well-established band structure showed a wide conduction band with almost spherical energy surfaces and a narrow hole band, the two separated by a very large direct band gap (for example 13 eV in solid Ar). Drift mobility experiments on this group of solids presented serious experimental problems, which were overcome by the patient and systematic efforts of two PhD students, Sid Miller and Stephen Howe. The gas under investigation was

condensed into a small Melinex chamber at a temperature close to the triple point (for example 82 K for Ar); with careful temperature control a fairly perfect transparent platelet crystal could be grown. The bottom electrode of the chamber consisted of a Mylar-backed gold film, thin enough to allow transient electron-beam excitation.

The results and their interpretation gave valuable information on the transport in simple wide-bandgap solids (9). At low applied fields the measured drift velocity in the rare-gas solids is proportional to the applied field. As expected, electron mobilities are high (for example $1000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for Ar at the triple point, rising to $4500 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for Xe) and their temperature dependence is controlled by acoustic-mode scattering. In all cases the electron mobility in the liquid phase near the triple points dropped to about half its value in the solid, which was consistent with the predictions of the theory of acoustic scattering. A remarkable feature of the results was the dependence of the drift velocity v on the applied field E . As the latter was increased, the linear change of v with E changed so that v varied as \sqrt{E} , associated with the ‘heating-up’ of the electron distribution. As envisaged by the Shockley theory, electrons gain energy from the applied field at a rate faster than can be dissipated by the relatively weak acoustic scattering. The most striking result, however, was the complete saturation of drift velocity at the highest applied fields, which was particularly brought out by the Xe curves. Spear and Le Comber showed by model calculations that the observed velocity saturation could be explained by taking account of the band structure and the increasing effective mass associated with the ‘heated’ electron distribution.

The years at Leicester were a happy and fruitful period in Walter’s life. From a modest beginning he had built up a flourishing research group, and his work on the physics of low-mobility solids was giving him an international reputation. When the Leicester Physics Department moved into its new building, he was able to further expand the range and size of his solid state group. He had close relations with undergraduate and postgraduate students, and Hilda’s student parties contributed to the life of the department. When the Spears moved to a larger house in Oadby, the Senior Honours class took the afternoon off to help with transporting the furniture. During the early 1960s their daughters, Gillian and Kathryn, were born (figure 1). Once the children had started nursery school, Hilda returned to a part-time lectureship at the university.

When Walter received a DSc degree from London University he was promoted to a readership, but he felt that after 14 years at Leicester and with his growing academic experience, it was time to make a fresh start elsewhere. In 1968 he was appointed to the Harris Chair of Physics at the University of Dundee in Scotland.

MOVE TO THE UNIVERSITY OF DUNDEE

The Spear family left their many friends in Leicester in September 1968 and moved to Dundee, on the east coast of Scotland. It was quite a new experience in comparison with life in the English Midlands. The town, with its mixture of tenements and large stone-built houses, is situated on the rising ground above the River Tay, with superb views to the south over the valley, estuary and Fife hills, and to the north over the Sidlaw mountains. After a year in university accommodation, the Spear family found an attractive and spacious house with a beautiful view over the river. They enjoyed walking and touring in the surrounding countryside and eventually bought an eighteenth-century forester’s cottage at the edge of the



Figure 1. Gillian, Hilda, Kathryn and Walter at home in Leicester in 1964. (Photograph taken by Edmund J. King.)

Highlands, within easy driving distance of Dundee. The building had been ‘condemned’ by Perthshire County Council, and Walter and Hilda undertook the lengthy task of improvement and restoration. After a decade the cottage became a comfortable weekend and holiday home, ideal for relaxation and a welcome change from town life.

Queen’s College, Dundee, had been established towards the end of the nineteenth century by some of the well-to-do citizens. It flourished and eventually joined forces with the ancient University of St Andrews, 15 miles south of Dundee, across the Tay, but the relationship was broken in 1967 when Dundee University received its own charter. Jack Standley and Walter were the two new professors appointed to the Carnegie Laboratory of Physics, which had been opened by J. J. Thomson FRS in the early days of the century. The department offered a traditional, broadly based Scottish four-year honours degree in physics and a three-year general degree as well as one-year courses to students from the biological and medical departments. During the 1970s, Walter and Standley succeeded in introducing a two-part degree structure in which the Senior Honours year was devoted to several advanced courses in physics and mathematical physics, selected by the students. The original research activities of the department were in the field of X-ray physics and crystallography, to which Standley had added a rapidly developing microwave group. Work on electron microscopy was also just starting.

Several PhD students and research fellows from the Leicester group had moved north with Walter. Peter Le Comber, who after completing his PhD at Leicester had spent two years in the USA, was appointed to a lectureship at Dundee University. During the next two decades



Figure 2. From left to right: Stewart Kinmond, David Jones, David Anderson, Walter and Peter Le Comber in the 'Old Jute Shed', Dundee University.

until his untimely death he was Walter's close friend, and his collaboration and outstanding scientific work contributed greatly to the development of the Dundee laboratory.

On his appointment, Walter was offered a research area consisting of an old, very solid, stone-built edifice, known as the 'Old Jute Shed' in the Physics quadrangle (figure 2). Dating from the beginning of the nineteenth century, it had indeed been used as a jute storage place and was left on site when the university was built. The interior was an empty shell, but with a generous university grant Walter and Le Comber were able to design the conversion to a research laboratory that was particularly suitable for their planned solid state research. The top floor comprised a large specimen preparation area (later upgraded into a clean room), studies for staff and research students, and a small chemistry laboratory, eventually to become a photolithography facility. The lower floor contained six well-equipped research rooms for optical and transport experiments as well as a small research workshop. With the agreement of Leicester University most of the major equipment of the group was transferred to Dundee, and within six months the optical and low-temperature facilities were ready in the new laboratory. Le Comber, together with Ron Loveland, continued the work on transport in low-temperature solids. They studied hole mobility in the narrow bands of the rare-gas solids and extended the research to solid N_2 , O_2 and CO . The interpretation of these results suggested that small-polaron localization is the dominant mechanism in most of these solids.

EARLY WORK ON AMORPHOUS SEMICONDUCTORS

During the 1960s Walter had been in close contact with Nevill Mott, mainly about their common interest in the physics of the non-crystalline state (figure 3). He was influenced and greatly stimulated by the new concepts and electronic models developed by Mott, and was convinced that they represented a significant step towards a deeper understanding of the electronic properties of disordered materials. Walter and Le Comber decided that one of the main research aims of the new Dundee laboratory should be to provide meaningful experimental information on non-crystalline materials, as experimental tests of Mott's concepts. The main problem was to find a suitable model material. They thought at the time that the study of amorphous Si and Ge (a-Si and a-Ge) would be informative through comparison with their crystalline counterparts, and they began to look at the electrical and optical properties of thin-film a-Si prepared by thermal evaporation and r.f. sputtering. Such specimens were fairly conductive and showed little temperature dependence of their electrical conductivity. It soon became clear that the properties of these a-Si films were almost entirely determined by structural defects, which obscured the phenomena associated with structural disorder that were relevant to Mott's work.

The situation proved to be far more promising in a-Si specimens deposited from silane gas in a radiofrequency glow discharge plasma, an approach pioneered by Sterling and his collaborators at STL in Harlow, Essex. Early in 1970 Walter and Le Comber set up the first plasma deposition unit in the new preparation laboratory. They were joined by Stewart Kinmond, who had just been appointed as senior technician to the group. During the next 20 years Kinmond became an undisputed master in this deposition technique, which, in view of the dangerous gases employed, required very cautious experimentation. In its simplest form the plasma reactor consisted of a vertical quartz tube enclosing a stainless steel substrate holder kept at about 270 °C. A carefully controlled flow of silane gas passed along the substrates, and a glow discharge plasma was produced by means of an inductively coupled 13.6 MHz generator, which had been borrowed from the Royal Radar Establishment. Electron collisions with the silane molecules created a weak plasma of Si/H fragments, hydrogen and electrons, all of which interacted with the growing film on the glass substrates.

The temperature dependence of conductivity of the thin-film specimens (about 1 µm thick) showed a well-defined activation energy of about half the optical gap, which indicated carrier transport in the region of the band edges. In comparison with the previous specimens, the structural defect density was orders of magnitude less. The main reason for this, established in subsequent years, was the presence of hydrogen in the plasma, which very effectively saturates dangling bond defects during growth. The films contained 5–10 at.% of hydrogen, bonded in the random Si network; the Si–H bonds are stable up to about 360 °C, which is generally sufficient for fundamental work and most applications of the material.

The Dundee workers realized at this early stage that a-Si prepared from the glow discharge plasma had considerable potential in the study of the basic electronic properties of disordered semiconductors. This was particularly borne out by the first measurements of electron drift mobility on a-Si in 1970 (10). Above about 200 K, the electron drift mobility showed a small activation energy that was associated with the interaction of the electrons, drifting in extended states at the band edge, with localized band tail states extending to about 0.18 eV below the mobility edge. At temperatures below 200 K the decrease in the observed activation energy



Figure 3. Walter worked closely with Sir Nevill Mott (second from the left on the back row).
(Photograph taken at Rensselaer College, Troy, New York, in 1970.)

suggested that the predominant transport path had dropped into the band tail states, where electrons propagated by a phonon-assisted hopping mechanism. Although this interpretation of the results was not generally accepted at the time, subsequent work proved it to be essentially correct.

The particular interest of the transport results was that they provided some experimental basis for new concepts such as the localized band tail states and the mobility edge, which Mott regarded as fundamental to an understanding of the disordered state.

The work during this period showed clearly that the density and distribution of localized states in the forbidden gap of an amorphous semiconductor is of crucial importance in determining its properties. In attempting to obtain some experimental information of this aspect, Walter used the field-effect method, which he had previously employed in surface studies on crystalline Ge. In this approach an electric field is applied normal to the thin-film surface and the conductivity along the specimen is measured as the perpendicular field is increased in a step-by-step process. Each step causes a change in the electron distribution in the localized states, which can be calculated with respect to the Fermi energy from the corresponding measured conductivity change. The first state distribution graphs for a-Si published in 1972 showed a deep minimum near the centre of the gap, where in the best plasma-deposited a-Si

the density of states was as low as $10^{15} \text{ cm}^{-3} \text{ eV}^{-1}$. Evaporated a-Si specimens led to state densities three to four orders of magnitude higher. The rapid rise in the curve towards the electron mobility edge suggested a width of the tail state distribution of about 0.2 eV, which was not inconsistent with the results of the transport experiments.

Although the first field-effect studies gave an indication of the state distribution in the gap of a-Si, the experimental techniques, specimen design and interpretation of the results were greatly improved in the following few years by Le Comber and his research student Arun Madan; their work also laid the foundations for the subsequent development of the a-Si field-effect transistor.

Walter had been elected to the Fellowship of the Royal Society of Edinburgh and, with the growing interest in non-crystalline materials, was asked to organize the 1972 Scottish Universities Summer School in Physics. The School, on the topic 'Electronic and structural properties of amorphous semiconductors', was held in Aberdeen and extended over three weeks. Many of the leading European and American workers in the field participated and gave detailed lectures on their work and ideas.

THE 1970S: DOPING IN THE AMORPHOUS PHASE

The 1970s saw many encouraging developments in the Dundee laboratory. Several additional members of the academic staff had joined the research group and, with growing support from the Science and Engineering Research Council (SERC) and industry, the number of research students, postdoctoral fellows and technicians had increased. The 'Old Jute Shed' was now an active and lively centre for amorphous semiconductor research, its success largely based on close and well-planned collaboration. Throughout this period considerable efforts went into the development of the plasma deposition technique. Several experimental reactors were constructed to explore the effects of geometry, frequency (eventually up to microwave frequencies), plasma intensity and other parameters on the specimen properties.

Perhaps the most important breakthrough in the field was achieved in 1975 (11) when, contrary to prevailing opinion, the Dundee group was able to demonstrate that plasma-deposited a-Si (and a-Ge) could be doped very effectively from the gas phase during deposition (figure 4). As described in detail in the *Philosophical Magazine* paper by Spear and Le Comber (12), the addition of small but accurately measured volumes of the gaseous hydrides phosphine (PH_3) or diborane (B_2H_6) to the silane flowing through the reactor could control specimen conductivities over 12 orders of magnitude. It was therefore possible to produce n-type and p-type a-Si continuously by successively flowing pre-prepared gas mixtures through the reactor. The possibility of substitutional doping in a-Si has led to an upsurge of applied interest in many laboratories throughout the world. Measurements on the first a-Si p-n junction (13) were published by the Dundee team; somewhat later, members of the RCA Laboratory reported results on the $\text{p}^+ \text{--i--n}^+$ junction (an undoped a-Si specimen with highly doped p and n surface layers), which became the basic configuration used in the extensive developments on a-Si solar cells.

During the 1970s, Walter and his colleagues began to extend the range of their experimental work to improve the fundamental understanding of this promising thin-film material. An important approach in this connection was through the photoconductivity of a-Si. Detailed studies of its spectral intensity and temperature dependence gave valuable information on

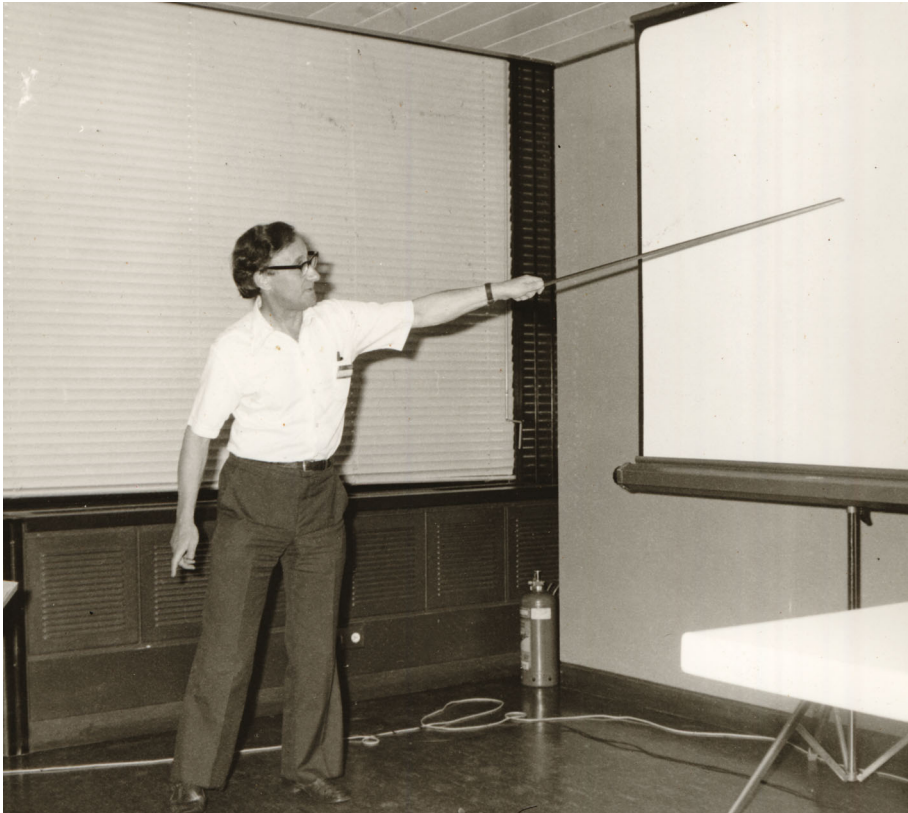


Figure 4. Walter presenting his results on the doping of amorphous silicon at the International Conference on the Physics of Semiconductors in 1976. (Photograph by Foto Sharaglia.)

the recombination kinetics of the generated excess electrons and holes and also on the localized states within the gap involved in the recombination process. Walter and David Anderson showed that by slight n-type doping the Fermi energy could be moved above a predominant range of recombination centres, which greatly sensitized the photo-response to approach unit quantum efficiency (15).

David Jones, one of Walter's colleagues, started work on thermoelectric power in the amorphous semiconductors a-Si and a-Ge. It is a difficult but highly sensitive experimental approach that provides direct information on the movement of the Fermi energy of the system with temperature, doping and deposition conditions. Hydrogen thermal effusion experiments, giving a quantitative measure of the hydrogen incorporated into the structure, gave useful information on the stability of the material and its electronic properties as a function of hydrogen content. Tony Snell, who had joined the group from the electronics industry, initiated an experimental study of the metal/a-Si barrier and n-p junction, attempting to interpret capacity and conductance data in terms of theoretical models.

In 1977 Le Comber and David Jones carried out an investigation of the Hall effect in n-type and p-type a-Si (14). They discovered the curious double reversal in the sign of the Hall coefficient—negative for p-type samples and positive for n-type specimens, opposite to that

expected from the classical theory and found in crystalline semiconductors. Walter recalled the excitement caused by the initial experimental results, followed by doubts about the correct direction of current flow and magnetic field in the experiments. The theoretical work of Friedman, based on small-polaron theory, had shown that the interpretation of the Hall effect in a solid lacking long-range order is fundamentally different from that in the crystalline material. He predicted a single sign reversal (the p–n anomaly), but the satisfactory explanation of the observed double reversal still poses theoretical problems.

At this time Walter began a fruitful collaboration with the Ion Implantation Group at the Max Planck Institute in Heidelberg, led by Siegfried Kalbitzer. The work showed that efficient doping of a-Si can also be achieved by implanting ions of Group III and V elements (16) and that interstitial implantation of alkali ions provided an effective method of n-type doping. The collaboration continued for a further five years, with support from the European Economic Community, on a programme to optimize implantation techniques. Another successful collaboration, extending over several years, was with Ifor Austin and his colleagues at Sheffield University. They investigated the photoluminescence of a-Si layers deposited in Dundee under different plasma conditions as well as the electroluminescence in a-Si junctions. The work was subsequently extended to plasma-deposited silicon nitride and silicon carbide.

In the mid-1970s Walter, with Alan Owen of the University of Edinburgh, launched a one-year MSc course on the physics and technology of amorphous semiconductors. It was a fairly advanced course, supported by the SERC, which attracted students from Britain, Germany and the Middle East and proved to be an excellent preparation for PhD work. Nevill Mott was a frequent visitor to the laboratory for lectures to the MSc students and for extensive discussions about research projects. His was a very stimulating influence on the progress of the research.

Although Walter always worked on some of his own research projects, this became increasingly difficult with growing departmental administration and external commitments. He gave lecture courses to honours and MSc students and was frequently invited to talk at meetings and conferences. The headship of the Physics Department was on a rotating basis, but with highly cooperative professorial colleagues the administrative burden was shared and became less arduous. Most of Walter's scientific effort went into the running of the growing research group, the supervision of research students and the collaboration with postdoctoral staff. Le Comber played an important supporting role in the development.

During the late 1970s Walter's achievements received wider recognition. In the same year he was awarded both the Max Born Medal by the Institute of Physics and the German Physical Society and the Europhysics Prize. Some time later the Royal Society of Edinburgh presented him with the Makdougall Brisbane Medal for his work in the field of amorphous semiconductors. The highlight of this period, however, was his election to the Fellowship of the Royal Society in 1980, which gave him great pleasure and encouragement.

RESEARCH DURING THE 1980S AND APPLIED DEVELOPMENTS

The work of the Dundee Laboratory during the 1980s continued on the fundamental side but also turned increasingly towards device applications of amorphous materials, especially a-Si. Walter was keen to round off his work on the basic understanding of a-Si as a model amorphous semiconductor by new and improved experimental approaches. A significant simplifi-

cation in the transport studies was the use of pulsed laser photoexcitation of excess carriers, rather than the more cumbersome electron-beam excitation. The rapid developments in the doping technology made it possible to produce good a-Si specimens with thin, highly doped n-type and p-type surface junctions on opposite sides of the undoped central region. In reverse bias these devices reduced leakage currents, so that mobility measurements could be extended into the high-temperature region. The transition from the multi-trapping to the scattering regimes could now be observed in a-Si (as in the early work on Se_8 and S_8) and led to extended state electron and hole mobilities of about $10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and $1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively.

On the low-temperature side, transport measurements were previously limited to the range above 150 K, when the hopping mobility at the bottom of the tail states became too low for a coherent transit signal. However, Carolyn Cloude and Walter (21) discovered well-defined electron transits at temperatures below 80 K, corresponding to surprisingly high mobility values. The latter depended on the density of the photogenerated electrons and could be further increased by synchronized pre-excitation. It was concluded that, in this temperature regime, states in the rapidly rising tail state distribution could be filled during or before the transit, so that the electron hopping path was moved to higher-energy regions of the distribution where hopping distances were greatly reduced, leading to the remarkably high hopping mobilities.

In collaboration with David Goldie, Walter conducted a detailed investigation of transport in compensated a-Si, which contains an approximately equal density of donor and acceptor sites, leaving the Fermi energy close to the centre of the gap, as in undoped specimens. The results showed that the electron mobility is at first reduced by the potential fluctuations introduced by the donors but then, with increasing donor density, changes markedly to a phonon-assisted hopping transport between donor sites.

Work by colleagues at the Xerox Laboratories and at Marburg University had led to the conclusion that the small fraction of dangling Si bonds, left unsaturated by hydrogen, represented the predominant defect in a-Si. These sites give rise to three electronic states, depending on the position of the Fermi level of the system. The neutral site, D^0 , contains a single electron but can accept a second electron (D^- site) or lose its electron to form D^+ . The D centre is thus a very likely recombination site, although its energetic position in the gap of a-Si was still a controversial problem. Walter felt that reliable electron and hole lifetime measurements as a function of doping in n-type and p-type specimens should provide crucial information on the recombination process. Together with his PhD student Hugo Steemers, he developed a time-resolved delayed-field technique that proved to be a direct and reliable method for studying the lifetime of each type of carrier with respect to capture in a dangling bond state. The results showed convincingly the expected sensitization of the majority carrier lifetimes and the desensitization of the minority carrier lifetimes as the Fermi level is moved towards the respective band edge (20). The analysis places the D^0 state about 1 eV below the electron band edge. Work on the temperature dependence of the D-centre capture cross-sections led to further interesting results.

Several Japanese groups had shown that by modifying the plasma preparation conditions normally used for a-Si, a material containing crystallites with dimensions in the nanometre range could be deposited. Gerhard Willeke and Walter investigated the electronic properties of this nanocrystalline material as a function of crystallite size and structure deduced from electron diffraction data (19). It was shown that with increasing crystallite size the width of the localized tail state distribution shrinks and, eventually, all these states, typical of the

amorphous phase, disappear. It was also observed that, with increasing long-range order, the anomalous sign of the Hall effect reverts to that predicted by classical theory at crystal-lite sizes of 2–3 nm. Other projects at the Dundee laboratory during that period included an investigation of the effect of applied and internal strain on the electronic properties of doped and undoped a-Si and work on preparation and properties of plasma-deposited a-SiC_x and a-SiN_x.

On the applied side, doping in the amorphous phase had opened up exciting new possibilities for plasma-deposited device structures. Thin-film a-Si layers composed of successive n, p and i sections as well as thin SiN_x or SiC_x insulating regions could be produced in a continuous deposition process on a range of substrate materials. There was no fundamental limitation to the size of deposited films—an attractive feature for large-area applications. As a leading laboratory in the field, the Dundee group was approached by an increasing number of British and European industrial laboratories for help and collaboration on new device ideas.

Although the financial rewards were beneficial to the development of the laboratory, Walter had serious reservations about the strict confidentiality conditions that several of the industrial collaborators tried to impose on his work. He felt that research results at a publicly funded institution should be made available to the scientific community through publication and discussion. However, he was also keen to explore possible applications of amorphous semiconductors in the Dundee laboratory and, if successful, pass them on to an industrial laboratory for development. In this connection he acted as consultant to several Swiss industrial concerns.

Two very promising developments were pioneered by Walter, Le Comber and Snell during the 1980s. The first was the a-Si field-effect transistor (FET), which nowadays is widely used as the switching element in large-area liquid crystal colour displays. Cyril Hilsum FRS, then a senior physicist at the Royal Signals and Radar Establishment (RSRE), originally drew their attention to this possibility. On the basis of the specimen configurations used in the earlier field-effect experiments, the Dundee laboratory produced the first a-Si FETs and reported their characteristics in 1979. The next step was the design and production of an array of minute FETs for application as addressable control elements in LCDs. In collaboration with Tony Hughes of the RSRE, the work progressed well and the Dundee laboratory produced 500-element prototype arrays for evaluation (17).

Throughout the 1980s Le Comber made many efforts to stimulate industrial interest in these displays by acting as a consultant to British and European electronics industries. Currently, within the flat-panel displays industry, amorphous silicon has become the bedrock that underpins the whole industry and has facilitated the very creation of the large-area, high-resolution, full-colour displays that we now rely on for so many consumer products. Amorphous silicon (and its slightly more expensive version poly-silicon) is used in LCD laptop computers and desktop monitors, and in LCD televisions. The technology allows high-quality video to be seen in mobile phone displays, and in the small displays used in digital cameras and camcorders. Without amorphous silicon, the explosion in consumer electronics would not have happened at such a rate, because the quality of the display would not have matched the speed and performance of the digital electronics. In terms of value, the flat-panel displays industry now has a global sales value of around US\$100 billion, and products totalling more than 70% of that value use amorphous silicon to underpin their performance. In recognition of their contributions to the field of addressable displays, Walter and Le Comber were among the recipients of the 1988 Rank Prize for Optoelectronics (figure 5).



Figure 5. Walter relaxing with Peter Le Comber at the reception, in the Dorchester Library of the Royal College of Physicians, that followed their receiving the Rank Prize for Optoelectronics in 1988. (Photograph by Mark Dennis.) (Online version in colour.)

Thin-film photovoltaics for the production of electrical power from sunlight is another crucial application that builds on the work of Walter and Le Comber. Although crystalline silicon accounted for about 90% of all solar cell production in 2008, amorphous silicon absorbs solar radiation about 40 times more efficiently than crystalline silicon and hence it can be effective in very thin layers 1–10 μm in thickness. These can be deposited on large-area lightweight substrates (such as stainless steel sheets) for the production of low-cost solar cells. These had

a global market value of the order of US\$1 billion in 2008, a value that is bound to expand swiftly in present circumstances.

The third fascinating device development arose from collaboration with Alan Owen and his colleagues at the University of Edinburgh. It was based on the discovery that, after forming, certain metal/a-Si junctions, such as a Cr-p⁺-n-i-Cr configuration, behaved as an electronically non-volatile memory element. Such a device exists in two states that differ in electrical conductivity by several orders of magnitude, the state remaining unchanged if the supply voltage is removed. The memory state can be changed by small voltage pulses of opposite polarity a few nanoseconds in duration. The initial work indicated that in terms of speed, retention time and stability these thin-film memory elements compared very favourably with the then current crystalline devices used for non-volatile, programmable storage.

The first joint paper on this work by the collaborating groups (18) was awarded the 1981/82 Maxwell Premium of the Institution of Electrical Engineers. Further work, particularly the development of arrays of addressable memory elements, received generous support from British Petroleum (BP) and also led to collaboration with members of the BP Research Laboratory at Sunbury. Subsequently, Le Comber initiated a new joint project with British Telecom, the SERC and the Edinburgh group on the application of the above memory elements in artificial neural networks.

During the 1980s Walter made several trips to Japan, where he was warmly received by industrial and university colleagues; he also visited China. The Royal Society sponsored two tours of Indian universities and scientific institutes, which gave him an opportunity to see a great deal of this fascinating country. In 1988 Walter was invited to present the Royal Society Bakerian Lecture (22). He chose the title ‘amorphous semiconductors: a new generation of electronic materials’ and included numerous demonstrations in his lecture. Two years later he was awarded the Rumford Medal of the Society.

RETIREMENT

Walter retired from the university in 1990 and Le Comber was appointed to the Harris Chair of Physics and took over the running of the Amorphous Semiconductor Laboratory. As Professor Emeritus, Walter stayed to complete research projects and the supervision of his remaining PhD students. He continued to visit the laboratory several times a week, keeping in close touch with how the work was developing and exchanging ideas with Le Comber, who sought to steer the work of the group further towards the applied side and industrial collaboration. Le Comber was an outstanding young scientist and was elected to the Fellowship of the Royal Society in 1992. His tragic death during that year was a considerable loss to the university and to the scientific community and was a great blow to Walter.

Walter then felt that it was time to retire more completely from the university scene and devote himself to other activities. He did not abandon his scientific interests, however, but instead broadened their spectrum, reading widely in journals such as the Royal Society’s *Notes and Records* and the Physical Society’s magazine. He took a considerable interest in the problem of energy needs and believed firmly that the only viable solution lay in nuclear power. He thought that research on nuclear fusion had been neglected by governments for research on less promising energy sources. His final publication was, in fact, a letter to the local paper on 11 March 2006 in which he regretted the lack of government support for fusion and extolled its potential.

During the early 1990s the Spears had decided that it was the time to move from their large stone-built family house into a smaller and more easily managed home. They divided their large garden and built a modern house, which they planned and designed themselves. With his practical mind, Walter took a great deal of interest in the building going up in his back garden. Once the outer shell had been completed, he installed some of the electrical refinements and did the internal carpentry in several rooms, a task that he particularly enjoyed. He lined their joint study with bookshelves to hold their several thousand books and later helped friends to fit out their houses and flats, making bookshelves in the Cathedral for the Bishop of Brechin and doing the same in a friend's church. He used to joke that he had been appointed joiner to the Episcopal Church in Dundee.

Walter also returned to his early love, chamber music, which might have offered him a career had he not chosen to concentrate on science. He was an accomplished and enthusiastic cellist and with regular and intensive practice he regained and improved his technique. Together with the pianist David Robb, a colleague of Hilda's from the English Department, Walter played his way through the sonatas of Bach, Beethoven and Brahms. He venerated them, all three. But he was equally keen to vary the diet with music for cello and piano by other nineteenth-century composers such as Schubert, Schumann and Chopin. He and Robb were not averse to exploring twentieth-century compositions and played sonatas by Shostakovich and Prokofiev, for example. In more recent years, however, along with the violinist Angela Green, they concentrated on the extensive repertoire of piano trios from Mozart onwards.

When Hilda retired in 1993 and they found themselves no longer travelling abroad to conferences, they decided to travel just for pleasure. They became keen Swan Hellenic cruisers, travelling to the Mediterranean, the Middle East, the Black Sea and the Baltic, and their last such trip in 2005 was to the Arctic. In between these journeys they returned to well-loved destinations in Switzerland and Italy, particularly Umbria and Tuscany. Italy became a favourite holiday destination, and Walter attended evening classes to improve his knowledge of Italian.

Walter also took to gardening in a modest way. He had always mown the lawn and cut the hedge but, with time on his hands, he weeded the flower beds and constructed cages for the vegetables, to resist the ravages of the local wood pigeons. He also found much pleasure in reading, particularly in a leisurely approach to the nineteenth-century British novel, and to some German and French literature.

Not long after their marriage Walter and Hilda had acquired an Airedale dog, to be a companion and accompany them on their walks (figure 6). While their daughters were small they had had smaller dogs, but in 1992 the second-generation Airedale was bought; at her death, the third Airedale joined the family, this last one sadly outliving her master. Walter especially enjoyed these last two dogs, as he had more time to relax, to play with them and to help in their upbringing. Similarly, he spent a lot of time with his two grandchildren, taking them on holidays, teaching his grandson to play the cello and encouraging both children to handle hammers, screwdrivers and other tools successfully.

Though he had suffered from angina for several years it was only during the final few months of his life that he felt the need to avoid exertion in cold or windy weather. His death, after only four days in hospital, was totally unexpected and a serious shock to his family. He is also missed by a large number of his former students and collaborators, whose careers and lives benefited from his energy and infectious enthusiasm for scientific research.



Figure 6. Walter after retirement near the eighteenth-century cottage that he renovated close to the Highlands. (Photograph taken by Hilda Spear in the late 1990s.) (Online version in colour.)

Walter's ashes were scattered in the grounds of Dundee Crematorium and he is remembered with the siting of a bench in the Rose Garden of Balgay Park, a few minutes' walk from the Spears' house.

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The frontispiece photograph was taken at Dundee University in the late 1980s. The photographer is unknown. (Online version in colour.)

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