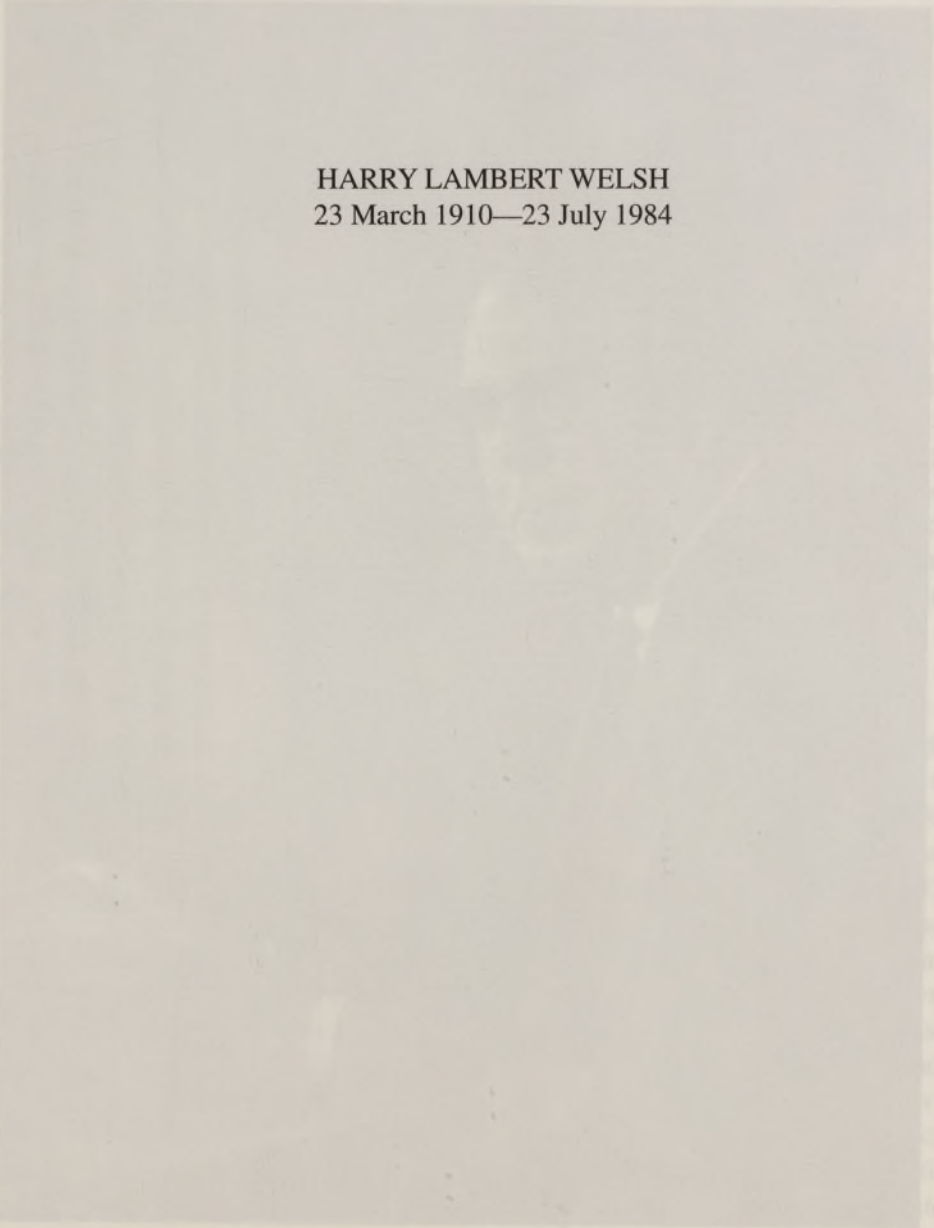

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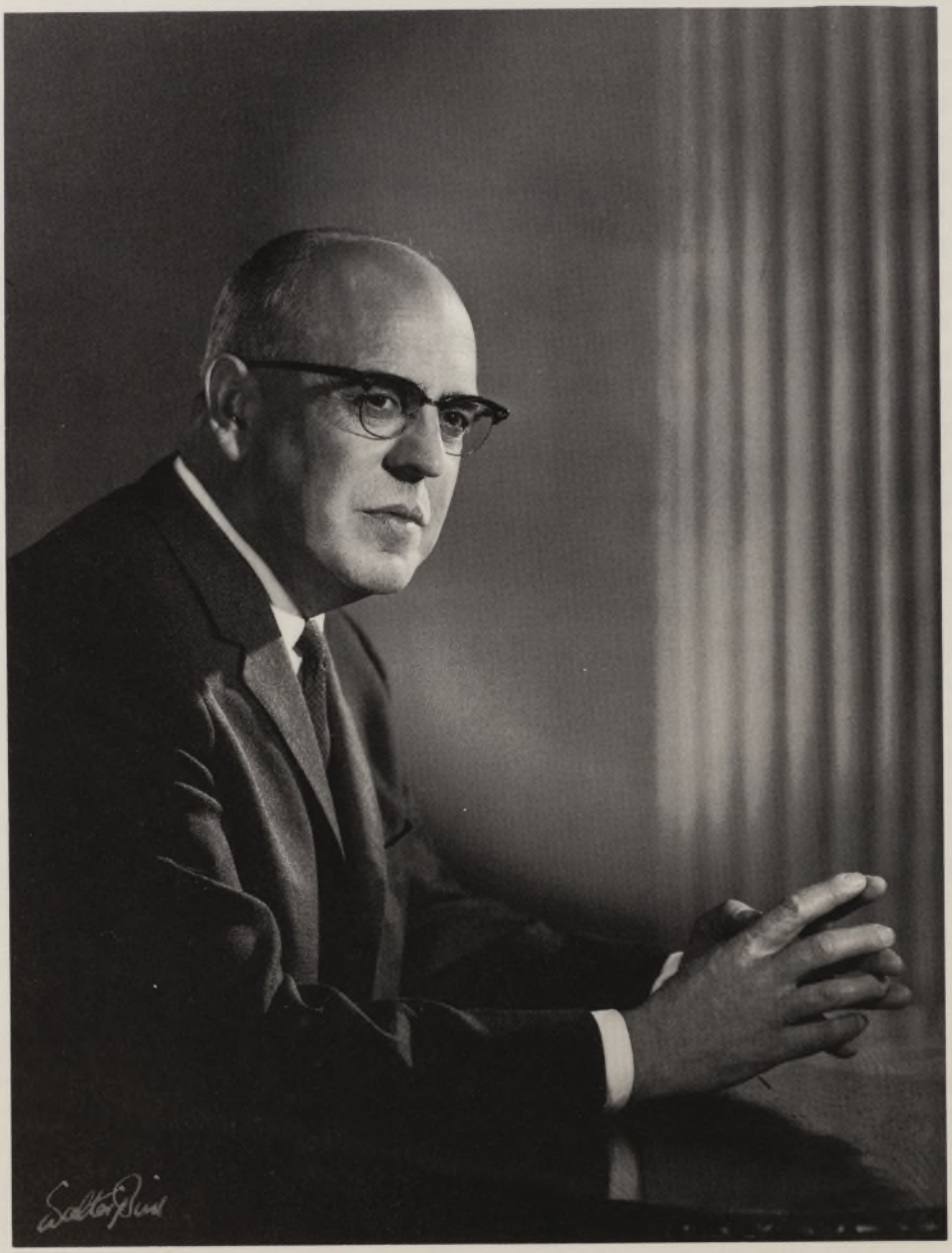

N. L. Walsh

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BY B.P. STOICHEFF, F.R.S.

HARRY LAMBERT WELSH began a long association with the University of Toronto with his enrolment in undergraduate physics in 1926. He brought fame to his Alma Mater with pioneering studies in molecular spectroscopy and intermolecular forces, and he played a major role in the development of the Physics Department with his introduction and establishment of research groups in theoretical, atmospheric, and high-energy physics. Undoubtedly, Harry Welsh's greatest achievement was the stimulation for scholarly research engendered in 65 Ph.D. students who had the privilege and pleasure to study under his supervision over a period of four decades. These scientists have made, and are continuing to make, important contributions to research in a variety of ways in universities, industry, and government institutions, across Canada and in other countries.

EARLY YEARS

Harry Lambert Welsh was born on 23 March 1910, on a farm 20 miles northeast of Toronto, near the town of Aurora. His father, Israel Welsh, was of Pennsylvania German descent. This side of the family arrived in York County, Canada, from the United States, in the early 1800s. His mother Harriet M. Collingwood, was of English ancestry. Her father came to Canada from Durham in 1845, and her mother from county Norfolk in 1836.

Israel and Harriet Welsh had two children: a daughter, Verna, and Harry. Although he lived on the farm until 11 years of age, Harry showed little enthusiasm for farming; he preferred school and reading. He attended a one-room primary school in the township of Whitechurch, County of York, from 1915–1921. A small travelling library, sponsored by the Women's Institute, visited Aurora regularly, and gave him the opportunity to read widely outside the school programme. At nine years of age, he read his first science book, an English translation of *Popular Astronomy*, by the French astronomer, Samarion, and this created a lasting impression on him. In later years, Welsh credited it for sparking his curiosity in astronomy, an interest which he maintained in his physics research. Harry inherited from his father a passionate love of music, particularly piano playing, which he pursued at a professional level throughout his life.

Upon the death of his father in 1921, the farm was sold, and Harry with his mother and sister moved to Aurora. He began his secondary school education at Aurora High School, which he completed in 1926, at age 16. His mother was the main influence during these

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years, providing encouragement in his scholastic activities and especially in his musical studies. He thoroughly enjoyed his years at high school. The teachers were very good, and he realized that teaching was an important and fine career. His scientific interests were confined to descriptive astronomy and mathematics, and to these he added the study of French, Latin and German (and Spanish, which he learned on his own). He also had a fine music teacher, and at that time, his chief ambition was to become a concert pianist. With high school completed, he worked that summer on a neighbourhood farm, and with his earnings bought the complete works of Chopin, which he dated 25 September 1926, and these remained in his collection.

At the urging of his mathematics teacher, Harry, at age 16, enroled in the challenging honours course of Mathematics and Physics at the University of Toronto. Harry's intention was to specialize in mathematics, but he was soon persuaded to switch to physics, by the masterful lectures of Professor John Satterly, which were always accompanied by vivid demonstrations of experimental physics. Fellow students in the mathematics and physics programme numbered more than 60, and included two of Harry's cousins, Carmen Miller and Harry Coleman, and close friends, Bob Ostrander and Leon Leppard. Two members of this class, John Tuzo Wilson and Byron Griffiths, along with Harry, later became professors at the University of Toronto, Wilson in physics and Griffiths in applied mathematics. Carmen Miller became a professor of mathematics at the University of Saskatchewan.

The Welsh family moved to Toronto in the autumn of 1926, and lived in a large house east of Riverdale Park, near the Teachers' Normal School. His mother took in boarders from the school to make ends meet, and this provided a lively house for the young Harry. In addition to his studies in physics, he registered in the Faculty of Music in his second year, taking the score study classes for three years. His daily schedule included physics in the mornings, usually by self study which he preferred, with attendance at only a few lectures. This was followed by music theory and piano practice, every afternoon. The celebrated Scottish pianist, Reginald Stewart, was his piano teacher during his years at university. Stewart had come to Canada to organize Gilbert and Sullivan operas, and founded the Promenade Symphony Orchestra giving concerts at Varsity Arena. At that time, Toronto and the University boasted a broad musical culture, including the Toronto Symphony Orchestra, the Hart House String Quartet, the Mendelssohn Choir, and Friday afternoon concerts at Convocation Hall.

In third and fourth years of undergraduate studies, Harry came under the influence of the dynamic John Cunningham (later Sir John) McLennan, F.R.S., Head of the Physics Department. McLennan thought that Harry spent too much time on the piano, and asked him one day 'When are you going to give up that damned music?' This was an exciting period in the Department, with visits by many giants of the time, including Cabannes, Fowler, Franck, Goudsmit, Hund, Kramers, Millikan, Van Vleck, among others. Harry completed the Bachelor of Arts degree in 1930. For six weeks that summer, he attended the famous Michigan Summer School, and heard lectures from Fermi, Ehrenfest, Goudsmit, and Uhlenbeck.

Graduate studies

Harry remained at the University of Toronto for graduate studies. At that time, research in the Department of Physics was mainly in atomic spectroscopy and cryogenics, Toronto being the second laboratory after Leiden to liquefy helium. Under the inspiring leadership of McLennan (the Director of the laboratory) the research activity at Toronto became widely known, and attracted postgraduate candidates from across Canada. Two investigations of the late 1920s are noteworthy. One was the excitation of the Auroral Green Line in the laboratory, with the establishment of oxygen as the carrier. The second was the observation of the Raman spectrum of liquid hydrogen, within a year of the discovery of the Raman Effect. This was the first report of a rotational Raman spectrum, and proved to be important not only because it showed that the molecules were rotating freely in the liquid, but also provided experimental proof that molecular hydrogen consisted of a mixture of two species, ortho- and para-hydrogen. In later years, research in Raman spectroscopy was to flourish at Toronto under the guidance of Harry Welsh, as was spectroscopy of molecular hydrogen in the gaseous, liquid, and solid states.

In 1930, the graduate students in Physics numbered about 20, with 12 working towards the Ph.D. degree, and most of them were supervised by McLennan. The professorial staff consisted of another five members, all of whom carried heavy teaching loads, yet managed to do some research in colloid physics, in heat and the physical and electrical properties of matter, X-ray diffraction, and geophysics. This group was assisted by a dozen junior staff, about half of whom were women. With his characteristic enthusiasm for research, McLennan made the rounds of the laboratory each day, with the greeting 'What's new!' In this atmosphere of research activity, Harry began experimental work on the Zeeman Effect of the Auroral Green Line, in September of 1930. A year later, he obtained the Master of Arts degree.

The personality and work of James Franck, Nobel Laureate in Physics (1925), who had lectured at Toronto in 1928, had so impressed the young Harry, that he decided to continue his graduate studies with Franck in Göttingen. He set out for Europe on the Mauritania, in September 1931, undeterred by the Depression and the political situation in Germany. On the way, he took the opportunity to visit London, Paris and Cologne. He was cordially received by Professor Franck, the Director of one of the three Physical Institutes at the University of Göttingen, and began research on the selective reflection of light by metal vapours, a topic he would return to in later years, in his own laboratory. While the physics courses were exceptionally good, the big event each week was the Colloquium, with lively discussions among Born, Cario, Eucken, Heitler, Kuhn, Nordheim, Pohl, Sponer, and Teller of the Institute, and many visitors. Harry also continued his piano studies, and attended operas in Berlin and Göttingen, with his many new musical friends.

With Hitler's coming to power in January 1933, followed by the distressing events he witnessed in Göttingen, and in particular, the resignation that April of Professor Franck, Harry realized that his stay in Germany was essentially over. After barely two years there, Harry left Göttingen. Although foreign students were not mistreated, these events marked Harry for life, and he declined to visit any country under a dictatorship. His next visit to Germany was to be made 20 years later.

On returning to Toronto in September 1933, Welsh found that Eli Franklin Burton was now Head of the Department of Physics and Director of the Laboratory. McLennan had retired in 1932 and was living in England. Otherwise, there was little change in senior or junior staff, but a noticeable reduction in research activity because of the continuing Depression, and more emphasis on teaching. Burton had been a member of staff in Physics for many years, with primary research interest in colloid physics. In his new role, he supported the continuation of research in low temperature physics and spectroscopy, and was soon to initiate work on electron optics, which eventually led to the building of the first electron microscope in North America. Welsh immediately began experimental research on atomic fluorescence, a new field for Toronto, but a topic which he had discussed with Franck. Spectrographs were plentiful, as well as a skilled glassblower for assembling sample cells and vacuum systems, but this work had to be carried out without a supervisor. He vividly remembered the visit of Dr Gerhard Herzberg in the autumn of 1935. Dr Herzberg had just arrived from Germany, and was on his way to the University of Saskatchewan, and a distinguished career in molecular spectroscopy and structure, culminating in the 1971 Nobel Prize in Chemistry. He was to become a very important friend through their common interests in physics and in music. Welsh completed the requirements for the Ph.D. degree in May 1936, with his thesis on 'Resonance fluorescences and sensitized fluorescences in the alkali vapours'.

University career

Upon obtaining the Ph.D. degree, Welsh accepted a job at the Duplate Glass Company, in nearby Oshawa, measuring the insulating properties of various materials. This work proved to be uninteresting, and he returned to the University in the autumn, as demonstrator, to assist H.J.C. Ireton and Elizabeth J. Allin in taking care of the fourth-year undergraduate laboratory. Both Ireton and Allin had received their degrees under McLennan; Ireton was soon to become Assistant Chairman of the Department, a post he held until retirement in 1958, and Allin and Welsh were later (1954) to begin a long series of investigations on liquid and solid hydrogen. Welsh took up research on the Raman effect in 1936 with Malcolm Crawford, then leader of the spectroscopy laboratory. They built an improved spectrograph for this work, and developed a method for obtaining Raman spectra of liquid drops. Together they received their first research grant just before the war, from the National Research Council (NRC), the federal granting agency for university research, and jointly supervised the first few graduate students in Raman spectroscopy.

With the outbreak of the World War II in 1939, research in the Department ceased, except for problems related to defense and the war effort. Under the direction of Burton, the staff became heavily involved in service courses for personnel from the Air Force, Army, Navy, and Medical Corps, in addition to teaching the regular undergraduate and graduate courses. Approximately 4200 attended special technical courses, whether in advanced radar, electronics, radio, or radiology, given from 1941 to 1943, in morning, afternoon and evening classes. Welsh played his part in this important training programme, by assisting in laboratory demonstrations and lecturing on electricity and magnetism. He was appointed a member of staff in 1941, and Assistant Professor in 1942.

On 13 June 1942, Harry Welsh married Marguerite Hazel Ostrander, the sister of his classmate Bob Ostrander. She was a school teacher with an aptitude for languages, and a special interest in French. With the completion of the service courses in September 1943, Welsh was asked to join the Directorate of Operations Research of the Royal Canadian Navy, in Ottawa. He served there with the rank of Lieutenant Commander for the duration of the War, with frequent visits to Washington, and two months in 1945 with the Admiralty in London. He worked chiefly on anti-submarine operations in Canadian waters and on convoy protection, with four other scientists. This work was challenging and interesting, but he felt that, at this stage of the War, it was perhaps not that important. He developed a high regard for one of his colleagues, Dr Andrew McKellar, a spectroscopist from the Dominion Astrophysical Observatory, and they became very good friends. The end of Welsh's war-work came shortly after VE-Day, his final responsibility being to interview the crews of two German submarines which had surrendered to the Canadian Navy. During their stay in Ottawa, both Marguerite and Harry found the time to acquire through private study, a knowledge of the Russian language and its literature. He also became proficient in the written language, and he later helped to translate articles from the Russian journal *Optika i Spektroskopiya,* published in English as *Optics and Spectroscopy*, by the Optical Society of America.

After the war, Welsh resumed his post at the University of Toronto. There he found a large influx of students, as young men returned from active service to take up postponed or interrupted studies. The number of undergraduates had increased from 8000 just before the war to 18 000 in 1947–48. Similar gains were realized in graduate studies in Physics, with 65 enroled for Masters or Ph.D. degrees in 1947–48, where half that number attended ten years before. Many of these chose to do their research with Crawford and Welsh, particularly in the new fields of infrared absorption and Raman scattering. Despite this increased activity, Welsh spent the summer of 1948 at the Dominion Astrophysical Observatory in Victoria, British Columbia, with his friend McKellar. He was offered a permanent position there, and came close to accepting, since he never lost his early interest in astronomy. Although he did not get along that well with Burton, Welsh chose to remain at Toronto. Later he was glad that he did, for he was extremely happy with his research during the many productive years ahead. He was promoted to Associate Professor that year.

Several changes in the administration of the Department took place in the brief period 1948–1950. After a leave of absence due to illness, Eli Burton died in the summer of 1948, and was succeeded by Edward Crisp (later Sir Edward) Bullard, F.R.S., of the University of Cambridge. Bullard's completely unbiased attitude immediately won him many friends, and his keen interest in computing and the Earth's magnetism helped the Computer Centre and the Geophysics Group, immensely. But his stay in Toronto was all too brief, with his return to England to become Head of the National Physical Laboratory in 1950. He was followed by William Heriot Watson, then Head of Theoretical Physics in the Division of Atomic Energy of the National Research Council, at Chalk River. The ever-increasing enrolment at the University at that time resulted in increased faculty, and the research activities of the Department were broadened to include Theoretical Physics and Nuclear Physics. By 1960, the number of professorial staff had risen to 26, twice that of the pre-war

years. Welsh's research flourished during this decade. He was appointed Professor of Physics in 1954, and he became Chairman of the Department in 1962, following Watson's resignation.

Welsh's term as Chairman was the period of the most rapid growth of the Physics Department. Along with the expansion of existing research groups, research in two new fields was established, namely high energy and particle physics, and meteorology. The faculty was increased to about 60, to supervise approximately 150 graduate students, and to teach over 3000 undergraduates enroled in physics courses. Welsh asked for an Associate Chairman to share the administrative duties of the Department, and chose George D. Scott for this new position. Scott was Crawford's and Welsh's first graduate student, and had joined the staff upon obtaining his Ph.D. degree in 1946. Welsh instituted more democratic procedures in the administration of the Department and its programmes. An executive committee, including five staff members, was elected to advise the Chairman, and a curriculum committee of members from the staff and student body was set up, and met frequently to discuss course content and student evaluation. With the ever increasing costs of research, and especially of 'big science,' Welsh also proposed the creation of a research board for the University, in order to review proposals and to make more democratic choices when large expenditures were requested. In the final years of Welsh's term, the long planning for a new and enlarged building for physics, astronomy and computer science, came to fruition on the opening of the McLennan Physical Laboratories and Burton Tower in September 1967, with Dr Herzberg turning the golden key. This special occasion was highlighted by lectures from six distinguished scientists, S. Chandrasekhar, F.R.S., R.H. Dicke, G. Herzberg, F.R.S., M. Kac, A. Kastler, and C.H. Townes. Welsh stayed on as Chairman until December 1968, when the move to the new building was completed.

In summarizing his six years as Chairman of the Department, Welsh has said 'These were happy years, we had plenty of money. Perhaps we overdid it in increasing the staff, but we didn't know at the time that the money would run out soon after. I am very satisfied with the new building, and happy about the development of the Department, especially about the new research areas in theoretical physics, nuclear and particle physics, and meteorology. I am proud of the quality of the staff, who all do research and take great care in preparation of lectures. I had no intention of staying on, even though I did not find administration to be a burden, thanks to Dave Scott who helped so much as Associate Chairman.' Thus, even during his years as Chairman of Physics, Welsh's research continued unabated, with new ideas and a stream of talented students. Now he was able to pursue his research interests full time. He accepted one additional administrative post at the University, serving from 1971–74 as the second Chairman of the Research Board, which had been established a few years earlier. In the summer of 1973, however, Welsh began to suffer mild heart attacks, with a more serious occurrence in early 1975. As a result, he decided to slow down his research activities.

In honour of Welsh's 65th birthday, former students and the Department of Physics inaugurated the H.L. Welsh Lectures, in May 1975, and these have continued as an annual celebration. For this event, a select few of the world's outstanding scientists are invited for two or three days each year, to Toronto to lecture at popular and technical levels on topics

of current interest and significance. At the first H.L. Welsh Lectures, over 500 friends and well-wishers, including 42 of Welsh's past students, came from all over Canada for the celebration. This gave Welsh much pleasure, as did the Banquet at Hart House, hosted by John R. Evans, President of the University of Toronto. An evening party with former students gathered around Harry and Marguerite was a fitting finale to this very special occasion, and a fond memory for all who attended.

By the time of his retirement in 1978, Welsh had supervised the research work of 64 Ph.D. students (another was added in 1979), an extraordinary contribution to the development of physics in Canada. In an interview that year, he explained that 'in looking back over my career, my biggest satisfaction came from these students, and through them, from my many "grandchildren" in physics.'

Scientific work

Welsh's interest in molecular physics and spectroscopy focused on intermolecular forces. For these investigations, his choice was molecular hydrogen, the simplest stable molecule for experimentation in the gaseous, liquid, and solid states. His excellent intuition for doing the right experiment was the hallmark of his distinguished research career. This characteristic was complemented by his innovative ideas of combining high-resolution spectroscopy with low-temperature and high-pressure techniques, as well as with low-pressure and light scattering experiments. Finally, it may be said that, while he appreciated the importance of precise measurements, Welsh undoubtedly derived the greatest joy from the search for, and observation and elucidation of, new phenomena.

Pressure-induced absorption

In 1946, Professors Crawford and Welsh purchased an infrared spectrometer with funds earned by the Physics Department for the courses taught to the Armed Services during World War II. For their first investigation, they planned to look for the infrared absorption spectrum of oxygen dimer molecules $(O_2)_2$, which had been suggested by G.N. Lewis to explain the magnetic susceptibility of liquid oxygen. They argued that, while the homonuclear O_2 molecule has no dipole moment and, therefore, no rotational or vibrational spectrum, the out-of-phase coupled oscillations of a pair of O_2 molecules would exhibit an absorption spectrum and provide evidence for the existence of $(O_2)_2$ dimers. Their graduate student, Jack Locke, took up the problem and readily observed a broad absorption band in liquid oxygen at the fundamental vibration frequency (1556 cm^{-1}) of the O_2 molecule. He found a similar spectrum in the compressed gas, as well as in compressed nitrogen and hydrogen at their fundamental vibration frequencies, 2331 and 4155 cm^{-1}, respectively. The absorption was found to vary as the square of the density, and Crawford, Welsh and Locke correctly interpreted their results, not in terms of bound dimers, but as a new phenomenon, an absorption due to the dipole moment induced by the distortion of the electron bond distribution by two molecules in close collision. Thus began a series of investigations which Welsh and his students carried on for three decades, leading to the discovery of a variety of spectra induced by intermolecular forces that are effective during molecular collisions. Professor Crawford took up related studies of infrared absorption induced by an applied

electric field.

The molecules, atoms and spectra examined in pressure-induced absorption at Toronto are summarized in table 1. Research was concentrated on hydrogen, partly because of the observed rotational structure in the vibrational spectrum, but mainly because hydrogen is amenable to theoretical interpretation. The rotational structure was explained by transitions obeying the selection rules $\Delta J=0, \pm 2$ (where J is the rotational angular momentum of the H_2 molecule), the same selection rules which hold for the Raman effect of diatomic molecules, thus confirming the interpretation of induced absorption.

Table 1. Spectra and systems studied in pressure-induced absorption.

spectra		molecules and atoms studied
vibration	(fundamental)	O_2, N_2, CO_2, H_2, D_2, $H_2 + N_2$, H_2 + rare gases, (He, Ne, Ar, Kr, Xe), $O_2 + N_2$ $(H2)_2$, $(D_2)_2$, H_2—D_2, H_2—Ne, H_2-Ar, H_2—Kr, H_2—Xe, H_2—N_2, H_2—CO, D_2—Ne,$(CO_2)_2$
vibration	(first overtone)	H_2, O_2, N_2
	(second overtone)	H_2
	(double transitions)	H_2, $H_2 + CF_4$
rotation		H_2, $H_2 + N_2$, H_2 + rare gases (He, Ne, Ar, Kr, Xe)
	(double transitions)	H_2
translation		H_2, H_2 + rare gases (He, Ne, Ar, Kr, Xe), mixture of rare gases (He—Ar, He—Ne, Ne—Ar)
electronic		O_2

Induced absorption spectra of the fundamental and first overtone of pure H_2 and of H_2 perturbed by the rare gases and by nitrogen, were investigated at a variety of pressures (as high as 5000 atmospheres) and temperatures (in the range of 300 to 18 K). From the band intensities, shapes and widths, it was clear that the observed spectra were composed of two different types of spectra. This was in complete agreement with the prediction of Jan Van Kranendonk, of Amsterdam, who showed in his Ph.D. thesis of 1952, that two mechanisms can produce the induced dipole moment in a gas of homonuclear molecules. One is a short-range induction due to overlap forces [$\mu \propto \exp(-aR)$] that contributes mainly to the broadest bands, corresponding to transitions with $\Delta J=0$. The other is a long-range induction due to the molecular quadrupole moment [$\mu \propto 1/R^4$] that contributes to much narrower components. This interaction is strongly dependent on the mutual orientation of the pair of molecules, and results in transitions with $\Delta J=0, \pm 2$. In addition to these main components that occur when vibrational and rotational transitions take place in only one molecule of a colliding pair, weaker components were observed as double transitions, that is, when both

molecules of a colliding pair make transitions.

The discovery of pressure-induced absorption in the fundamental and first-overtone vibrational spectra of hydrogen, at Toronto, prompted other groups to take up research in this field. Two important experiments will be briefly mentioned here. At the National Research Council in Ottawa, Gerhard Herzberg postulated that a broad feature at 8200 Å, observed by Gerard Kuiper in the spectra of the planets Uranus and Neptune, might be explained by the pressure-induced, second-overtone band of hydrogen. He carried out a long-path experiment at liquid nitrogen temperature, and confirmed his idea in 1952, a year after the announcement of the first-overtone spectrum. This was the first proof of the presence of hydrogen in the atmospheres of the major planets. In 1955, at the University of Amsterdam, Ketelaar and colleagues observed the pressure-induced pure rotational spectrum of hydrogen for the first time. While investigating this spectrum in more detail, Welsh found that the rotational spectrum was superimposed on a continuum arising from a purely translational spectrum, induced by intermolecular forces. This result led to the discovery of translational spectra for mixtures of the rare gases, by Zoltan Kiss and Welsh in 1959. Such spectra occur for any pure gas of non-spherical molecules or any mixture of gases. However, translational spectra cannot occur for a pure atomic gas (or a gas of identical spherical molecules) since the colliding pair would have a centre of symmetry and hence no dipole moment.

When the hydrogen pressure-induced spectra were examined at low temperatures and high dispersion, Welsh and his students found that the maximum of each of the components exhibited a superimposed fine structure. By working at temperatures of 20 K and at one atmosphere pressure (with long path lengths), clearer spectra of this fine structure were obtained, and these were shown to be due to the formation of bound and quasi-stable molecular complexes. This was the first direct spectroscopic evidence of an $(H_2)_2$ molecule. Spectra of the complexes H_2–Ar, H_2–Kr, H_2–Xe, were also observed in mixtures of hydrogen with argon, krypton and xenon, at temperatures just above the boiling points for the rare gases. Much of the research on these complexes was carried out by a graduate student, A. Robert McKellar, the son of Welsh's close friend, Andrew McKellar.

Pressure-induced absorption was also detected in the electronic spectrum of O_2. It is well known that oxygen exhibits absorption in the infrared and visible regions arising from electronic transitions, which are forbidden by electric dipole selection rules, but occur as magnetic dipole radiation. These band systems were studied in O_2 at pressures of 50 to 150 atm, and in O_2—N_2, O_2—Ar, and O_2—He mixtures up to 3000 atm total pressure. From these studies, Allin and Welsh concluded that the intrinsic absorption in the red system is at least ten times greater than that of the infrared system, and that the induction effect in O_2—O_2 pairs alters the potential energy curves considerably, but not in O_2-foreign gas collisions. The collision-induced absorption in oxygen was assumed to be an electric dipole absorption and therefore distinct from the magnetic dipole absorption. This was borne out in the observed spectra: the induced absorption for the O–O band is detectable over a broad range of 800 cm^{-1}, while the corresponding magnetic dipole spectrum extends over approximately 200 cm^{-1}.

Here it should be mentioned that in the experimental research at high pressures, Welsh

was ably assisted by Dr Jan Stryland, of the Van der Waals Laboratory in Amsterdam, whom he attracted to Toronto as a staff member in 1954. They were to co-operate in a series of experiments on high-pressure effects in Raman and infrared spectra, until Welsh's retirement. Welsh was also able to persuade Dr Jan Van Kranendonk to join the staff at Toronto, in 1958. He was instrumental in helping to interpret the many new observations and to provide a comprehensive theory of pressure-induced absorption. This close collaboration was to continue with the study of the infrared and Raman spectra of solid hydrogen, and of line broadenings and frequency shifts in the Raman spectra of compressed gases.

Infrared and Raman spectra at high pressures

As the early experiments on induced infrared absorption progressed from studies of liquids to compressed gases, Welsh formed the idea of doing research in high-pressure spectroscopy. He was encouraged in this pursuit by A. Michels of Amsterdam, an authority in the field of high pressures, who visited Toronto on several occasions. Welsh began research in the spectroscopy of compressed gases with a $5000 NRC grant for high-pressure equipment, designed and built in Amsterdam. He was assisted, briefly, by a junior staff member, P.E. Pashler (who left to join the General Electric Company in 1950), and graduate students Andrew F. Dunn, working on the infrared spectrum of methane, and myself, experimenting with the Raman spectrum of carbon dioxide.

As already discussed, much of the research in infrared spectroscopy at high pressures was centred on hydrogen and other homonuclear diatomic molecules, since rotational and vibrational spectra are not active for the free molecules, and their appearance at high pressure is a result of intermolecular forces. Investigations of the effects of high pressures on allowed (or active) infrared transitions from polar diatomic or polyatomic molecules were also carried out, to look for intensity changes, line broadenings or frequency shifts. Experiments with CO and mixtures of CO–He, and with CH_4 and mixtures of CH_4–He, CH_4–Ar, and CH_4–N_2, were attempted, up to pressures of 3000 atmospheres. Frequency shifts were not detectable in the pressure-broadened bands. However, the intensity of each allowed band showed enhancement when the absorbing molecule was surrounded by polarizable foreign-gas molecules at high densities. This increase varied linearly with density, and extrapolation to zero foreign-gas density yielded absolute intensities of the infrared fundamental bands for the free molecule.

Very few systematic studies on frequency shifts and broadenings of Raman lines with pressure had been carried out before such investigations were initiated in Toronto in 1950. It was known that rotational ($\Delta J = \pm 2$) and rotation–vibrational ($\Delta J = \pm 2$, $\Delta \nu = 1$) Raman lines arise from anisotropic (depolarized) scattering, and are broadened by collisions. In contrast to infrared spectra, the dominant features of vibrational Raman spectra are sharp, line-like Q-branches ($\Delta J = 0$, $\Delta \nu = 1$) associated with isotropic (polarized) scattering. Crawford and Welsh found that these remain relatively narrow at any gas density, and in liquids and solids. Investigations of pressure effects on both types of spectra were carried out over a range of pressures, and compared with theories developed by Van Kranendonk, and by Gray, at Toronto.

Initial, qualitative studies on rotational spectra were superseded as high-resolution Raman techniques were developed. For the nonpolar molecules O_2, N_2 and CO_2, a linear dependence of linewidth with pressure was established, with the broadening coefficient (width/pressure) decreasing monotonically with increasing rotational quantum number J. For the polar molecule HCl, much larger broadening coefficients were found, with a maximum at low J and more rapid decrease with increasing J. These different behaviours for polar and nonpolar molecules were in accord with theory. For H_2, H_2—He, H_2—Ar mixtures, with total pressures up to 2000 atm, the rotational lines showed symmetrical Lorentzian line shapes, but with broadening coefficients an order of magnitude smaller than obtained for O_2 and N_2. Frequency shifts were measured and found to fit a 'virial-type' expansion in the relative density, with linear and quadratic terms. A similar dependence was found for the individual Q-branch lines at the high pressures. At lower pressures, the linear term for frequency shifts with pressure for the Q-branch lines of H_2 and D_2 was shown to depend on temperature, in the form $a_J = a_i + a_c\ (n_J/n)$. Here a_i is due to isotropic intermolecular forces, and is temperature dependent, while a_c arises from vibrational coupling through dispersion forces, and is nearly independent of temperature, and n_J/n is the fractional population of the initial rotational level J. An analysis based on a (6–12) intermolecular potential showed that the dispersion forces lead to a negative shift and the overlap repulsive forces to a positive shift and cause a near cancellation of the linear term. This is in agreement with the small observed shifts at room temperature at low densities, and with the quadratic dependence at the higher densities. The results of this analysis are also in agreement with the shifts observed in solid hydrogen.

Spectra of liquids and solids

Crawford and Welsh first began research in Raman scattering with work on condensed media. In long exposures, each of the spectra of the tetrahedral molecules CCl_4, $SnBr_4$ and $SnCl_4$ revealed from 15 to 20 faint lines in addition to the four fundamental vibrational lines, and all were assigned as overtones and combination tones of the fundamentals. A careful investigation of the intensity contour of the Raman spectrum of rock salt (NaCl) taken at high dispersion was interpreted in terms of the Born theory of lattice dynamics, and was one of the earliest confirmations of this theory. Rotation–vibrational Raman bands of liquid O_2, N_2, CH_4 and solid CH_4 were examined. In all of the spectra, the Q-branches associated with isotropic scattering were sharp, while Q-branches and rotation–vibrational wings associated with anisotropic scattering exhibited broad continuous bands whose intensity contours were consistent with partly free molecular rotation.

In 1954, Allin and Welsh began a research programme on infrared and Raman spectra of molecular crystals at low temperatures, and were the first to observe the Raman spectrum of solid hydrogen. The research on solid hydrogen was a natural extension of the work on the pressure effects in the spectra of the compressed gases. It was also unique, since there is practically free rotation in solid hydrogen, and the population is confined to the two lowest states, or ortho- and para-molecules, so that one type behaves as an impurity in the lattice structure of the other molecular form.

In solid H_2 and D_2, four Raman lines were observed, two rotational lines of width

approximately 10 cm^{-1}, and two sharp vibrational (or Q-branch) lines of width 0.4 cm^{-1}, one of each arising from para molecules and from ortho molecules. For HD, only two lines were observed, one rotational and one vibrational, since at the temperature of 2 K, the population in HD is all in the lowest rotational level, J=0. The widths in HD were 5 cm^{-1} and 0.2 cm^{-1}, respectively. For all three solids, the frequencies are close to those of the liquids, but the rotational lines are shifted by a few cm^{-1} and the Q-branch lines by about 10 cm^{-1} from those of the free molecules. These shifts agree with an extension of the theory, taking into account the isotropic molecular interaction and vibrational coupling used to explain the frequency shifts in the compressed gases. In pure para-H_2 (that is, J=0) and pure ortho-D_2 (J=0), the rotational Raman line becomes narrower and splits into three almost equally-spaced components. This was explained by Van Kranendonk and Karl by the existence of travelling waves or rotational exciton bands, due to coupling between pairs of rotating molecules. Similarly, vibrational and rotation-vibrational exciton bands were postulated. These were necessary to explain the observed intensity contours and intensity anomalies for various ortho-para concentrations. The anomaly of interest concerned the intensity ratio Q(1):Q(0) of the J=1 and J=0 vibrational lines, which for H_2 should be unity, based on the population of the two levels at the temperature of the solid. But large deviations from this value were found. For example, the intensity ratio varied from 3.8 at very low ortho concentration to 2.3 for the largest ortho content of 75%. In deuterium, the effect was much more marked, being approximately 50 times the concentration ratio. Quantum mechanical calculations of the enhancements based on excitons and the effects on rates of change of polarizability agreed with the measured values.

In the infrared spectrum of solid hydrogen, three main groups of maxima were observed in the region of the fundamental band. These are correlated with the Q-branch and the para and ortho rotation-vibrational lines of the collision-induced spectrum of the gas. Each group consists of a sharp and a broad component. The sharp components are due to induction by the molecular quadrupole moments, and the broad components are summation tones of translational lattice frequencies with the molecular vibration and rotation. At higher resolution, the sharp components each consist of two distinct lines. One is a weak line at the frequency of the Raman line and originates in a transition in a single molecule, and the second is a much stronger line whose frequency corresponds to a double transition, one being a vibrational transition in one molecule, the other a rotational transition in another molecule. In liquid hydrogen these lines are more diffuse, but the greater intensity of the double transitions is as marked as for the solid. When the ortho-para ratio of solid hydrogen was altered, the observed spectrum changed in quite remarkable ways, with further splitting of the vibrational and rotation-vibrational lines and large changes in relative intensities of the lines. Evidence for quantum diffusion of ortho-molecules in almost pure solid para-hydrogen was also found, in the form of an intensity increase as a function of time after solidification, for a band identified to be due to the reorientation of neighbouring ortho-H_2 pairs. The infrared fundamental band of H_2 dissolved in solid Ar and solid Kr, and of D_2 in solid Ar were also examined, and show the three main maxima, each with similar patterns of five components. All of the above features observed in the spectra of solid hydrogen were satisfactorily explained in terms of travelling vibrational, rotational and orientational

excitations (or vibrons, rotons and librons), and by including the order-disorder phase transition which occurs at 2.8 K for pure ortho-hydrogen. This research inspired a stream of students at Toronto, and many additional experimental and theoretical studies on the spectrum of solid hydrogen, at other institutes around the world.

High-resolution Raman spectroscopy

With the above research programmes on pressure-induced infrared absorption, on spectroscopy of compressed gases, and of liquids and solids, all having been started in the late 1940s and early 1950s, and being carried out concurrently for two to three decades, it comes as a surprise to find that Welsh was involved in yet another extensive research project, namely high-resolution Raman spectroscopy. Twenty students (including myself) were to receive their Ph.D. degrees in the period 1950 to 1979, helping to develop new techniques, and to use these to investigate the rotational and rotation–vibrational Raman spectra of the molecules listed in Table 2. During the initial studies of the spectra of compressed gases, it became apparent that the high-intensity, water-cooled mercury arc developed at Toronto was an almost ideal source for exciting the Raman spectra of gases at low pressures. Further improvements of the arc (which became known as the Toronto lamp), and the conception and development of the multiple-reflection Raman cell, made high-resolution Raman spectroscopy possible. The group also designed and built high-speed, high-dispersion spectrographs using blazed diffraction gratings, increased the effective light-gathering power of the spectrograph with a cylindrical lens placed in front of the photographic plate, and baked the photographic plates to increase their sensitivity. With these various developments, high-resolution Raman spectroscopy became an important complement to infrared and microwave studies for the determination of molecular constants and structures. In the 1970s, the mercury lamp was superseded by the laser, which provided even higher intensity, and substantial increase in resolution.

Table 2. Molecules studied by Raman spectroscopy

	spectra	molecules
gases	rotation	O_2, N_2, H_2, D_2, CO, CO_2, HCl, C_2H_2, C_2D_2, C_2H_4, C_2H_6, C_3H_6
	vibration	H_2, D_2, N_2, O_2, HBr, HCl, CO_2, NH_3, CH_4, CH_2Cl_2, CH_3Br, CH_3Cl
	rotation–vibration	H_2, D_2, C_2H_2, C_2D_2, C_2HD, NH_3, CH_4, CD_4, CH_3D, C_2H_4, C_2H_6, C_2D_6, $C_2H_3D_3$, C_3H_6, C_6H_6, C_6D_6
	electronic	NO
liquids		H_2, D_2, HD, N_2, O_2, CO_2, NH_3, CH_4, CCl_4, $SnBr_4$, $SnCl_4$, C_2H_4
solids		H_2, D_2, HD, NaCl, NH_3, C_2H_4

Raman spectroscopy proved to be especially valuable in two general areas of molecular studies: rotational spectra of nonpolar molecules (forbidden by selection rules in infrared and microwave spectra), for the determination of ground state structural parameters; and rotation–vibrational spectra of highly symmetric polyatomic molecules for which the rotational and vibrational selection rules for Raman scattering and infrared absorption may be different.

Methane, CH_4, is an example of a highly symmetrical molecule for which the Raman and infrared spectra are very different, and it was chosen for the first detailed investigation. The rotational selection rules for the triply degenerate vibration, ν_3, are $\Delta J=0, \pm1, \pm2$, for the Raman effect. Since each of the upper state levels is split into three, a total of 15 branches is allowed in the Raman spectrum, leading to a rich and complex spectrum. In contrast, only three branches are active in infrared absorption. Over the years, progressively improved Raman spectra were recorded, and once a complete theory became available, these data were used to evaluate molecular constants and the internuclear distance

r(C—H)=1.0939 Å. Three of the four fundamental bands, ν_1, ν_2, and ν_3 were recorded and analysed for both CH_4 and CD_4, while ν_4 was too weak to be observed.

Another important example for which the Raman spectrum provided unique data was the structure of the ethane molecule, C_2H_6. The selection rules for a doubly degenerate vibration lead to one set of Q-branches for the eclipsed configuration of the two CH_3 groups, and to two sets of Q-branches for the staggered configuration. The identification of two series of Q-branches in the Raman spectrum of C_2D_6 (and its confirmation in C_2H_6) established the staggered structure unequivocally. The Raman spectrum of ethane also led to the direct determination of the small moment of inertia about the symmetry axis (along the C–C bond) of the molecule, and to the internuclear distance r(C–H)= 1.095±0.002 Å. Corresponding data for C_2D_6 yielded r(C–D)= 1.093±0.001 Å.

In summary, the rotational and rotation–vibrational Raman spectra of the polyatomic molecules listed in table 2 were investigated in considerable detail (with those of the acetylenes and benzenes being obtained with laser excitation). Analyses of the spectra, using the extensive computing facilities of the Department, yielded molecular parameters for the ground and vibrational levels, as well as internuclear distances and bond angles for these important molecules.

Honours

The scientific achievements of Harry Welsh brought international recognition to him, to his laboratory, and to the University of Toronto. As we have seen, many graduate students were attracted to his research, as were many postdoctoral fellows and visiting scientists. The fields of research which he initiated at Toronto developed worldwide followings, and several international conferences and summer schools have been devoted to these topics. Welsh was elected a Fellow of the Royal Society of Canada in 1952, and of The Royal Society of London in 1962. He was a Fellow of the American Physical Society (1957) and of the Optical Society of America (1964). He was a Member of the Royal Astronomical Society of Canada and served as President of the Toronto Centre of the Society in 1956. An active Member of

the Canadian Association of Physicists, he was awarded its Medal for Achievement in Physics in 1961 and elected its President for 1973–74. In 1963, he received the Tory Medal of the Royal Society of Canada, and in 1974, the W.F. Meggers Medal of the Optical Society of America. In 1972, Welsh was appointed an Officer of the Order of Canada, Canada's highest distinction for outstanding achievement and service to the nation. Welsh received Honorary D.Sc. degrees from the University of Windsor and Memorial University of Newfoundland. In May 1984, on the occasion of the tenth in the series of H.L. Welsh Lectures, he received the L.L.D. degree from his Alma Mater, the University of Toronto.

Personality

Harry Welsh was widely respected in the physics community across Canada, and by molecular spectroscopists internationally. His colleagues found him to be a quiet man with a gentle wit, and in all respects a gentleman. He was a scholar whose knowledge extended beyond science to music, languages, literature and history. In his scientific work as well as in the administration of the Department, he combined the qualities of leadership, good judgement and integrity.

Welsh was regularly invited to speak at international conferences, and he participated in numerous scientific gatherings in Europe as well as in India and Israel. He and Marguerite particularly enjoyed a sabbatical year in France in 1956–57. Welsh rarely missed the Annual Congress of the Canadian Association of Physicists (CAP), and Meetings of the Royal Society of Canada. His students remember well the convoy of cars on annual drives, with occasional picnic stops, to a nearby CAP Meeting, or to the Molecular Spectroscopy Symposium in Columbus, Ohio, or further afield, to Gordon Conferences in New Hampshire. These were opportunities for students to present their research (after careful rehearsal at home) and to meet their peers, as well as the leaders in science; truly heady and stimulating experiences for budding young scientists.

Harry and Marguerite enjoyed the company of friends, and their home was often the scene of a party on a Saturday evening. Usually these began with dinner, prepared by Marguerite, sometimes followed by a musical recital with Harry as piano accompanist. He had many musician friends, and it was known that he practised on a regular basis with vocalists, violinists and cellists, many of whom were colleagues, visiting scientists, or the talented children of his students. It also became a tradition that the final few drafts of a Ph.D. thesis or paper were revised in the quiet of the Welsh home. Students vividly recall their study of grammar and fine points of syntax with Harry far into the night, while Marguerite kept up a bountiful supply of refreshments.

Welsh suffered several years of illness, and endured numerous physical discomforts in his last few years. This he did without complaint, as he continued to come to his office on a daily basis, and to keep up with science and music as best he could in his weakened condition. In this respect, he was a source of strength and inspiration to all, and a true gentleman to the end. He died of a stroke on 23 July 1984.

Harry Welsh will long be remembered by his many friends and colleagues for his kind and considerate nature, his sincerity and integrity, and especially for his interest in the well-being of his students, as well as for his outstanding scientific accomplishments. With

his death, the scientific community lost a leading figure in molecular physics and spectroscopy, and Canada lost one of its most distinguished physicists.

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Bibliography

The complete bibliography appears on the accompanying microfiche. A photocopy is available from the Royal Society library at cost.