SYDNEY LEACH
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Sydney Leach will be remembered as an outstanding and inspirational scientist, an irreplaceable friend to many—artists and musicians as well as academic colleagues. He encouraged and influenced numerous scientists as a mentor. After graduating from King’s College London and carrying out war work at Farnborough, he spent all his scientific life based in Paris, working principally at Orsay and, in his later years, at the Observatoire de Paris–Meudon. Sydney was a major influence in establishing chemical physics in France after World War II, founding the highly influential Laboratoire de Photophysique Moléculaire (LPPM) at Orsay, where much of his pioneering work was performed. The ‘Sydney lab’ lives on in the newly created Institut des Sciences Moléculaires d’Orsay. Early experiments often took place at the synchrotron source (ACO, Super-ACO), just a few hundred yards from LPPM. He was a pioneering advocate of synchrotron radiation, and a driving force for its use in spectroscopy and photodynamics, along with free-electron lasers, supersonic jets, coincidence spectroscopy and matrix isolation—techniques that were applied and refined over decades and used to explore fundamental processes such as photoionization, vibronic coupling and radiationless transitions. Sydney’s seminal studies of polyatomic molecular ions led him towards fresh horizons in planetary atmospheric and space science. His work opened

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new vistas in cometary spectroscopy, polycyclic aromatic hydrocarbons, fullerenes and their possible roles in the chemistry of the interstellar medium and, finally, biologically relevant species, helping to instigate the newly developing subject of astrobiology—a perfect example of his sustained prescience in the world of science.

EARLY LIFE AND EDUCATION

Born in 1924, Sydney Leach came from a family without privilege or academic background. His subsequent career, founding a major laboratory at Orsay and publishing more than 300 articles, many of these ground-breaking, should be seen against this backdrop. At three years old, personal washing in the kitchen sink was his lot, with a shared toilet on the landing, in his tenement block in London—though his family subsequently moved to a better neighbourhood in 1933. Sydney’s father was a kind and compassionate man who, while preserving his independent spirit, kept deep and affectionate links to his large family and community. He worked at first for a stunt bicycle manufacturer, ran bicycle races and met his future wife through a cycling club. After surviving the trenches in World War I, he joined the family tailoring business and, following Sydney’s mother’s death at the age of 42 when Sydney was 12, he brought up their three young children on his own. This was a feat all the more remarkable given the time he was living in, and most certainly he played a critical role in their education. Sydney’s sisters, Joyce and Sylvia, now living in the USA and in Canada, were nine and five years old when their mother died. Sydney and his father remained very close until his father’s death in 1953, following the 1952 London smog, his lungs weakened by World War I gas attacks. Sydney also felt very affectionately towards his mother, a lady whom he described as adventurous in spirit, outgoing, trusting in her children and an avid reader with a passion for crossword puzzles. These are hints of the source from which Sydney inherited his intellect.

Scattered memories give reminders of the era in which Sydney grew up: the R101 over London in 1929, talk of the Manchurian war in 1932, frightening images of World War I, seeing the glow of the burning Crystal Palace in 1936, the historic performances of world-class cricketers Don Bradman and Frank Woolley at Blackheath in 1934 or, more mundanely, the open upper deck of a tram. There was a larger family; its members included a dizzy aunt who married a Maltese sailor, a relative who emigrated to America and one who served long years in the British Army in the Sudan.

Sydney’s life-long love of music was instilled by his parents from a very early age, with a horn gramophone, of the HMV type, but no radio until 1934—too expensive a luxury. Outside school, he had violin lessons. One may find early hints of his precocious mind: being able to locate specific gramophone records at age three or—shades of things to come half a century later—hoping, a hope frustrated, to build a telescope on the roof of his new home in New Cross, where they moved in 1933. That year marked a turning point. His new teachers in Monson Road School varied from a sadist who caned him every Friday, ‘for no reason at all’, to a real teacher, a Miss Evans, where he was top in English but second in mathematics. Practical science classes during this time made an early impression on Sydney. One such showed how a ball of metal could pass through a ring when cold, but could not when it was hot.

It was between the ages of 10 and 11 that Sydney’s abilities became apparent. He won a top scholarship in the London County Council examinations, as well as a Stanhope Scholarship.
for the Addey and Stanhope School of Science. This was one of only two mixed schools in London, and was accompanied by progressively minded teaching, with high standards in the sciences, including practical labs from the start. These inspired Sydney’s tinkering with a chemistry set at home, with experiments in the back yard. The names of his teachers were etched into his mind, readily recalled many years later, as they introduced him to Mozart and to mathematics, physics and chemistry. There was a school trip to Switzerland in spring 1938, more or less coinciding with the Anschluss, and he felt the threat of war hanging everywhere. Accompanying these formative experiences were less portentous visits to see Flash Gordon at the cinema. He read Olaf Stapeldon’s *The last and first men* and wrote that this science fiction projection into the future, re-read several times, became of service in providing a timeframe for thinking, decades later, of the origin of life on Earth.

During his school years, Sydney was chosen as a violinist to join a children’s orchestra, giving further impetus to the power of music in his life. He must have been good, since he won a scholarship to attend the Royal Academy of Music. These trips to South Kensington led him to the great museums, the first week to the Geological Museum, the next to the Science Museum, from where he went no further but visited every successive Saturday, becoming another brick in the house that made Sydney. Violin lessons thus led down unexpected paths—a reminder of the importance of such public institutions in forming the minds of all of us. A further such institution was the public library, just 50 yards from his New Cross home, where he avidly read adventure, exploration and popular science books. Sydney wrote that much of his pre-war education was linked to this library. Moreover, next door was a second-hand bookshop, a source of freely given ‘unsaleable’ books such as *Canoeing in the Congo*, Conrad in the making. During the wartime years, his high school was evacuated to Wales and Sydney took care of his two sisters, who were housed in a nearby village, writing regular reports to his father, keeping him constantly informed. His sisters recall how he was more than a brother to them, a sentiment that would find later professional parallels among scientists who saw him as more than just a collaborator or laboratory director.

Moving on, Sydney found that it was the physics of chemistry that interested him and, after ‘Higher School Certificate’ (the old A-level), he studied physics at King’s College London. Evacuated to Bristol in 1942–1943, he was greatly inspired by Arthur Tyndall’s lectures and, in his own words, ‘caught more than a glimpse of the pleasure of science’. Sydney’s interest in spectroscopy was ignited by a summer job at Kodak in 1943. Chance and his widening knowledge of literature intervened in 1944 and led to a life-changing encounter and conversation with C. P. Snow. Following a discussion on the poet Richard Aldington, Snow eased Sydney’s passage into Farnborough to work on spectroscopy rather than radar, where all of his other 21 classmates were directed.

Sydney’s first project at Farnborough was to study the spectrum of lead tetraethyl, the anti-knock component of aviation fuel. But the beautiful five-banded electronic spectrum reported by ‘Tommy’ H. W. Thompson in 1934 could not be reproduced! Despairing at his failure, he found that Thompson was visiting Farnborough. Plucking up his courage, Sydney told Thompson of his results, only to be informed by Thompson that ‘there was a benzene impurity in my lead tetraethyl’, which was the source of the five bands. Sydney saw this as an important lesson learnt. Further wartime science at Farnborough schooled Sydney in experimental technique and developed his characteristic self-reliance. Spectroscopy of metals, on spark-plug refractories, piston-ring deposits, etc. all yielded practical results, including pioneering work on a double-beam IR spectrometer and on a German CO detector sent to him.
after the end of the war. He wrote years later that Farnborough taught him that common sense was just as important as physics in solving the wide variety of practical problems that came his way. It also taught him that basic and applied research can be equally satisfying and are two sides of the same coin—but there were perhaps more coins to be had in applied research. More details of the Farnborough work may be found in ‘In my time: scenes of scientific life’ (22).

Sydney’s first encounter with the academic world of science was attendance at the very last wartime Faraday Society discussion meeting, on the ‘Application of infrared spectra to chemical problems’, held in January 1945 at King’s College London. This was important both for the display of advances in experimental techniques and theoretical understanding and for the significant wartime applications in the petroleum and polymer industries. Sydney also recorded that he encountered for the first time the rivalries which beset research, in the form of a ‘heated discussion’ of the relative contributions of Oxford and Cambridge in wartime science.

The final phase of Sydney’s making of a scientist as a young man was what he has called his ‘flight to France’. Determined to see more of the world, and following interviews at the French embassy in London, he departed these shores in 1946, to work with Edmond Bauer and Michel Magat at the Laboratoire de Chimie Physique in Paris. With an offer in the bag from Canada, his career might have taken a very different turn at this point, but it was France that would reap the full benefits of his ensuing insights and work. He came to the laboratory in Paris, which was stripped of the resources to conduct science but peopled with superb researchers. He resorted to the use of what he could recover from Pierre Auger’s pre-war laboratory, building such things as a McCleod gauge in the Dickensian squalor of a tiny glass-blowing workshop in the Rue de la Montagne Sainte-Geneviève. So began Sydney’s scientific journey, in which he is rightly credited with a major role in recreating chemistry, physics and spectroscopy in France after the depredations of war. Indeed, there was little or no spectroscopy in post-war France. Thus Sydney’s foray into this area became a voyage of self-tuition. This led over the years to the creation of study groups in France and then to workshops to develop spectroscopy in France and across Europe. So was born a spontaneous progenitor of the modern European Training Network and it is fair to say that Sydney was indeed a pioneer in this development, so beneficial to many of us in the last decades.

Sydney met his companion for life, Ida, in Paris at this time. Ida had come to do research for her PhD at Oxford. They married in 1952. Ida worked as an editor in Italy and France. As we write, they have two children, Stéphane, a composer and pianist, and Francesca, a teacher and translator, and four grandchildren (figure 1).

PRELUDE: PARIS, 1946–1959

Throughout his ‘scientific life’, Sydney embodied Peter Medawar’s Advice to a young scientist: ‘If you want to make important discoveries, you first have to ask important questions’ (Medawar 1979). Sydney was continually open to new scientific ideas and directions, especially their interconnections. As his encyclopaedic knowledge grew, the boundaries of his curiosity-driven interests steadily expanded until they embraced the (not quite) empty space between the stars. Indeed, the Royal Air Force motto, per ardua ad astra (through adversity
to the stars), runs like a \textit{leitmotiv} through Sydney’s scientific career. Perhaps the first dormant seed was sown during Sydney’s early apprenticeship spell at the Royal Aircraft Establishment. Certainly it sprang to life when, following a few weeks spent in Herman Schüler’s laboratory in the French-occupied zone of post-war Germany, he moved to Paris and began an exploration of molecular emission spectra, recorded in those days on photographic plates.

The spectra were excited in a gas-filled, cooled hollow-cathode discharge tube—the Schüler tube. Among the spectra was that of the so-called $\alpha$-band system of ammonia, which had been detected in the luminescence of comets and which Sydney was able to assign to the NH$_2$ radical. Its subsequent observation in absorption at high resolution by Herzberg and Ramsay, following the flash photolysis of NH$_3$ and its isotopic variants, confirmed the assignment. Other studies identified emission spectra associated with a range of diatomic free radicals, among them S$_2$, OH, SH(D), and NH(D) in both the singlet and triplet manifolds, enabling a first determination of its singlet–triplet energy separation, and lastly, the generic aromatic hydrocarbon, benzene. This last was a portent of many things to come, from the laboratory to the interstellar medium.

The photophysics and photochemistry of benzene and related molecules, in frozen matrices as well as the gas phase, became one of Sydney’s key interests, partly for their own sake but more importantly because they provided an entry into ‘a strongly felt aesthetic component (of molecular spectroscopy) where it would be the diagnostic instrument, rather than the object’ (22). Matrix studies were conducted initially in frozen glassy solutions (77 K) and extended to crystalline organic and frozen rare-gas matrices (20 K), following a year spent in Berkeley (figure 2), where he worked with George Pimentel developing and exploiting the matrix isolation technique—part of Herb Broida’s ‘Free Radical Trapping’ program.
In 1960, Sydney moved with Michel Magat’s group from the Laboratoire de Chimie Physique in Paris, then under the direction of Yvette Cauchois, to Orsay, where he was to become one of the leading pioneers of matrix spectroscopy and photochemistry—the latter, perhaps, an initially serendipitous by-product of the former. Its central themes were isolation of the dispersed molecules, and trapping of their photochemical products in the matrix. Ultraviolet excitation of substituted benzene derivatives in frozen matrices led to the photolytic formation and trapping of organic free radicals, for example, benzyl and its deuterated isotopes, sensitively detected via their luminescence spectra. The similarity of its vibronic band structure to that of the parent molecule, toluene, suggested very similar force fields, which facilitated the assignment of its ground and electronically excited states—a signal achievement at the time (1). Of course, in the condensed phase the aromatic molecules were not isolated from their local environments (or even from themselves when clusters were formed) and early ultraviolet spectroscopic studies revealed a range of interesting ‘conversations’, both physical and chemical, between the photoexcited aromatic molecules and their immediate locale—potentially modifying their properties. The photoexcited solute could act as a ‘spectroscopic probe’ of crystalline matrix environments reporting back on their structures and dynamics (3). In glassy organic matrices, electronic excitation of benzene led to ring opening and reaction with the solvent to form a substituted triene (4).

Typically, while exploring all these avenues, together with Eva Migirdiyan, Ricardo Lopez-Delgado, Claudine Cossart and others, Sydney never lost sight of the central theme (and the aesthetic component)—at this stage, the role of vibronic interactions in molecular electronic spectroscopy, notably Fermi resonance, Herzberg–Teller and Jahn–Teller effects in the photophysics of aromatic molecules and the consequences of lowering their symmetry.
A particularly striking example of the latter was provided by an analysis of the perturbed vibronic structure in the luminescence spectrum of benzene in frozen cyclohexane, which revealed a matrix crystal field-induced distortion of its lowest-lying triplet state (2). More than half a century later, one of the authors (J.P.S.) still recalls a breathtaking ‘David and Goliath’ discussion of this conclusion, between Sydney’s fearless (and compact) colleague, Ricardo Lopez-Delgado, and the intellectually (and physically) towering figure of John van der Waals from Leiden, when this research was presented at a conference in Bordeaux in 1964. Many years later, a vibrational analysis of the fluorescence from C₆F₆⁺ and its symmetrically substituted relative, 1,3,5-C₆F₃H₃⁺, chosen because of the degeneracy of their ground (2E′) electronic states, provided benchmark examples of distortion and consequent symmetry-lowering through the dynamical Jahn–Teller effect (7).

While the earlier work was in progress, Sydney, with Marcel Horani and Joelle Rostas, laid the foundations of his seminal studies of polyatomic molecular ions. They began with the construction of a crossed, low-energy-electron-diffusive molecular beam source, rather than the less selective electrical discharge tube. Like the photoexcited neutrals, the ions were also detected, initially, via their emission spectra—several of them for the first time: these included H₂S⁺ (and SH⁺), CO₂⁺ and its iso-electronic partners, N₂O⁺, COS⁺ and CS₂⁺. The ion CO₂⁺ had already been identified as a component of planetary atmospheres and, as Sydney noted (22), all of them were ‘of direct or potential interest in cometary spectroscopy’—another early pointer to ‘a bigger picture’. Later extensive spectroscopic studies of CO₂⁺, including comparisons with the photoelectron spectrum of the neutral molecule, led back to the role of vibronic coupling, this time between neighbouring electronically excited states, and the key role of non-radiative transitions in molecular photophysics (see below).

Before closing this section, mention must also be made here of Sydney’s ground-breaking exploration of refractory species detected via their luminescence, excited during laser vaporization from the surface of solid materials (5). This was an innovation later exploited by many others to study atomic and molecular clusters.

**Allegro Vivace: Orsay 1970–1993**

The creation of the new Laboratoire de Photophysique Moléculaire (LPPM) in 1967, with Sydney as its founding Director, opened the floodgates and LPPM rapidly became a major European centre of molecular physics—especially of molecular ions. Why ions? In Sydney’s words (14),

> Structural and dynamic information on (molecular) ions was not only of intrinsic interest but was vital for interpreting physicochemical processes operative in collision physics and chemistry; plasmas; radiation chemistry; mass spectrometry; photoelectron spectroscopies; astrophysics and aeronomy, especially planetary atmospheres and ionospheres, comets, cool stars, the interstellar medium and possibly . . . the birth and death phases of stars.

Reason enough for their study.

Ions generated in the laboratory, in electronically excited states, through electron impact or vacuum ultraviolet photoionization, could be observed through their fluorescence but often this was weak and hard to detect—which raised many interesting questions, not least, the nature of the competing ‘dark’ decay pathways and their intramolecular dynamics. In
the absence of collisions, these pathways involved radiationless transitions promoted by inter-electronic state coupling with neighbouring iso-energetic bound electronic states, or to continuum states leading to dissociation. Prior to the 1960s the non-radiative pathways had been very poorly understood but during that decade a ferment of activity rapidly provided a quantitative theoretical basis, the theory of radiationless transitions.

Sydney, with his great perception and fearless grasp of both theory and cleverly crafted experiment, very quickly recognized the unique advantages of using molecular ions to test the new theory, particularly their open-shell electronic structures and the relative ease of their manipulation and detection, compared with the equivalent neutral species. If their (exponential) fluorescence lifetimes, $\tau_F$, and quantum efficiencies, $\phi_F$, could be measured, their non-radiative transition rates, $k_{nr}$ (averaged over the spread of vibrational levels accessed in the initial excitation process), could be determined through the simple relationship $k_{nr} = (1 - \phi_F) \tau_F^{-1}$ (6,11).

To achieve this, an elegant armoury of photoion– or photoelectron–fluorescence photon coincidence techniques, PIFCO or PEFCO, was developed, in collaboration with Sydney’s gifted and green-fingered colleagues, John Eland (FRS 2006), Michel Devoret and, later, Gerald Dujardin. Measurements of the radiative (and non-radiative) quantum yields and decay rates of a wide range of ions, of increasing molecular complexity, provided rigorous quantitative tests of the three general cases of radiationless transition theory:

(a) the statistical case, where the accessible ‘dark’ quantum states were so densely spaced that they presented, effectively, a quasi-continuum (e.g. the polyatomic ions, C$_6$H$_6$$^+$, C$_6$F$_6$$^+$, C$_6$F$_4$H$_2$$^+$, ...);

(b) the resonance case, where the quantum states were widely spaced and sparsely distributed (e.g. the triatomic ions, CO$_2$$^+$, N$_2$O$^+$, SO$_2$$^+$, CICN$^+$, BrCN$^+$, ICN$^+$, ...); and

(c) the intermediate case, where the density of states within the accessible band width could be described as coarse-grained (e.g. the tetratomic ions, CICCH$^+$, CICCCI$^+$, ...).

In the ‘large molecule’ statistical case (a), exemplified by the photoexcited C$_6$F$_6$$^+$ ion, Sydney and his colleagues were able to confirm the two theoretically predicted ‘energy gap laws’: (i) an approximately exponential decrease in $k_{nr}$ as the electronic energy gap between the initial and final states increased, and (ii) an increase in $k_{nr}$ with increasing vibrational energy (12). Their vibrational-state-resolved data also revealed mode-selective inter-electronic coupling, a result facilitated by the availability of a tuneable synchrotron light source at Orsay coupled with zero (threshold) kinetic energy electron detection: T-PEFCO.* The local light source, the first in France, was constructed following Sydney’s prescient insistence in the 1970s and he subsequently played a leading role in establishing the European Synchrotron Source at Grenoble. At the other extreme, case (b), the combination of high-resolution spectroscopy with fluorescence quantum yield and lifetime measurements on individually resolved rotational levels of the CO$_2$$^+$ ion provided an exquisite benchmark example of the resonance case (8). Its relevance to the Mariner spacecraft observations of the Martian atmospheric air-glow was

* Threshold electron detection also gave access to indirect photoionization (autoionization) following excitation into neighbouring Rydberg states: the states detected had the same energy as the absorbed photon.
also ‘the cause of the gradual switching of Sydney’s interests towards astrophysical themes’ (22). Operation of the intermediate case (c) was demonstrated in a landmark investigation (with John Maier (FRS 1999) and William Gelbart) of the chloroacetylene ions, CICCH$^+$ and CICCCl$^+$, which revealed their signature non-exponential fluorescent decay and ‘offer[ed] a fertile new ground for testing strong and weak coupling theories of non-radiative electronic relaxation’ (9).

In most, but not all, of these examples, the competing non-radiative decay paths led directly to bound vibrational states of the ion. But what of the other pathways, where inter-state coupling led to fragmentation and to vacuum ultraviolet photochemistry—and ultimately, to chemistry between the stars? Sydney’s first steps along this path began in 1981 with a series of ultra-sensitive (and meticulously analysed) mass-selected ion-fluorescence (PIFCO) coincidence measurements of SO$_2$. Its photoionization in the energy range 10–21 eV excited fluorescence from SO$_2^+$ (10) and (much more strongly) its daughter ion, SO$^+$: a forerunner of Sydney’s pioneering studies of fragmentation of doubly charged molecular ions. This began three years later, again with SO$_2$, now photoionized by synchrotron radiation in the energy range 34–54 eV (13), this time using dual (fragment) ion–ion coincidence detection (PIPICO). It was soon followed by a ground-breaking, state-to-state investigation of CH$_4^{++}$ (17) and of the multiple fragmentation channels in the series CH$_3$X$^{++}$, X = OH, SH, CN, NH$_2$, CHO (18) using a triple, electron–ion–ion coincidence technique, PEPIPICO (developed in collaboration with John Eland at Oxford and Irène Nenner at Saclay). The expanding range of techniques reflected the need for enhanced energy resolution to unravel the potential complexity of the increasing number of possible direct, or sequential two- or three-body exit channels, e.g.

\[
\begin{align*}
ABC^{++} & \rightarrow AB^{++} + C \text{ (or } A + BC^{++}) \\
& \rightarrow A^+ + BC^+ \\
& \rightarrow A^+ + B^+ + C \text{ (or } A + B^+ + C^+) 
\end{align*}
\]

In the case of methane, two channels were identified. Excitation into a triplet state of its doubly charged ion led to CH$_3^+ + H^+$. At higher energy, excitation into an excited singlet state led to CH$_2^+ + H^+ + H$: Coulomb repulsion in this exit channel sent the two ions flying apart in diametrically opposite directions with the neutral H atom left as a stationary ‘spectator’. Fragmentation of the series, CH$_3$X$^{++}$, could proceed through a multiplicity of exit channels, in some cases through the likely intermediacy of isomeric, ylide dications, CH$_2$XH$^{++}$.

One of the fundamental drivers of Sydney’s interest in these exotic species, apart from his innate curiosity and the belief that good scientific research had a value in its own right as well as for its relevance in other fields, was, paradoxically, its potential relevance in other fields—notably, astrophysics and the ‘cosmo-chemistry’ of the interstellar medium (the ISM). The apparent absence of fluorescence from a molecular or fragment ion in a comet tail could simply reflect its vanishingly low quantum yield, $\phi_F$—but absence of evidence did not necessarily reflect evidence of absence: hence the value of the ultra-sensitive PEPICO technique which enabled measurements of $\phi_F \geq 10^{-5}$. Publication (18) ended with speculations about CH$_3$X$^{++}$ as a possible source of some of the (singly charged) fragment ions, HCO$^+$, HOC$^+$, HCS$^+$, HCNH$^+$, already observed in the ISM, and perhaps several others yet to be detected.

Sydney’s astronomical curiosity (in more senses than one) was more strongly manifested in the extended series of investigations of neutral and ionized polycyclic aromatic hydrocarbons (PAHs) which began in 1989, following a thoughtful earlier ‘prequel’ (15) that explored their
possible association with a series of unidentified infrared (UIR) bands observed in emission in the ISM, and, less likely, the diffuse interstellar bands (DIBs) observed in absorption, in the visible region of the spectrum. Its conclusion was characteristically cautious:

... if [authors’ italics] polycyclic aromatic hydrocarbons (PAHs) are present in the interstellar medium, conditions could exist for the formation of their doubly-charged cations. However, the fragility of doubly-charged polycyclic hydrocarbons, arising from Coulomb explosion processes involving direct dissociation and/or tunnelling effects, could also lead to destruction of these species or at least to deprotonation and loss of other charged or neutral fragments. There exists very little laboratory work of relevance to the fragmentation processes and yields of the doubly-charged cations of polycyclic aromatic hydrocarbons ...

—watch this space! The work began with an exploration of the photostability of the two simplest PAHs, naphthalene and its isomer, azulene. Rare-gas or synchrotron light sources (ACO at Orsay and BESSY in Berlin) provided the high-energy photons (8–35 eV), and threshold (T-PEPICO) and triple coincidence (PEPIPICO) techniques probed the fragmentation kinetics of their singly and doubly charged ions. In the rarified, collision-free environment of the ISM, could photoexcited ions survive long enough, typically ≥10 ms, to allow their relaxation through IR emission? In the naphthalene mono-cation, the principal fragmentation channel, unimolecular dissociation following non-radiative transfer into highly excited vibrational levels of its ground electronic state (internal conversion), could indeed be slow enough provided the photon excitation energy was less than about 14 eV. Subsequent measurements, extended to much larger PAHs, gave the first experimental evidence of increasing photostability, of neutrals as well as ions, with increasing molecular size and complexity (19).

SYMPHONY OF THE STARS: L’OBSERVATOIRE DE MEUDON 1993–2019

Sydney recognized from an early stage the way in which physical chemistry and spectroscopy could be brought to bear on questions relating not only to indigenous organic molecules in space, but also to those organics sent into space from Earth. In 2006, he began a collaboration with David Tepfer to study the effects of extra-terrestrial conditions (in low Earth orbit) on plant seeds. Using the European Space Agency (ESA)’s EXPOSE facility on the outside of the International Space Station, they were able to examine the effects of wavelengths of ultraviolet (UV) radiation not encountered naturally on the surface of the Earth (110–200 nm), but present above the Earth’s atmosphere, on biological materials (31).

Using seeds from Arabidopsis thaliana, they were able to show the efficacy of short-wavelength light in causing damage to the seeds. Spectroscopy revealed that the plant pigment quercitrin was able to screen longer wavelengths of UV light (with one absorption peak at 200 nm), confirming the inability of these pigments, and thus the seed coat, to protect the seed interior from wavelengths <200 nm. However, DNA integrity was low for seeds not exposed to the UV radiation, leading them to suggest that the site of damage was not the nucleic acid, but targets such as ribosomes or other UV-absorbing biomolecules (including those with π-electron systems). These data were an elegant example of the intersection of spectroscopy and space biology in addressing novel questions about the damage caused by extraterrestrial radiations using space technologies and wavelengths that cannot be easily investigated on Earth across the full spectral range. With the challenging limitations of small sample size and
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conducting experiments on space stations overcome, it became possible to publish several further papers on this topic.

Sydney was instrumental in encouraging a diversity of related activities. He was a frequent attendee and supporter of the ESA topical team, Geobiology for Space Settlement and Exploration (GESSE), a six-year ESA-funded team that investigated the role of microbe–mineral interactions in space applications. This ultimately led to the ESA project BioRock, which launched to the International Space Station in 2019 and studied microbial interactions on basalt surfaces in microgravity and simulated Martian gravity.

While engaging in these forays into astrochemistry and astrobiology, Sydney developed a powerful synthetic grasp of the role of physical chemistry in these wider fields. In a magisterial summary ‘Physical chemistry: extending the boundaries’ in the Springer book he co-edited on Astrochemistry and astrobiology, as part of the Physical Chemistry in Action series, he stated, speaking of early applications of physical chemistry in space sciences:

there also occurred the extension of physical chemistry from Earthbound considerations to those of the Solar System, with the interpretation of spectroscopic observations of comets and planetary atmospheres, in the first half of the twentieth century, amplified later by satellite studies and space probes and finally to the Universe as a whole . . . this has led in recent years to the determinations of the partial content and physical characteristics of exoplanet atmospheres, requiring physical chemistry knowledge . . . . These developments have also led to the birth of the field of astrobiology . . . ‘

(28). Thus, in his latter research years, Sydney saw, and actively encouraged, the expansion of physical chemistry as a tool to understand questions about biology and its place in the Universe in its widest context.

Accompanying all this activity in what he liked to term the ‘interstellar–Earth connection’, Sydney, at the normal age of retirement—and much beyond—also launched into diverse studies of molecules both large and small, exploiting his range of knowledge of spectroscopy to the full (figure 3). Among smaller species, he studied the photoelectron spectroscopy of formic acid and deuterated formic acid (23), yielding highly accurate ionization energies, illustrative of his abiding interest in precise, classical spectroscopy. The importance of formic acid, as a building block of biomolecules in space, of course did not go unnoted (see below).

This work was one of his early collaborations with Martin Schwell, at that time a young postdoc in Sydney’s group at Meudon, and with whom Sydney wrote more than 30 articles. Sydney went on to publish work on the photophysics of formic acid the following year (26), showing that below the astrophysical limit of 13.6 eV, photofragmentation is dominated by formation of $\text{OH} + \text{HCO}$ and $\text{CO}_2 + 2\text{H}$. Results demonstrated that the spectroscopy and photophysics of formic acid have potential applications in astrophysics and especially for exobiology. Formic acid in principle provides a route to interstellar amino acids, as found in the Murchison meteorite. This work, and numerous other related publications, were the result of collaboration with the Freie Universität Berlin, in particular with Helmut Baumgärtel and Hans-Werner Jochims.

In parallel, turning to much larger molecules, there are more than 35 publications involving fullerene-related species, where fullerene itself was the brainchild of his old friend Harry Kroto (FRS 1990) (among others). The most recent work involved laser flash photolysis and the photophysical properties of triplet $\text{C}_{60}$ in solution in cyclodextrin, showing how the triplet state is identical to that in $\text{C}_{60}$ in toluene solution (29). These and many other publications
illustrated how far Sydney had moved in applying his knowledge beyond that of his roots in classical spectroscopy. Returning to more conventional spectroscopic territory, a valuable overview of the electronic spectroscopy of C\textsubscript{60} and its adduct derivatives (27) emphasized their similarity in the visible and UV absorption regions. Their spectra were only mildly affected by changes in molecular symmetry or the size of the \( \pi \)-conjugation region. Astrophysical applications in the C\textsubscript{60} field were not neglected, as illustrated by (24), which discussed the most fruitful spectral regions, and associated astronomical contexts, in which to seek C\textsubscript{60} (or C\textsubscript{60}\textsuperscript{+}) in space, undetected at that time. Carbon-rich, dusty regions were suggested, involving a search for the 3980 Å band in C\textsubscript{60}. However, these conditions of dustiness and UV radiation are somewhat mutually exclusive, since short wavelengths do not penetrate dusty regions and cannot therefore be observed. Nevertheless, things were not so far from the mark, since C\textsubscript{60}\textsuperscript{+} was indeed identified in hot dusty regions, namely the photon-dominated region NGC7023, but in the 18–19 \( \mu \)m region of the spectrum, at which such regions are translucent. In passing, Sydney became very interested in \( \text{H}_2 \) formation in space and published on \( \text{H}_2 \) near-infrared emission in a very similar region, NGC2023 (20). These observations, at 2 \( \mu \)m, became the frontispiece of the book of abstracts for his so-called Gourmet’s Delight of Science, his title for the more than 200-strong meeting at the Paris Observatory, on the occasion of his so-called retirement in 1993. Coincidentally, he also published similar near-infrared spectra and images for NGC7023 (21), which proved to be the subsequent site of C\textsubscript{60}\textsuperscript{+} detection, but, as noted, found in the mid-infrared, rather than the near-infrared or the optical.

Moving sideways—as is so necessary in appreciating Sydney’s scientific career—he also worked as a visitor at the Foundation for Research and Technology Hellas (FORTH) in Heraklion. In collaboration with others, principally S. Couris and E. Koudoumas, he published 13 articles between 1996 and 2002 concerning the non-linear optical response of C\textsubscript{60} and higher fullerenes (25). Values of the second-order hyper-polarizability, \( \gamma \), were found to be much larger than expected from the theoretically predicted smooth dependence of \( \gamma \) on fullerene size. This behaviour could be rationalized in terms of the number of carbon
atoms in the fullerenes studied, the associated molecular symmetry, the aromaticity, molecular dimensions and resonance contributions, taking into account fast intersystem crossing in C$_{60}$ and C$_{70}$ and relatively weak crossing in C$_{76}$ and C$_{84}$, following 532 nm (10 ns) and 800 nm (100 fs) pulse excitation.

To the end, Sydney’s work continued to address basic aspects of molecular physics, for example (30), on cationic heats of formation, or again, in his very last publication (32), classic synchrotron-based electronic absorption spectroscopy of iso-quinoline. Juxtaposed, in 2016, one finds ‘An ESA roadmap for geobiology in space exploration’: he was truly a renaissance man of science. It is fitting that Sydney’s last published paper involved the use of synchrotron radiation, which he helped to pioneer in molecular physics (22) and which has subsequently become such a major tool.

**Sydney Leach: his legacy**

Sydney worked for 75 years in science, from the spectroscopy of anti-knock in aviation fuel, at Farnborough in World War II, to femtosecond laser spectroscopy of fullerenes and the electronic intricacies of coupling in polyatomic molecules and ions. He applied his knowledge in areas which cover a broad landscape, from the origin of life on Earth (and elsewhere, perhaps) to molecular physics in planetary atmospheres and in the interstellar medium. He has left his mark on numerous subjects, as we have sought to show, and this is felt through his scientific children, grandchildren and great-grandchildren, who populate the world’s laboratories. But his contribution was not just in the results that he achieved, but also in the style of their making. His benevolent manner of doing research, reflecting his own generous nature, has taught generations of budding scientists to listen with humility to Nature (figure 4). This approach was reflected in Sydney’s introduction to his magisterial, 70th birthday autobiography (22),* ‘Let me confess that I do science for the pleasure it gives to me—and to others.’* As a director of science, he was noted for the way he fostered creativity and helped promising new scientists through the administrative challenges of becoming established. He also exerted an important influence on the course of scientific research both in France and worldwide, through his founding and leadership of a major international laboratory (LPPM), or the ‘Sydney lab’, as it remained affectionately known for years. Indeed his influence continues to be felt through his foresight of where science is headed.

No description of Sydney would be complete without reference to his love of music and especially jazz—he played himself (figure 5)—and his literary and poetic production, some of which is very fine, but as yet unpublished. Literature was indeed very important to him, and he and Ida’s discussions centred mostly around art, literature and music. Sydney influenced many of his artist friends, and he collaborated with the composer Giuseppe Englert, with whom he published a paper (16), involving the transcription of the photoelectron spectrum of phosphabenzene into music, which at the time was most avant garde. In 2004 he wrote the libretto of an opera *Can machines think?* based on a text

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*To which the reader is enthusiastically referred for a more detailed and comprehensive survey of Sydney’s ‘pre-retirement’ scientific life, from the ‘horse’s mouth’.*
Figure 4. Sydney exchanging views with Pierre-Gilles de Gennes (Nobel Prize in Physics 1991), in the early 2000s. (Family collection; credit unknown.) (Online version in colour.)

by Alan Turing, which was publicly staged in David Tepfer’s Atelier de la Main d’Or in Paris.

Figure 5. Sydney at the keyboard. (Photograph by Aidan Un.)
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The portrait photograph was taken in 1993 by A. C. Copper and is Copyright © The Royal Society.

Author Profiles

David Field

David Field is Professor Emeritus of Experimental Physics at Aarhus University, Denmark. After completing his PhD at Cambridge, he moved to Göttingen as an ICI Fellow. It was during these Göttingen days, in 1974, that he first met Sydney at a memorable gathering in southern Germany, which included a very early encounter with isentropically cooled beam spectroscopy—a revelation at the time. Although David was much his junior, Sydney took him under his wing and they became friends for life after that first brief meeting. From Göttingen, David moved to Bristol, with extended visits to Nanjing and Paris, where, at the latter, he was appointed to numerous visiting fellowships at both Orsay and the Observatory. He was an avid user of the synchrotron sources, ACO and super-ACO, at Orsay. He left Bristol for Aarhus in 1998. Scientific interaction with Sydney was prompted, during visits to Meudon, through a common interest in how molecular hydrogen forms in space. This led to observational work using the Canada–France–Hawaii telescope, with several joint publications. When the Astrophysical Chemistry Group of the Royal Society of Chemistry was created in 1998, Sydney was its first president and David its first secretary. Following in Sydney’s mould, David was instrumental in 2008 in establishing the new synchrotron radiation source at Aarhus, where indeed the data for Sydney’s very last publication were gathered. Sydney was an invaluable mentor and ever-closer personal contact was maintained up to his very last days.

John Simons

John Simons FRS is Dr Lee’s Professor of Chemistry Emeritus at Oxford. After his PhD at Cambridge supervised by R. G. W. Norrish FRS, he moved first to Birmingham, then to Nottingham University, before his final appointment at Oxford. He first met Sydney Leach at a Free Radicals Symposium in Uppsala in 1961, and a few years later spent six months in Sydney’s laboratory at Orsay, as a young lecturer enjoying a precious spell of ‘study leave’. It led to a lifelong friendship, at first scientific, then professional and, in the course of time, personal. Although they never published jointly, at various times they shared several common research interests, particularly in molecular electronic spectroscopy, photophysics and photochemistry. During his spell as Honorary Secretary of the Faraday Division of the RSC, John was
delighted when Sydney agreed to serve as a European Representative on its Council. Later, the compliment was returned when John was asked to serve on the Comité de Direction of the Laboratoire de Photophysique Moléculaire. In 2001 John was invited to present the Humphry Davy Lecture at the Académie Française—following in Sydney’s earlier (1993) footsteps. In more recent years, they began to share common cultural interests: a friend for life.

Charles Cockell

Charles Cockell is Professor of Astrobiology at the University of Edinburgh. After his DPhil at Oxford, he moved to the NASA Ames Research Centre, California, before returning to the British Antarctic Survey, Cambridge. It was while working here that he first met Sydney at a meeting of the European Astrobiology Network Association. After this meeting, he maintained discussions with Sydney on a variety of topics that led to Sydney becoming an attendee at meetings of the European Space Agency topical team ‘Geobiology in Space Exploration and Settlement’ that Charles established in 2009, a group formed to consider how microbes could be used in space. Sydney offered valuable scientific and technical insights that eventually contributed to two flight experiments on the International Space Station. Later, Charles moved to the Open University and then to Edinburgh University. During this period, he, Sydney and Ian Smith FRS co-organized a Royal Society discussion meeting on the ‘Conditions for the emergence of life on the early Earth’ (2006). The three of them later co-authored the multi-author volume *Astrochemistry and astrobiology* published by Springer as part of their Physical Chemistry in Action series (2013). Most recently, in 2018, Sydney gave considerable depth of insight and lively critique on drafts of Charles’s popular science book, *The equations of life*, which explored the links between physics and biology.

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