

The Laboratory Founded by Van der Waals¹

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During the past century, the Van der Waals Laboratory at the University of Amsterdam has been the principal provider of reliable fluid property data over large ranges of pressure and temperature. This paper describes the history of the laboratory, starting in 1898 when funding for it was obtained. In the early period, under Van der Waals and Kohnstamm, the high-pressure direction was chosen, and the first PVT and phase equilibria data were published. The main focus of this paper is the Michels period, from 1921 to 1960. In this period, the laboratory acquired its own building and assumed a unique position in the world because of its highly accurate thermodynamic, transport, and other property measurements in fluids at high pressures. In the 1950s, a second laboratory was built by Michels at the University of Maryland, per request of the U.S. Navy. Under Trappeniers, 1961–1987, the laboratory incorporated new techniques, such as NMR, undertook a major expansion of the pressure range, and extended its interest to phase transitions in molecular solids. The position of the Van der Waals Laboratory in the world of high-pressure science is highlighted.

KEY WORDS: high pressure; history; molecular physics; NMR; phase equilibria; PVT data; thermal conductivity; Van der Waals Laboratory; viscosity.

1. INTRODUCTION

During the past century, the Van der Waals Laboratory at the University of Amsterdam has been a major provider of accurate thermophysical property data at high pressures. The organizers of the Fourteenth Symposium on Thermophysical Properties considered it desirable to highlight

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the history of this laboratory at a time when it is undergoing major transformation, which includes termination of much of its traditional work.

In this paper, such a history is presented. In addition to describing facts, dates, and outputs, the paper shows the motivation for the undertakings. What did the founder have in mind? What models did he and his successors emulate? Why were the directions chosen the way they were? What were the major achievements? and What was the impact? The paper is written in four sections. Section 2 describes the early period of Van der Waals and Kohnstamm, 1898 to 1920. Sections 3 and 4 concern the Michels period, 1920–1960; and Section 5, the Trappeniers years, 1961–1987, and thereafter. The main emphasis is on the Michels period, in which the laboratory carved out its work area, acquired its own building, built its reputation, and became known worldwide.

2. THE VAN DER WAALS FUND

For a proper understanding of the origin of the Van der Waals Laboratory, it is necessary to consider the circumstances of thermodynamics in The Netherlands in the late 1890s [1]. J. D. Van der Waals (1837–1923) had his major achievements well behind him: the equation of state, the theorem of corresponding states, the theory of mixtures, and the theory of capillarity. His reputation was solidly grounded, and the experimental study of phase separations in mixtures was spreading all around him. He had had a pioneering role in experimental studies of fluid mixtures. Thus, he was exploring the phase behavior of mixtures of carbon dioxide and air in Cailletet tubes in the early 1880s, being as mystified by the results as Cailletet himself, who did similar experiments in France at that same time.

His close friend H. Kamerlingh Onnes (1853–1926), however, had since built a famous physics laboratory at the University of Leiden, where he was well on his way toward the liquefaction of helium. He had tested Van der Waals' theory of mixtures in glass vessels at pressures up to approximately 100 bar, obtaining the first reliable and interpretable results by introducing a stirrer.

The Chemistry Department in Amsterdam, under the leadership of H. W. Bakhuis Roozeboom (1854–1907), was actively studying phase equilibria in liquids and solids in Cailletet tubes, vertical glass capillaries sealed at the top and immersed in mercury at the bottom. By injecting mercury, phase transitions could be observed, and crude volumetric data obtained at pressures up to about 100 bar.

In France, E. H. Amagat was a pioneer in high-pressure studies. He was able to generate pressures up to 3000 bar, and for calibrating his manometers he had installed mercury columns in deep mine shafts. Also,

he had modified the Cailletet tube, enclosing it in an autoclave, and providing it with platinum contacts at intervals along the tube. This way, *PVT* data could be obtained up to high pressures.

While this experimental activity was going on all around him, Van der Waals had barely any facilities he could call his own. This was increasingly felt to be embarrassing. Thus, at the 25th anniversary of Van der Waals' 1873 thesis, colleagues and friends decided to establish the Van der Waals Fund "to supply monetary support for about the next ten years for experimental research related to the theories put forward by the eminent scientist and to be conducted in Amsterdam under the supervision of the founder of the theory."

At first, several institutions contributed, while, later, individuals paid a yearly subscription fee of five Dutch guilders. The history of this fund can be found in Ref. 2.

It must not have been easy for Van der Waals, as the Director of the Physics Laboratory while heavily loaded with teaching, to get an experimental laboratory off the ground and moving. Fortunately, a capable Ph.D. student under his supervision in the 1890s, Ph. A. Kohnstamm (1875–1951), took an interest in the laboratory. Van der Waals decided to dedicate the laboratory to studies of phase equilibria and molecular interactions at high pressure. This was an obvious choice in The Netherlands, in view of the pressure limitations experienced by Kamerlingh Onnes and by Bakhuis Roozeboom, while French high pressure research was booming. In 1904, a large grant from the Van der Waals Fund was used to buy a Schaeffer and Budenberg press, autoclave, and several deadweight gauges. The upper range was 5000 bar. The laboratory was a 7×7 -m room in the Physics Lab, plus a small corridor serving as a machine shop. The Van der Waals Fund began publishing around 1910. At that time, the Belgian scientist J. Timmermans, still well remembered as an expert in liquid mixtures, was a guest at the laboratory, and one of the earliest papers by Kohnstamm and Timmermans was on liquid miscibility up to 3000 bar [3].

Around 1915, a new development occurred. Kohnstamm and Walstra constructed an Amagat-style glass piezometer with platinum contacts, suitable for pressures up to 3000 bar. They measured *PVT* data for hydrogen around room temperature. This is the beginning of a method of *PVT* measurement which the Van der Waals Laboratory would practice throughout the 20th century.

The Bakhuis Roozeboom student, F. E. C. Scheffer (1883–1954), worked as an assistant at the laboratory. He studied three-phase equilibria in Cailletet tubes. Scheffer later became a professor at the Technical University of Delft. At Delft, studies of phase equilibria by means of Scheffer's method persist to the present day.

From 1915 to 1922, no publications resulted from the Van der Waals Fund. Although The Netherlands was not involved in the First World War, students did have to serve in the military, so graduate students were scarce. Kohnstamm's interest began to shift from experimental physics to pedagogy, to which he would devote the later part of his life. Worse, however, was the general belief that Van der Waals' work on molecular science was outdated. The available students flocked toward the rising star, the Amsterdam physics professor Pieter Zeeman (1865–1946), who became a Nobel laureate as early as 1902, not yet 40 years old, and 8 years before Van der Waals was thus honored.

Two events are worth noting. In 1911, an instrument maker trained by Kamerlingh Onnes joined the laboratory. His name was J. Ph. Wassenaar, and he would become responsible for the machine shop that made the Van der Waals Laboratory unique during much of the 20th century. And A. M. J. F. (Antonius; nickname, Teun) Michels (1891–1969), destined to lead the Van der Waals Laboratory to fame, began his math and science studies at the University of Amsterdam, earning his way by tutoring high-school students.

3. HISTORY OF THE MICHELS YEARS: 1920–1960

3.1. From World War I to 1930

Michels soon became associated with Kohnstamm, but his studies were interrupted by his military service. Around 1919, he resumed his studies and laboratory work in Amsterdam, while supporting himself as a part-time high-school teacher and tutor. In the early 1920s, he began to put his considerable energy, plus an appreciable part of his own earnings, into rebuilding the moribund laboratory. He made a major improvement in the design and operation of the bottom-loaded differential pressure gauge [4] and began to build a collection of differential pistons suitable for the various pressure ranges, up to the maximum of 3000 bar. He quickly sensed that the lab could not be put on a solid footing without proper funding for his first priority: a pressure calibration facility.

3.2. Westertoren Project

Unlike the larger European nations, The Netherlands had never had a standards laboratory. Thus, Michels had no convenient way of getting the deadweight gauges of his design calibrated, resulting in a wide gap between the “precision” and the “accuracy” of pressure measurement. While temporarily working for Kamerlingh Onnes 1 day a week, Michels

convinced the Nobel laureate of the necessity of a pressure calibration facility. In 1925, Kamerlingh Onnes embarked on a spirited fund-raising drive advertised in the newspapers. The drive yielded a considerable sum for the Van der Waals Fund. Several Dutch companies then constructing pressure equipment did contribute. Michels used the funding for one of his more colorful projects. Following the example of Amagat, he would erect a tall mercury column. For lack of mine shafts in a city built on mud, Michels decided to use one of Amsterdam's architectural assets: a tall church spire, the 17th-century Westertoren (Fig. 1). For years hence, this tower would house a 27.5-m-tall mercury column, the only pressure standard in the world. It was used to calibrate the effective area of low-pressure differential pistons up to the 30-bar range. Once the differential area of such a piston gauge was calibrated, it was cross-floated with a

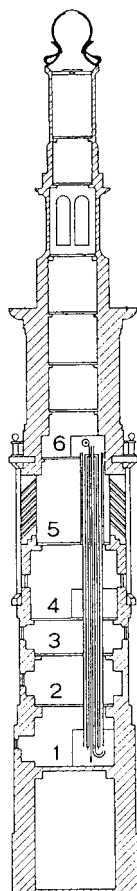


Fig. 1. From the 1920s until well after the Second World War, the 17th-century Amsterdam Westertoren housed a 27.5-m-high mercury column, spanning six floors in the tower. It was used as a pressure standard.

piston suitable for the next-higher pressure range, and thus, the effective differential areas of many differential pressure gauges were obtained for ranges up to 3000 bar. The sensitivity of the Michels-design piston gauges was exquisite: loaded with a maximum mass of over 250 kg, they could still sense a load change of a few grams. In the low-pressure range, the goal of 1 part in 10,000 uncertainty was attained. Due to error accumulation in the step-up to the high-pressure range, where, in addition, the effective area became pressure dependent, the accuracy must have deteriorated as the pressure increased to the 3000-bar limit.

3.3. New Direction

Michels obtained his Ph.D. with Kohnstamm in 1924. His thesis topic was the accurate measurement of PV isotherms. In 1928, Kohnstamm took a leave of absence, and Michels obtained a full-time position as a lecturer at the University of Amsterdam. High-school teaching days were over. The new course Michels set for the laboratory was as follows.

- A substantial increase in the accuracy of all physical variable measurements
- Large pressure and temperature ranges
- *De facto* deemphasis of phase equilibria studies in mixtures
- Emphasis on molecular science
- A broad spectrum of physical properties of interest

3.4. The Entrepreneur

In the late 1920s, Michels' reputation as an expert in high pressure came to the attention of Imperial Chemical Industries (ICI), which was then engaged in exploring pressure as a medium for new chemistry. They hired Michels as a high-pressure consultant. Company engineers would be sent to Amsterdam for training and for obtaining advanced degrees. The company was unwilling to fund Michels through the university administration. A creative legal solution was found: the Boyle Foundation was established to receive industry support for instrumentation of the Van der Waals Laboratory. Through this foundation, the laboratory quickly became one of the best equipped in the world.

ICI was working on the polymerization of ethylene. The story is that in the early 1930s, in the course of PV measurements on ethylene at the Van der Waals Laboratory, the piezometer lost pressure. When the glass blower cut the piezometer open, it contained a sticky residue that could not

be removed with chromic acid: the first sample of polyethylene. ICI's manufacturing of polyethylene contributed to the outcome of the Second World War: its low dielectric loss at high frequencies enabled the development of centimeter-wavelength radar by the British, who were then able to monitor the launching of planes and missiles from Germany.

The pressure-generating capabilities became prototypes for the design of high-pressure chemical plants in various countries. Also, collaborative ventures with Dutch companies resulted for construction of the necessary equipment. After the Second World War, one such Dutch Company, 't Hart, began to sell Michels-designed pressure equipment to the U.S. Navy, to American universities, and to companies around the world. As a university lecturer, Michels was breaking new ground. Although consulting with industry was common at the Technical University in Delft, at venerable academic institutions such as the University of Amsterdam, such practices were unheard of.

In 1930, Kohnstamm retired and Michels succeeded him. His position remained that of a university lecturer, and it would not be until 1939 that the university considered him worthy of a professorship. By that time, Michels had earned a solid reputation abroad and had found the means to build a first-rate high-pressure laboratory, with no more than lukewarm support from the university.

3.5. A Building of Its Own

By 1930, the laboratory had outgrown the wooden barracks to which it had temporarily moved from cramped space in the Physics Laboratory, and Michels started planning for a new building. The Amsterdam University was in the process of designing buildings to house the geology and the chemistry departments. Between the two buildings, room for an additional wing was available, and Michels did not waste any time claiming it. The new facility, bearing the name of Van der Waals, was occupied in 1935 and remained in use through the mid-1960s. The four-story building had a suite of temperature-controlled vibration-free basement rooms and an additional floor of experimental rooms filled with pressure generating and measuring equipment. Emulating Kamerlingh Onnes' instrument-makers school, Michels and Wassenaar designed a fully equipped machine shop, in which all pressure equipment needed by the laboratory was constructed by a group of several dozen trained machinists and apprentices. The building contained a glass shop, a chemical preparation and purification laboratory, a hazards lab for pressure testing, and, later, an air liquefaction facility. There was a library filled with the major journals, supplied by Michels' personal subscriptions, while the meager university contribution was used for

books only. A standards-quality metrology laboratory was available for calibration of platinum thermometers, precision length measurement, and precision mass calibration of the 25-kg weights carried by the “deadweight” gauges. The new facility was undoubtedly the best-equipped high-pressure laboratory in the world.

In 1947, Michels established a nonprofit foundation for calibration and precision measurement in industry. It was funded in part through an industrial consortium and in part by the government. Meetinstituut occupied several of the basement rooms, and it specialized in nonroutine precision length measurements and pressure calibration for industry.

Throughout his tenure, Michels, with his strong interest in molecular science, sought the advice and collaboration of molecular physics theorists. The Amsterdam professor of theoretical physics, Jan de Boer, had obtained his Ph.D. with Michels. The authors of the De Groot and Mazur classic on nonequilibrium thermodynamics were weekly visitors at the laboratory in the 1950s. The recent physics Nobel prize winner, Martinus Veltman, was a teaching assistant to Michels for a while during his student years.

3.6. The American Connection

Right after the Second World War, Michels, as a colonel for scientific and technical intelligence of the Dutch Army, began to serve on defense-related missions to the United States. He used this opportunity to broaden his scientific contacts there, and traveled to the United States several times a year during the period of 1946 to 1965. He visited numerous companies, such as Dupont, Dow Chemical, Phillips Oil, Corning Glass, and Norelco. He lectured at many universities on his high-pressure research and conversed with famous statistical physicists such as Margenau, Montroll, Onsager, and Slater. He returned home scientifically invigorated, with industrial support for his laboratory, with samples of new materials not yet available in Europe, and with scientific contacts that brought many American visitors to the Van der Waals Laboratory. Support from U.S. industry enabled him swiftly to re-equip his laboratory after the Second World War and set it on a course of growth and high productivity.

Around 1950, the Office of Naval Research and the Naval Ordnance Laboratory of the American Navy, much concerned about possible lags of American high-pressure science, approached Michels, with the request to build a copy of the Van der Waals Laboratory in the United States. Michels accepted this challenge. The new facility was built at the University of Maryland in College Park. At groundbreaking in 1953, the first shovel of dirt was dug by the physicist J. D. Van der Waals, Jr., the son and one of the successors of Van der Waals. The facility was opened in 1954.

Presently, it is part of the Institute of Physical Science and Technology at the University of Maryland. It specializes in theoretical statistical physics and contains experimental light scattering facilities.

A steady stream of American visitors to the Van der Waals Laboratory originated from U.S. Navy laboratories, as well as from universities. They came to receive training in high-pressure techniques, to obtain advanced degrees, or to prepare themselves for work at the new facility in College Park, Maryland. The Van der Waals Laboratory thus assumed an international atmosphere long before the globalization of science penetrated other academic institutions in The Netherlands.

4. WORK IN THE MICHELS YEARS

The Van der Waals Laboratory became a unique source of reliable thermophysical property data for fluids by combining a large pressure range, a high level of experimental accuracy, a wide scope of properties studied, and a well-deserved reputation for reliability of its work. An overview of the most significant work follows. Since full referencing of over 300 publications and Ph.D. theses is clearly impossible, the author has severely and arbitrarily limited referencing. Virtually all of the experimental data on thermodynamic and transport properties have found their way into existing databases, in which the references can be found.

4.1. *PVT* Data for One-Component Fluids Above 0°C

Throughout the Michels years and beyond, measurement of *PV* isotherms of gases were carried out in Amagat-type glass piezometers enclosed in autoclaves. The typical experimental range was from 0 to 150°C in temperature, and up to 3000 bar in pressure, as set by Amagat. The target uncertainty was 1 part in 10,000 in both pressure and density. Due to the accumulation of error during calibration, this target was not reached at the higher pressures. Nevertheless, the final uncertainty in the density, involving both pressure and volume calibration uncertainties, was usually well below 1 part in 1000 over much of the range. The sensitivity of the temperature measurement was 1 mK, and the temperature gradients in the piezometer probably no larger than 0.01 K, at least for the lower half of the temperature range. In the Michels years, the laboratory built its own platinum thermometers and calibrated them in a water triple-point cell of Michels' own invention in 1927 [5], long before this fixed point became internationally accepted. For the rest of the temperature range, the thermometers were calibrated on the absolute temperature scale by extrapolating the *PV* data to zero density. It was believed that this could be done

Table I. Fluids Studies in Glass Piezometers Above 0°C

Substance	Year(s)
Hydrogen	1915, 1932, 1941, 1959
Nitrogen	1928, 1934
Carbon dioxide	1930, 1935
Carbon dioxide	1937
Ethene	1936, 1942, 1976
Methane	1935, 1979
Deuterium	1941, 1959
Helium	1941
Argon	1949
Carbon monoxide	1952
Methyl fluoride	1952
Propene	1953
Oxygen	1954
Xenon	1954
Ethane	1954
Neon	1960
Krypton	1964

with an uncertainty of the order of 0.01 K. Table I contains a list of fluids studied during the Michels years and beyond, together with the year of publication of each of the experimental data sets.

Although it is impossible to describe individual publications here, an exception may be made for carbon dioxide. The data, obtained in an Amagat-style piezometer, extended Andrews measurements in large measure, with unsurpassed accuracy. The 1937 paper [6] reported on the critical region in great detail. These data were obtained by weighing mercury displaced by volume variation of a sample of gas confined to a steel vessel. This work was based on the Ph.D. research of Michels' wife, C. Michels-Veraart (Fig. 2), whose Ph.D. adviser was J. D. van der Waals, Jr., the son of Van der Waals. The Van der Waals Laboratory was an exception at the Amsterdam Physics Department in its hospitality to women graduate students and to female technicians.

In general, the publication of the experimental data was followed by a calculation of thermodynamic properties derived from the *PVT* data by numerical and graphical differentiation and integration. Computational experts, employed by the laboratory and outfitted with hand-cranked (later, electrically powered) calculators, thought nothing of doing least-squares fits to *PVT* data by means of up to sixth-degree polynomials in density. A full calculation of thermodynamic properties of a substance over the entire experimental range might take such an expert as long as a full



Fig. 2. Dr. C. Michels-Veraart joins her husband, Prof. A. Michels, in front of the laboratory he built at the University of Maryland in College Park. The picture dates from around 1954.

year. Figure 3 shows the pronounced maximum in the isochoric heat capacity, derived from the PVT data for CO_2 in 1937. We now know that this was an indication of a weak divergence, whose measurement by Voronel and co-workers 25 years later initiated the rebirth of interest in criticality of fluids.

These numerical representations of the experimental data became the principal source of experimental second and third virial coefficients. The virials, in turn, yielded accurate information on intermolecular forces, as envisioned by the Founder.

4.2. PVT_x and Phase Equilibrium Data for Mixtures

At various times, data were obtained for mixtures at one or a few fixed compositions in the same piezometers as used for one-component fluids. We mention the system methane–ethane, of interest to the gas industry

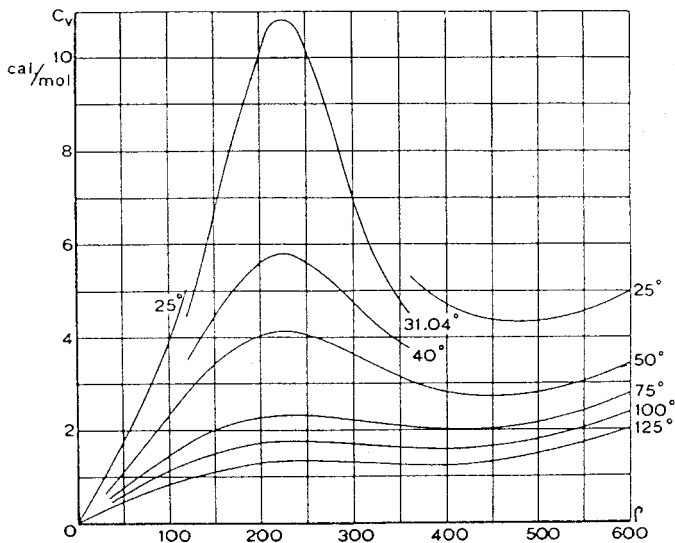


Fig. 3. The heat capacity C_V of CO_2 , calculated from the *PVT* data in 1937, shows a pronounced maximum in the supercritical regime. The horizontal axis shows the density, with the density at 0°C and 1 atm as the unit. The critical density of CO_2 equals 237 in these units. 1 cal = 4.184 J.

(1939), ammonia–nitrogen–hydrogen (1949, 1951), related to ammonia synthesis at the Dutch State Mines (DSM), and air (1955).

In the Michels years, phase equilibrium studies played a surprisingly small role. The methanol–hydrogen system was studied in 1953, and three ammonia systems, those with nitrogen and hydrogen, with krypton, and with argon, were studied between 1959 and 1961. An autoclave provided with sampling valves was used.

4.3. Vapor Pressures and *PVT* Data Below 0°C

After the Second World War, an expansion of the temperature range was initiated under the direction of Tom Wassenaar, son of J. Ph. Wassenaar. For this application, a steel vessel was built, which was inserted in a cryostat filled with a stirred, temperature-controlled, low-melting-point organic fluid such as isopentane and connected by a fine capillary to a differential pressure gauge, a membrane transducer, maintained at room temperature. This was not an absolute instrument: existing *PVT* data at 0°C and above were used to calculate both the fill density and the fraction of the sample present in the transducer. By means of liquid nitrogen as a

coolant, temperatures as low as -170°C were reached. The pressure range was up to 1000 bar. The target uncertainty was again 1 part in 10,000 in pressure and density, but some degradation occurred due to the indirect way of obtaining the density and due to a decrease in the sensitivity of the transducer when the fluid studied was in a range of low compressibility.

In the early 1950s, the apparatus was used for measuring vapor pressures of argon, krypton, carbon monoxide, and nitrogen. After this, *PVT* data were obtained for air, argon [7], hydrogen, and deuterium.

4.4. Transport Properties

4.4.1. Viscosity

The decision to include transport property measurements in the program of the laboratory was made by Michels at a fairly early stage. The first industrial associate from ICI, R. O. Gibson, obtained his Ph.D. in 1931 in Amsterdam on viscosity measurements of nitrogen up to 1000 bar [8]. The apparatus (Fig. 4) was a glass capillary of carefully calibrated diameter. It was attached to a reservoir at the bottom. The reservoir was provided with four platinum contacts at different heights. The capillary-reservoir assemblage was mounted in parallel to a wider glass tube, and the system was enclosed in a pressure vessel. All volumes between the platinum contacts were calibrated using known *PVT* relations for the gas. After pressurizing the gas by pumping mercury into the bottom of the viscometer, the mercury would rise quickly in the wide tube, thus pressurizing the gas in the reservoir below the capillary and establishing a pressure differential across the capillary. The gas then transpired through the capillary upward from the reservoir. The time it took for the mercury to travel from one platinum contact to the next one up was a measure of the fluid viscosity. The range of the apparatus was from 0 to 75°C and up to 2000 bar. The density of the fluid followed from the measured average pressure and temperature. For fluids in regions of moderate compressibility, where the pressure gradient does not affect the density unduly, the method yielded viscosities accurate to within a few tenths of a percent. For pressurized gases, this was at the time the best instrument available in the world. After the Second World War, viscosities were measured for hydrogen and deuterium (1953), carbon dioxide (1957), neon (1964), and krypton (1965).

4.4.2. Thermal Conductivity

Rather late in the Michels years, the decision was made to build a thermal conductivity apparatus. The intended range was 0 to 75°C , at pressures up to 2600 bar. The apparatus was of the parallel-plate type, with

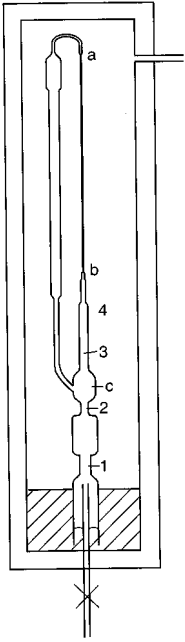


Fig. 4. A high-pressure capillary viscometer [5] was operational in 1931. The capillary is indicated by a and b. Platinum contacts are numbered 1–4.

adjustable spacings down to less than 1 mm and a guard ring ensuring a linear profile of the gradient. The top plate was heated, so that, at least in fluids of low compressibility, a stationary state was obtained free from convection. Near a critical point the tendency toward convection increases enormously, and it took graduate student J. V. (Jan) Sengers several rounds of reconstruction of the apparatus to eliminate convection to within 0.1 K from the critical point in carbon dioxide. This effort paid off: in 1962, a critical anomaly (Fig. 5) was reported in the thermal conductivity [9] of a type different from that predicted by the generally accepted Van Hove theory. This discovery led to the modern theory of critical dynamics, the mode-mode coupling theory, which has since been verified for a multitude of long-range dynamic fluctuation phenomena.

4.5. Other Properties

In the mid-1930s, an interferometer suitable for refractive index measurements between 0 and 75°C, up to 2600 bar, was built. It was used for the fluids carbon dioxide (1937), ethylene (1947), nitrogen (1947), and argon (1949). For these fluids, the equation of state had been measured earlier. This was the first time the weak density dependence of the Lorentz–Lorentz function was studied in detail over a large density range.

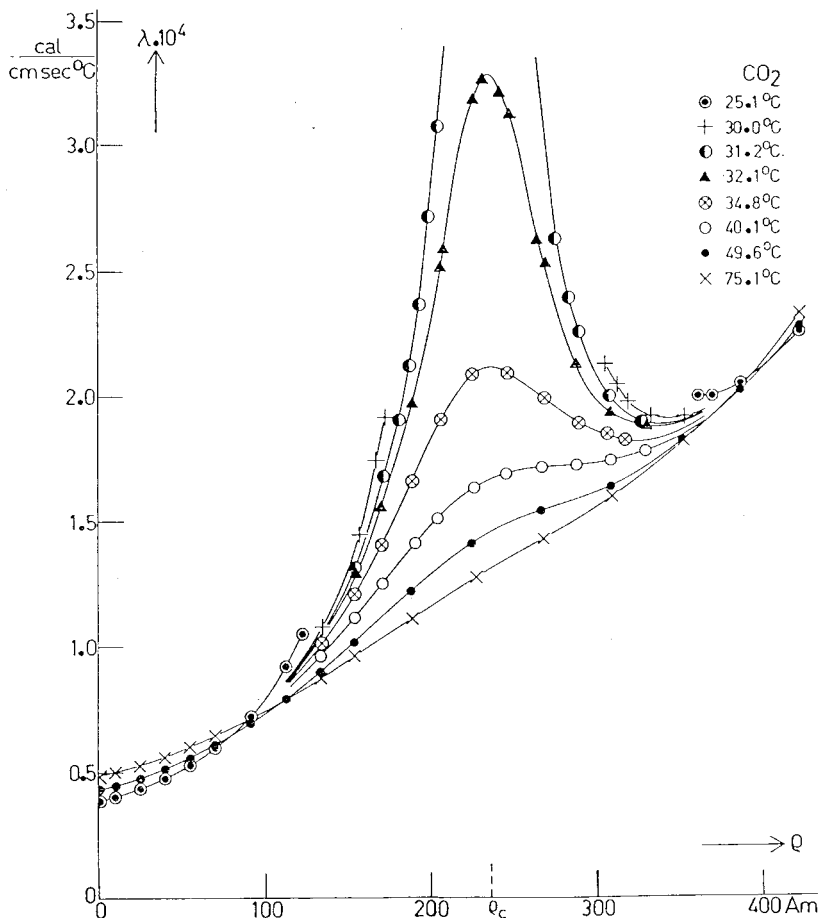


Fig. 5. The thermal conductivity of CO₂ along near-critical isotherms as a function of the density shows a critical anomaly of only about half the strength originally predicted by the Van Hove theory [6]. The density unit is as in Fig. 3. 1 cal = 4.184 J.

In the early 1930s, work began on measurement of the dielectric constant of compressed gases. The experimental range was from 0 to 150°C, at pressures up to 3000 bar. In the 1930s, data were obtained for hydrogen (1934), nitrogen (1935), and carbon dioxide [10]. After the Second World War, data were taken for argon. The Van der Waals laboratory thus was the first to explore the density dependence of the dielectric constant and to demonstrate the weak variation of the Clausius–Mossotti relation in supercritical fluids from vapor-like to liquid-like densities.

In the mid-1950s, the laboratory branched out toward studying optical phenomena induced by pressure. At that time, the theories of pressure broadening of spectral lines and of pressure-induced absorption were being developed at the Institute for Theoretical Physics in Amsterdam and elsewhere. The time was ripe for experimental verification of this new branch of molecular physics: the influence of Van der Waals forces on optical absorption [11]. As usual, the range of the apparatus was chosen to be from 0 to 150°C, at pressures up to 3000 bar. Initially, the effect of various pressurized gases on a mercury absorption line was measured. Later, evidence was found for weakly bound mercury-inert gas compounds [12].

To conclude the overview of the Michels years, I mention a body of measurements of the melting curves of the noble gases [13] and an interesting study of the adsorption of nitrogen on alumina at high pressures [14].

5. NEW DIRECTIONS IN THE TRAPPENIERS YEARS

5.1. Nuclear Magnetic Resonance

Nestor J. Trappeniers, who had obtained his Ph.D. with J. Timmermans, one of the early guests at the Van der Waals Laboratory in the Kohnstamm years, succeeded Michels in 1961. A Professor of Physical Chemistry in Groningen, he brought to Amsterdam a large group of Groningen graduate students well versed in the technique of nuclear magnetic resonance. Large magnets began to appear in Amsterdam, and a new and unique capability was forged in the Van der Waals Laboratory, that of NMR at high pressures and, later, at low temperatures. The design capabilities of the shop were stretched to the limit, and creative solutions were found for competing requirements: close spacing of magnetic poles, access for probes of the magnetic field, heavy walls for pressure containment, and space for thermal insulation. The need was felt for cryogenic work well below the liquid nitrogen range, and facilities for hydrogen liquefaction were developed, soon to be followed by the use of liquid helium. Much of the NMR work has been reviewed by K. O. (Klaas) Prins [15]. The early measurements were those of self-diffusion in several fluids. The coefficient of self-diffusion of methane [16] showed an absence of a critical anomaly (Fig. 6). Then the emphasis shifted to the study of phase transitions in molecular solids, such as ethylene, ammonium chloride, and methane. The work in solid methane, at temperatures from 1.5 to 60 K, became a spectacular success when a number of high-pressure phases were detected in a pressure range up to 9 kbar, which owed their existence to quantum effects [17].

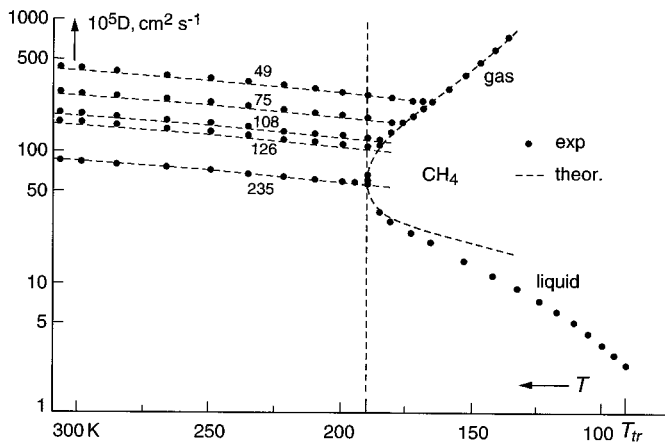


Fig. 6. The coefficient of self-diffusion of methane, measured by NMR, shows no critical anomaly [16]. Isochores are labeled in terms of the density ratio to that at 0°C and 1 atm. The critical density of methane is 226 in these units.

5.2. Expansion of the Pressure Range

Early in the Trappeniers years, the artificial barrier of 3000 bar, dating back to Amagat, was surmounted. In part, studies of solid phase transitions require much higher pressures, with less of a demand for ultraprecise pressure determination. On the other hand, the quest for high pressures was fueled by the studies of phase equilibria in mixtures, carried out by J. A. (Jan) Schouten [18]. The expansion of the pressure range occurred in two steps. More or less conventional means were used to reach pressures up to 15 kbar. To confine and characterize gaseous mixtures under such conditions is by no means conventional, however, and the Van der Waals Laboratory was one of the first to acquire this capability. The next major step was the use of the diamond anvil cell, and eye-catching results began to appear in the literature on fluid–solid phase equilibria obtained in systems with gas–gas phase separation. Some of this work is referred to in the paper by Schouten in these Proceedings. As a result of the new activities, the Van der Waals Laboratory enhanced its reputation as a premier high-pressure laboratory. The establishment and flourishing of the international AIRAPT conferences, initiated by Trappeniers in collaboration with Boris Vodar in France and E. Ulrich Franck in Germany, are testimony to the reputation and clout of the laboratory.

5.3. Other Topics

The optical work begun in the Michels years was expanded to include dynamic light scattering near the critical point and concentrated on the contributions of multiple scattering to the depolarization of light [19]. The Van der Waals laboratory continued to participate in microgravity experiments after the retirement of Trappeniers. A thermostat designed by A. C. (Teun) Michels was used by several other groups conducting experiments in space.

Since the 1980s, the laboratory has carried out a program, initiated by Samir Biswas, on accurate speed-of-sound measurements at pressures up to 10 kbar (1 GPa) and in the range of 100 to 350 K. Data were obtained in a combined *PVT*/speed-of-sound mode. Gases studied include helium [20], argon, methane, and nitrogen.

J. P. J. (Jan) Michels introduced molecular simulation at the Van der Waals Laboratory, as a complement to experimental studies of phase separation, transport properties, and Raman spectroscopy. His paper in these Proceedings shows the benefits of combining experiment and simulation for reaching an understanding of Raman line broadening and relaxation times in fluid mixtures at high pressures.

In collaboration with Jan Sengers at the University of Maryland, Hans van den Berg and colleagues began a study of thermal conductivity of fluid mixtures near the critical point. Recently, it was reported [21] that the thermal conductivity of an equimolar mixture of methane and ethane, though appearing to rise on approach to the critical line, levels off and remains finite, as anticipated by theory.

For more details on the work at the Van der Waals Laboratory in and after the Trappeniers years, the reader is referred to papers by Michels, Prins, Schouten, and Van der Gulik in these Proceedings.

6. CONCLUSION

Two forces propelled the Van der Waals Laboratory to its unique place in the world of thermophysical property measurement: the strong Dutch tradition of molecular science, and the singular energy, vision, engineering skill, and entrepreneurship of its director, Michels.

This is not the place to comment on the fate of the Van der Waals Laboratory after it took leave of its last director, Trappeniers, in 1987. Forces well beyond the control of the laboratory engulfed the physical sciences at the University of Amsterdam, and mergers and reorganizations resulted.

We should remember that when Michels began his career at the laboratory in 1920, he found it near its death. It took one visionary to revive the laboratory and launch it on a long and strikingly successful course spanning most of the 20th century. We may well gain hope when listening to the new voices now sounding from the Van der Waals–Zeeman laboratory, represented by Hijmans at this Symposium. There is no doubt that current work such as the Bose–Einstein phase transition in atomic hydrogen, and the trapping of light in photonic crystals, would have intrigued and delighted the two Nobel prize winners after whom the Laboratory is presently named.

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