

The discovery of superconductivity

Dirk van Delft and Peter Kes

A century ago Heike Kamerlingh Onnes set a new standard for physics research laboratories. But careless notebook entries have confused the story of his greatest discovery.

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On 10 July 1908, in his laboratory at Leiden University, the great Dutch physicist Heike Kamerlingh Onnes (1853–1926) experienced the most glorious moment of his career. That was the day he first liquefied helium and thus opened an entirely new chapter in low-temperature physics. (See the article in *PHYSICS TODAY*, March 2008, page 36.)

In a triumphant report to the Royal Netherlands Academy of Arts and Sciences (KNAW), Kamerlingh Onnes documented his achievement in great detail. Therefore it is remarkable that reliable details about his serendipitous discovery of superconductivity three years later have been hard to come by. Lack of information has led to speculations about the discovery. In particular, it has perpetuated an apocryphal tale about the role played by a sleepy young apprentice in Kamerlingh Onnes's lab. That tale was treated as established fact in a September 1996 *PHYSICS TODAY* article by Jacobus de Nobel (page 40). There have even been rumors of the possible disappearance of Kamerlingh Onnes's laboratory notebooks.

The lab notebooks

Enough reason, then, to have a close look at the Kamerlingh Onnes Archive, housed at the Boerhaave Museum in Leiden, to see whether any new clues could be found about the discovery of superconductivity—that most important consequence of the ability to reach liquid-helium temperatures.

Of course, it's roughly known when the first two superconductivity experiments were carried out. Kamerlingh Onnes's two earliest reports to the KNAW about zero resistance and "supraconductivity," as he preferred to call it, are dated 28 April and 27 May 1911. Figure 1 shows his laboratory at about that time.

According to the archive's inventory, two notebooks (numbers 56 and 57) should cover the period 1909–1912.¹ But on the cover of number 56 is written "1909–1910," and 57 begins with an entry dated 26 October 1911. So it does indeed seem as if a crucial notebook is missing. That would explain why so many speculations began to circulate.

Another obscuring factor is Kamerlingh Onnes's terrible handwriting. He wrote his lab notes, in pencil, in small household notebooks. They are very hard to read. After a few desperate hours trying, one tends to give up. And that's a pity because, the cover notwithstanding, notebook 56 does indeed announce the 1911 discovery of superconductivity (see figure 2). Translated, the entry reads, "The temperature measurement was successful. [The resistivity of] Mercury practically zero." A more literal rendering of the breezy Dutch tagline

Kwik nagenoeg nul would be "Quick[silver] near-enough null."

When Kamerlingh Onnes took lab notes, he always started by writing down the date. For that entry he wrote 8 April, but not the year! He dated the second experiment on the resistivity of mercury 23 May, again without giving the year. It gets worse: Between those dates, he and Albert Perrier, a visitor from Lausanne, performed an entirely different experiment on the paramagnetism of liquid and solid oxygen. For that experiment, the written date—19 May 1910—did specify a year, but the wrong one! It should have been 1911.

Why did Kamerlingh Onnes make that mistake? It's probably because an extensive series of similar experiments with Perrier had been carried out at the end of 1909 and during the first months of 1910. In any case, that little slip of the pencil has led many astray. It's the most likely reason that researchers exploring the archive have, until now, not looked more closely at the lab notes. Had they made the effort, they would have found the excitement over the first successful transfer of liquid helium to a separate cryostat for measuring resistivity, the exact dates of the first superconductivity experiments, who was involved, and what their roles were. The notes also reveal that some nice, oft-retold stories about those events will always remain nice but will never become true.

"Mercury practically zero." That penciled note heralded the birth of a new field. But probably at that moment Kamerlingh Onnes was simply thinking how right he had been to choose mercury. Zero resistance was what he expected to find in extremely pure metals at liquid-helium temperatures.² After he liquefied hydrogen in February 1906, he started a program to investigate the resistance of metals at low temperatures. He had a practical reason—thermometry. But he also had a purely scientific interest.

One of the issues in those days was the question of what happens to the resistivity of a metal as its temperature approaches absolute zero.³ It was accepted that electrons were responsible for electric conductance and that resistance was due to the scattering of electrons by the ions of the metal crystal. Would the scattering amplitude decrease fast enough with falling temperature to yield zero resistance at zero temperature? Or would the mobility of the electrons also diminish at lower temperature, thus resulting in zero *conductivity* at absolute zero? If nature followed the latter prescription—put forward by Lord Kelvin in 1902—the resistance of a pure metal would first fall with decreasing temperature, then bottom out at some minimum, and finally climb to infinity at absolute zero.

In the earliest investigations at liquid-hydrogen temperatures in Leiden, Kamerlingh Onnes and his assistant Jacob

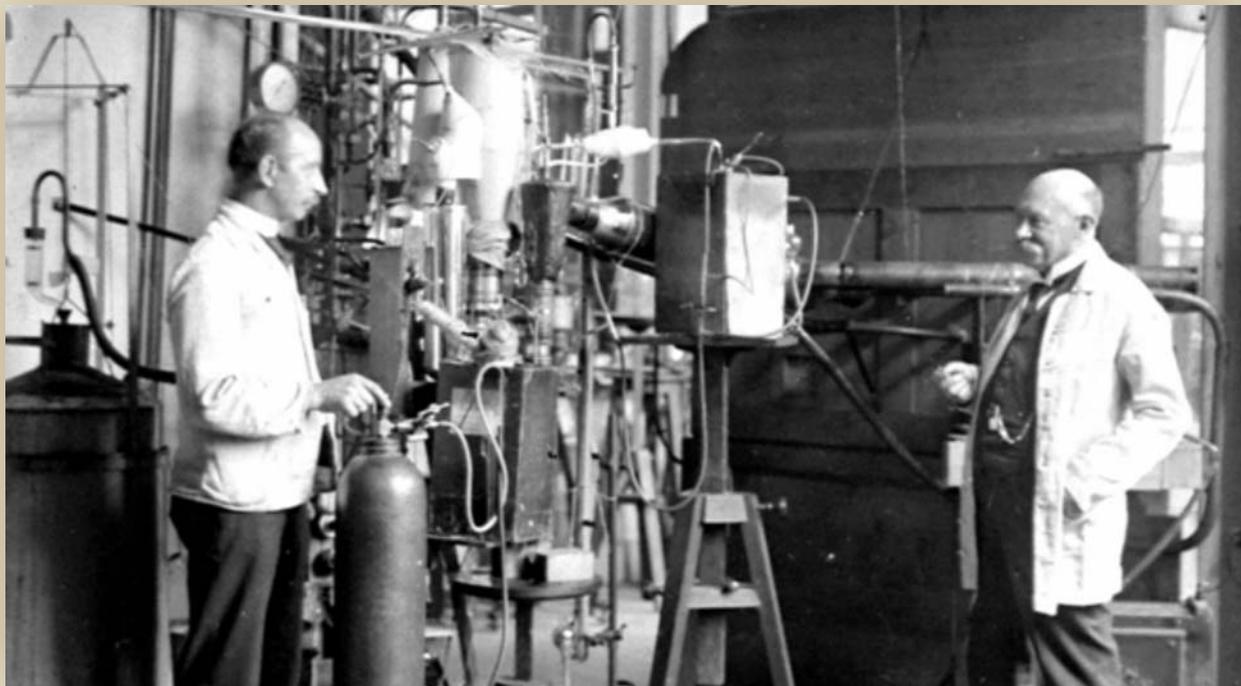


Figure 1. Heike Kamerlingh Onnes (right) and Gerrit Flim, his chief technician, at the helium liquefier in Kamerlingh Onnes's Leiden laboratory, circa 1911.

Clay studied resistance R versus temperature T in very thin gold and platinum wires.⁴ Before July 1908 the lowest available temperature was 14 K, at which solid hydrogen sublimates under reduced pressure. That was low enough to observe that the almost linear decrease of R with T at higher temperatures starts to level off to an almost constant value. In one of his KNAW reports, Kamerlingh Onnes even mentioned a trace of a minimum in the $R(T)$ plot, which indicates that he originally believed in Kelvin's model.

The almost linear $R(T)$ behavior of platinum above 14 K made that metal suitable as a secondary thermometer. It was much more convenient than the helium gas thermometer Kamerlingh Onnes had been using. But a disadvantage was the platinum thermometer's rather large size: 10 cm long and about 1 cm wide.

The resistance of the metal wires depended on the chemical and physical purity of the materials. For instance, Kamerlingh Onnes showed that the resistance increase due to adding small admixtures of silver to the purest available gold was temperature independent and proportional to the concentration of added silver. So, improving purity would yield metal wires of very low resistance that could serve as secondary thermometers at temperatures far below 14 K.

Those very low temperatures came within reach after the successful liquefaction of helium in July 1908. The next important requirement was transferring helium from the liquefier, which lacked adequate space for experiments, to a separate cryostat. In those days, accomplishing that transfer was a real challenge. Thanks to the notebooks, we can follow quite closely the strategy followed by Kamerlingh Onnes; his technical manager of the cryogenic laboratory, Gerrit Flim; and his master glassblower, Oskar Kesselring.

Experimenting in liquid helium

The first entry about the liquid-helium experiments in notebook 56 is dated 12 March 1910. It describes the first attempt

to transfer helium to a cryostat with a double-walled container and a smaller container connected to an impressive battery of vacuum pumps. "The plan is to transfer, then decrease pressure, then condense in inner glass, then pump with Burckhardt [pump down to a pressure of] 1/4 mm [Hg], then with Siemens pump [to] 0.1 mm."

Because there was nothing but glass inside the cryostat, the experiment worked well and a new low-temperature record was registered: roughly 1.1 K. The goal of the next experiment, four months later, was to continue measuring $R(T)$ for the platinum resistor that had previously been calibrated down to 14 K. But the experiment failed because the extra heat capacity of the resistor caused violent boiling and fast evaporation of the freshly transferred liquid helium. So it was decided to drastically change the transfer system. And that would take another nine months.

Meanwhile, interest in the low-temperature behavior of solids was growing rapidly. Specific-heat experiments carried out in Berlin and Leiden exhibited unexpected decreases with descending temperatures. For the first time, quantum phenomena were showing up at low temperature. Kamerlingh Onnes, playing with theoretical models himself, didn't want to wait until the new liquid-transfer system was ready. He decided to expand the original liquefier so that it could house a platinum resistor. Thus, on 2 December 1910, he made the first measurement of $R(T)$ for a metal at liquid-helium temperatures.⁵ Cornelis Dorsman assisted with the temperature measurements and student Gilles Holst operated the Wheatstone bridge with the galvanometer. That ultrasensitive setup for measuring the current was placed in a separate room, far from the thumping pumps.

The experiment's outcome was striking. The resistance of a platinum wire became constant below 4.25 K. There was no longer any doubt that Kelvin's theory was wrong. The resistivity had fallen to a residual value that presumably depended on the purity of the sample. Kamerlingh Onnes concluded that

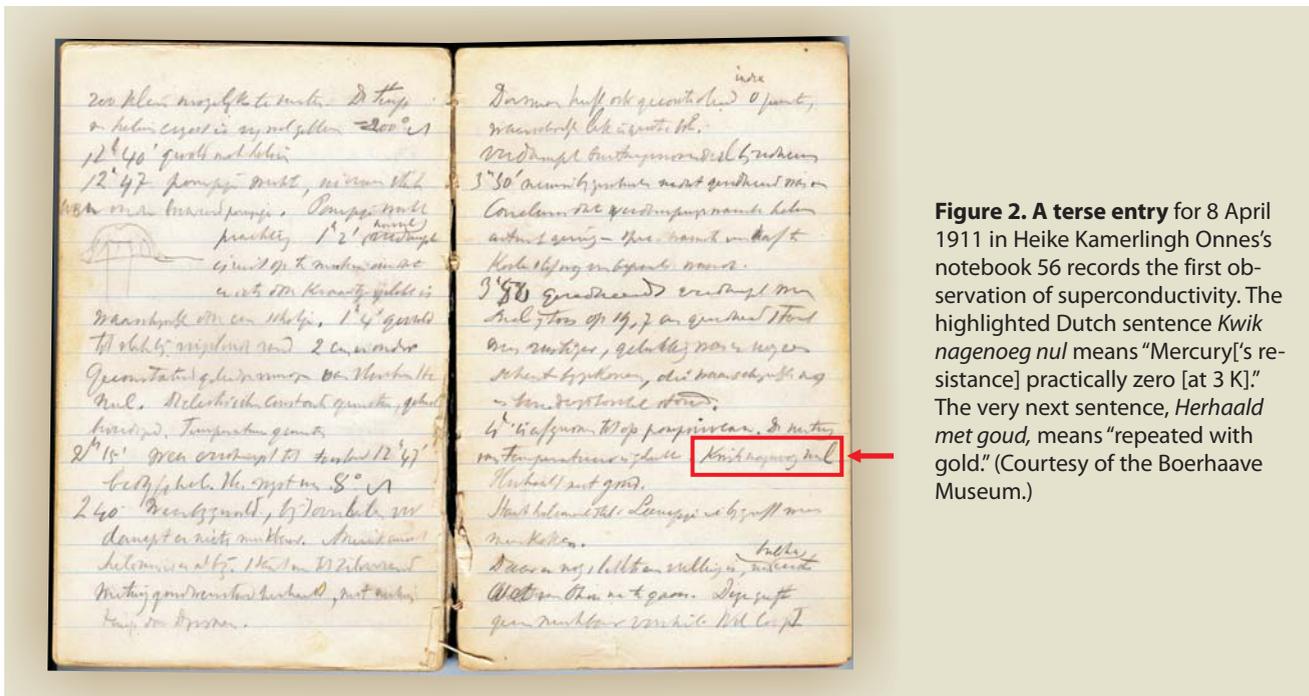


Figure 2. A terse entry for 8 April 1911 in Heike Kamerlingh Onnes's notebook 56 records the first observation of superconductivity. The highlighted Dutch sentence *Kwik nagenoeg nul* means "Mercury [s resistance] practically zero [at 3 K]." The very next sentence, *Herhaald met goud*, means "repeated with gold." (Courtesy of the Boerhaave Museum.)

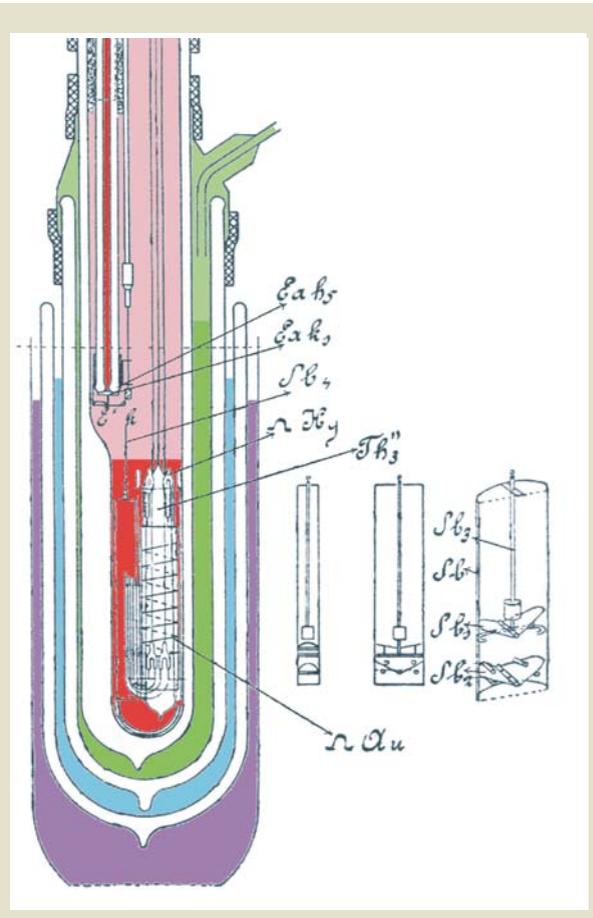
the resistance of sufficiently pure samples of platinum or gold should become zero at liquid-helium temperatures.

Why was he so eager to continue those investigations with mercury in particular? It must have been the large slope of the $R(T)$ curve for mercury at 14 K and the fact that, at that temperature, the slope showed no sign of leveling off. Furthermore, the Leiden lab had a lot of experience with the purification of mercury by distillation, and the material would not be contaminated by the necessity of drawing a thin wire. (The liquid mercury in a capillary simply freezes at 234 K [-39 °C].)

At the beginning of April 1911, the new cryostat was ready for its first cooldown. It was a masterpiece of technical design, demonstrating amazing levels of glassblowing skill and fine mechanical construction⁶ (see figure 3). The transfer tube had been replaced by a double-walled, vacuum-pumped glass siphon, externally cooled by a counterflow of liquid air forced through a copper capillary coil wound around it. The liquefier and the cryostat could be separated from each other simply by closing a valve. Another important innovation, necessary for establishing a well-defined temperature at the site of the measurement, was a stirrer connected to a magnet at the top of the cryostat that could be moved up and down by a motor. The action of valve and stirrer could be directly followed through uncoated strips in the silvered vacuum glass.

Figure 3. Bottom of the cryostat in which Heike Kamerlingh Onnes and coworkers carried out the 8 April 1911 experiment that first revealed superconductivity. The original drawing is from reference 6, but colors have been added to indicate various cryogenic fluids within the intricate dewar: alcohol (purple), liquid air (blue), liquid and gaseous hydrogen (dark and light green), and liquid and gaseous helium (dark and light red). Handwritten by Gerrit Flim are labels for the mercury and gold resistors (Ω Hg and Ω Au), the gas thermometer (Th_3), components at the end ($\mathcal{E}a$) of the transfer tube from the helium liquefier, and parts of the liquid-helium stirrer (Sb), which is also shown enlarged in several cross sections at right.

The main purpose of the 8 April 1911 experiment was to test the transfer of liquid helium to the experimental cryostat. But according to notebook 56, the helium gas thermometer and the gold and mercury resistors needed to perform the mercury-resistance measurements were already installed in



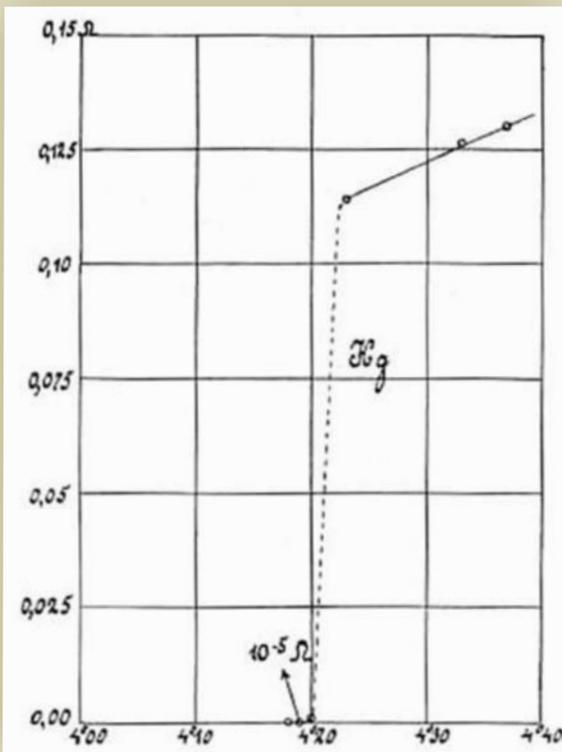


Figure 4. Historic plot of resistance (ohms) versus temperature (kelvin) for mercury from the 26 October 1911 experiment shows the superconducting transition at 4.20 K. Within 0.01 K, the resistance jumps from unmeasurably small (less than $10^{-6} \Omega$) to 0.1 Ω . (From ref. 9.)

the cryostat—just in case the helium transfer worked.

The mercury resistor was constructed by connecting seven U-shaped glass capillaries in series, each containing a small mercury reservoir to prevent the wire from breaking during cooldown. The electrical connections were made by four platinum feedthroughs with thin copper wires leading to the measuring equipment outside the cryostat. Kamerlingh Onnes followed young Holst's suggestion to solidify the mercury in the capillaries by cooling them with liquid nitrogen.

The first mercury experiment

To learn what happened on 8 April 1911, we just have to follow the notes in notebook 56. The experiment was started at 7am, and Kamerlingh Onnes arrived when helium circulation began at 11:20am. The resistance of the mercury fell with the falling temperature. After a half hour, the gold resistor was at 140 K, and soon after noon the gas thermometer denoted 5 K. The valve worked “very sensitively.” Half an hour later, enough liquid helium had been transferred to test the functioning of the stirrer and to measure the very small evaporation heat of helium.

The team established that the liquid helium did not conduct electricity, and they measured its dielectric constant. Holst made precise measurements of the resistances of mercury and gold at 4.3 K. Then the team started to reduce the vapor pressure of the helium, and it began to evaporate rapidly. They measured its specific heat and stopped at a vapor pressure of 197 mmHg (0.26 atmospheres), corresponding to about 3 K.

Exactly at 4pm, says the notebook, the resistances of the gold and mercury were determined again. The latter was, in

the historic entry, “practically zero.” The notebook further records that the helium level stood quite still.

The experiment continued into the late afternoon. At the end of the day, Kamerlingh Onnes finished with an intriguing notebook entry: “Dorsman [who had controlled and measured the temperatures] really had to hurry to make the observations.” The temperature had been surprisingly hard to control. “Just before the lowest temperature [about 1.8 K] was reached, the boiling suddenly stopped and was replaced by evaporation in which the liquid visibly shrank. So, a remarkably strong evaporation at the surface.” Without realizing it, the Leiden team had also observed the superfluid transition of liquid helium at 2.2 K. Two different quantum transitions had been seen for the first time, in one lab on one and the same day!

Three weeks later, Kamerlingh Onnes reported his results at the April meeting of the KNAW.⁷ For the resistance of ultrapure mercury, he told the audience, his model had yielded three predictions: (1) at 4.3 K the resistance should be much smaller than at 14 K, but still measurable with his equipment; (2) it should not yet be independent of temperature; and (3) at very low temperatures it should become zero within the limits of experimental accuracy. Those predictions, Kamerlingh Onnes concluded, had been completely confirmed by the experiment.

For the next experiment, on 23 May, the voltage resolution of the measurement system had been improved to about 30 nV. The ratio $R(T)/R_0$ at 3 K turned out to be less than 10^{-7} . (The normalizing parameter R_0 was the calculated resistance of crystalline mercury extrapolated to 0 °C.) And that astonishingly small upper sensitivity limit held when T was lowered to 1.5 K. The team, having explored temperatures from 4.3 K down to 3.0 K, then went back up to higher temperatures. The notebook entry in midafternoon reads: “At 4.00 [K] not yet anything to notice of rising resistance. At 4.05 [K] not yet either. At 4.12 [K] resistance begins to appear.”

That entry contradicts the oft-told anecdote about the key role of a “blue boy”—an apprentice from the instrument-maker's school Kamerlingh Onnes had founded. (The appellation refers to the blue uniforms the boys wore.) As the story goes, the blue boy's sleepy inattention that afternoon had let the helium boil, thus raising the mercury above its 4.2-K transition temperature and signaling the new state—by its reversion to normal conductivity—with a dramatic swing of the galvanometer.

The experiment was done with increasing rather than decreasing temperatures because that way the temperature changed slowly and the measurements could be done under more controlled conditions. Kamerlingh Onnes reported to the KNAW that slightly above 4.2 K the resistance was still found to be only $10^{-5}R_0$, but within the next 0.1 K it increased by a factor of almost 400.

Something new, puzzling, and useful

So abrupt an increase was very much faster than Kamerlingh Onnes's model could account for.⁸ He used the remainder of his report to explain how useful that abrupt vanishing of the electrical resistance could be. It is interesting that the day before Kamerlingh Onnes submitted that report, he wrote in his notebook that the team had checked whether “evacuating the apparatus influenced the connections of the wires by deforming the top [of the cryostat]. It is not the case.” Thus they ruled out inadvertent short circuits as the cause of the vanishing resistance.

That entry reveals how puzzled he was with the experimental results. Notebook 57 starts on 26 October 1911, “In

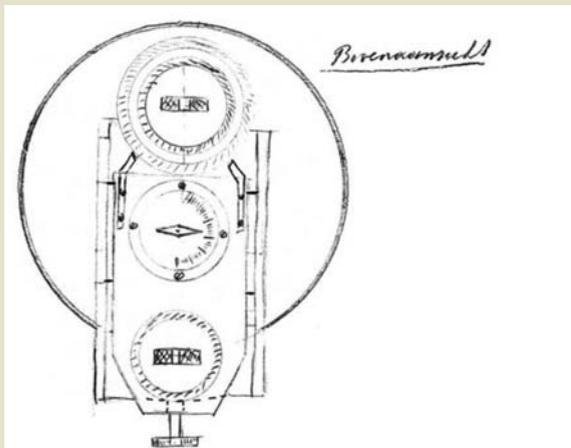


Figure 5. Gerrit Flim's drawing of the setup for a persistent-current experiment in May 1914. In this top view (*bovenaanzicht*), one sees a compass needle pointing north between a superconducting lead coil (west) immersed in liquid helium in a double-walled dewar and a normally conducting copper coil (east) of equal size immersed in liquid air in a single-walled vessel. The copper coil, whose connection to a current source and galvanometer is not shown, calibrates and monitors the persistent current in the superconducting coil. When both currents are equal, the compass points due north. (Courtesy of the Boerhaave Museum.)

helium apparatus, mercury resistor . . . with mercury contact leads." That is, the team had spent the whole summer replacing the platinum feedthroughs and copper leads by mercury wires in glass capillaries that went all the way through the cryostat's cap. And they studied how the new setup could be cooled down in a controlled fashion without breaking the mercury wires or the glass.

That was quite a challenge. In retrospect, the effort turned out to be a waste of time, but it was motivated by the important question of how small the resistance actually was. To improve voltage resolution, they sought to minimize the thermoelectric effect in the leads. The idea was to do that by using the same material for both sample and leads. It didn't work, because the transition from solid to liquid mercury actually turned out to be the source of a large thermoelectric voltage.

Still, the October experiment produced the historic plot, shown in figure 4, of the abrupt appearance of mercury's resistance at 4.20 K. The part of the plot above the transition temperature is of particular interest because it shows a gradual increase with temperature beyond the jump. To obtain those data, the Leiden team had to go beyond the normal boiling point of helium. They did that by closing the helium inlet valve so that the vapor pressure could rise and thus raise the boiling temperature.

From the sudden jump it was clear that a totally new and unexpected phenomenon had been discovered. Just one week later, Kamerlingh Onnes reported his discovery in Brussels to the elite of the physics world at the very first of the historic Solvay Conferences.⁹

Further on in notebook 57, we notice preparations in March 1912 for specific-heat experiments near the superconducting transition of mercury. Holst was put in charge. But there turned out to be experimental difficulties that couldn't satisfactorily be resolved. Eventually, Holst and Kamerlingh

Onnes published a paper together on the specific heat and thermal conduction of mercury, but the accuracy of the measurements at helium temperatures was not sufficient to reveal any features at the transition temperature.

A notebook entry dated 20 June 1912 is very interesting. "Discussed with Holst . . . alloying mercury with gold and Cd. Decided [to use] very small concentrations." A few days later, the experiment had already been carried out. But the results weren't published until the following March. In the notebook, Kamerlingh Onnes wrote early in 1913,

To my surprise, the resistance [of the mercury alloy] disappeared in the same way as with pure mercury; much of the time spent on the preparation of pure mercury . . . might therefore have been saved. . . . Even with the amalgam that's used for the backing of mirrors, the resistance was found 0 at helium temperatures. Later, Dec. 1912, it was found that it disappeared suddenly, as with the pure mercury, but at a higher temperature.

A few months earlier, the Leiden team had discovered that lead and tin were also superconductors, with transition temperatures near 6 K and 4 K, respectively.¹⁰ Those discoveries came as something of a relief. Now the team could do superconductivity experiments without worrying about laboratory problems peculiar to mercury: double distillation, broken threads, and very cumbersome cooldown procedures.

An experiment on 17 January 1914 revealed the destructive effect of magnetic fields on superconductivity. For lead, the critical field at 4.25 K was only 600 gauss.¹¹ That must have been a great disappointment for Kamerlingh Onnes. Before that sobering discovery, he had on several occasions dreamt aloud about coils made of superconducting material that could produce magnetic fields as high as a hundred thousand gauss (10 tesla). Nowadays the 8-tesla bending magnets of the Large Hadron Collider at CERN approach that dream, but they do so with a niobium-titanium alloy whose critical magnetic field far exceeds those of the superconductors known in Kamerlingh Onnes's time.

Persistent currents

Kamerlingh Onnes next concentrated on the question of how small the "microresidual" resistance actually was in the superconducting state. He designed an experiment to measure the decay time of a magnetically induced current in a closed superconducting loop—a small multiloop coil of lead wire cooled to 1.8 K. To probe the decay of the current circulating in the closed loop after the induction magnet had been removed, he used a compass needle placed close to the cryostat and precisely to its east. To calibrate the supercurrent, he positioned an almost identical copper coil on the other side of the compass (see figure 5).

Kamerlingh Onnes reported his first results to the KNAW on 24 April 1914:

During an hour, the current [0.6 A] was observed not to decrease perceptibly. . . . A coil cooled in liquid helium and provided with current at Leiden might, if kept immersed in liquid helium, be conveyed to a considerable distance and there be used to demonstrate the permanent-magnetic action of a superconductor carrying a current. I would have liked to show the phenomenon at this meeting in the same way I brought liquid hydrogen here in 1906. But the equipment at my disposal does not yet allow the transportation of liquid helium.¹²

Two decades later, such a traveling show had become possible. In 1932, six years after Kamerlingh Onnes's death, Flim flew to London with a portable dewar containing a lead ring immersed in liquid helium and carrying a persistent current of 200 A. He made the trip to demonstrate the most sensational effect of superconductivity at a traditional Friday evening lecture of the Royal Institution. That was the same august venue at which James Dewar had demonstrated the liquefaction of hydrogen in 1899.

The 1914 observation of a persistent current was the ultimate proof that superconductivity was indeed an entirely novel phenomenon. The excitement spread quickly. Paul Ehrenfest, who had witnessed the experiment, told Hendrik Lorentz that he was flabbergasted.

I attended a fascinating experiment at the laboratory. . . . Unsettling, to see the effect of this "permanent" current on a magnetic needle. It is almost palpable, the way the ring of electrons goes round and round and round in the wire, slowly and virtually without friction.¹³

Ehrenfest suggested to Kamerlingh Onnes that the experiment could equally well be performed with a lead ring in place of the multiloop coil. That worked perfectly. The Leiden team measured a persistent current of 320 A in a lead ring with a rectangular cross section of $3.0 \times 3.5 \text{ mm}^2$.

Kamerlingh Onnes correctly concluded that current density was an important limiting quantity. But the current density he computed, 30 A/mm^2 , was far too low. As we now know, supercurrents are confined within a thin surface layer a few hundred nanometers thick—the superconducting penetration depth introduced by theorists Fritz and Heinz London 20 years later.¹⁴

Another two decades would have to pass before John

Bardeen, Leon Cooper, and Robert Schrieffer produced the first satisfactory microscopic theory of superconductivity.¹⁵

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