

*Citation:*

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when it was found that the resistance of tin disappeared suddenly too, we succeeded in making a less doubtful experiment than is possible with mercury, with a flattened out constantan wire, which was covered with a thin layer of tin<sup>1)</sup>. The resistance of the layer of tin disappeared with a weak current and at a low temperature, while the constantan remains an ordinary conductor at that temperature.

Thus we may for the present adhere to the usual laws of current division, and in this extreme case continue to assume that in so far as the appearance of the potential difference is to be explained by a local heating in consequence of a local change in difference of the chemical nature of the conductor from pure mercury this disturbance must extend over the whole section of the current path. Thus the conclusion drawn in § 9 concerning the probability of the existence of a micro-resistance remains valid.

*(To be continued).*

**Physics.** — *“The radiation of Radium at the temperature of liquid hydrogen”*. By Madame P. CURIE and H. KAMERLINGH ONNES. Communication N<sup>o</sup>. 135 from the Physical Laboratory at Leiden.

One of the most remarkable peculiarities of radio-active substances, is that the radiation is independent of the temperature. Neither do the radio-active constants change with the temperature. These two facts are related to each other; they prove that the radio-active transformations are not affected by the influence of temperature, which plays such an important part in the chemical transformation of the molecules.

According to the theory of radio-active transformations, the intensity of radiation of a simple substance is proportional to the rapidity of the transformation, so that a change in one of these quantities involves a change in the other.

The experimental investigations of the influence of temperature have been concerned with the measurement of the radio-active constants and the intensity of radiation of certain substances. P. CURIE has shown that the law of transformation for the emanation does not change at a temperature of 450° C. nor at the temperature of liquid air<sup>2)</sup>. Various observers have proved that the penetrating

<sup>1)</sup> It is to be noted, however, that the current density in the thin layer had to be made very weak Comp the following part of this Communication VIII, § 16.

<sup>2)</sup> P. CURIE, C. R. 1903.

radiation of radium and uranium have the same value at ordinary temperature and at the temperature of liquid air <sup>1)</sup>. The influence of high temperatures on the radium emanation and its transformation products, particularly Radium C, has also been the subject of various investigations. The results have given rise to differences of opinion. Nevertheless it would seem to be justifiable to conclude that the dependence upon temperature which was observed in some cases must be attributed to secondary phenomena of less importance, and that the radioactive constants of the above substances are not appreciably altered when the temperature is raised to 1500° <sup>2)</sup>.

As the question is of great importance it was desirable to extend the results already obtained, by extending the experiments over a wider range of temperature and by increasing the accuracy of the measurements, which in the above mentioned investigations could not have been greater than 1% at the most.

Our object was to descend to the temperature of liquid hydrogen. By using a compensation method we were able to determine very slight changes in the radiation intensity. Our measurements were concerned with the penetrating radiation of radium. The results, within the limits of accuracy which may be placed at 0.1%, do not confirm the existence of a quickly acting influence upon the radiation, in consequence of this strong decrease of temperature.

The investigations were made in the first part of 1911. The preliminary measurements were partly made in Paris, and partly in Leiden, while the final measurements took place in Leiden in July 1911. We intend to continue and extend the experiments, which is the reason of the publication having been postponed. But as the continuation of the work has been prevented so far by the long indisposition of one of us, we thought it best not to wait any longer in publishing our results.

*Apparatus and arrangement of the measurements.* After some preparatory experiments we decided to use the following apparatus. The apparatus consists of a vacuum glass *A*, in which a copper vessel *B* is placed, which contains the low temperature bath. The vacuum glass, which is fairly wide at the top ( $a_1$ ) consists underneath of a tube-shaped portion, the length of which is about 16 cm. and the two diameters 8,5 and 13 mm. The copper vessel which fits into the vacuum glass, is also provided with a tube-

<sup>1)</sup> BECQUEREL, CURIE, DEWAR, RUTHERFORD.

<sup>2)</sup> CURIE and DANNE, C. R. 1904. BRONSON, Phil. Mag. 1906. MAKOWER and RUSS, Le Radium, 1907. ENGLER, Ann. d. Phys. 1908. SCHMIDT, Phys. Zeitschr. 1908.

shaped portion, which is shorter than that of the vacuum glass. This copper tube is closed underneath ( $B_1$ ) by a metal stopper  $C_1$  to which a tube  $C_2$  of thin aluminium is attached (thickness 0.3 mm.); this tube contains a sealed glass tube with the radium. The narrow space at the bottom of the vacuum glass in which this tube is placed, is cooled to a temperature that differs very little from that of the bath: the difference could hardly be established, when the copper vessel was filled with liquid air. This

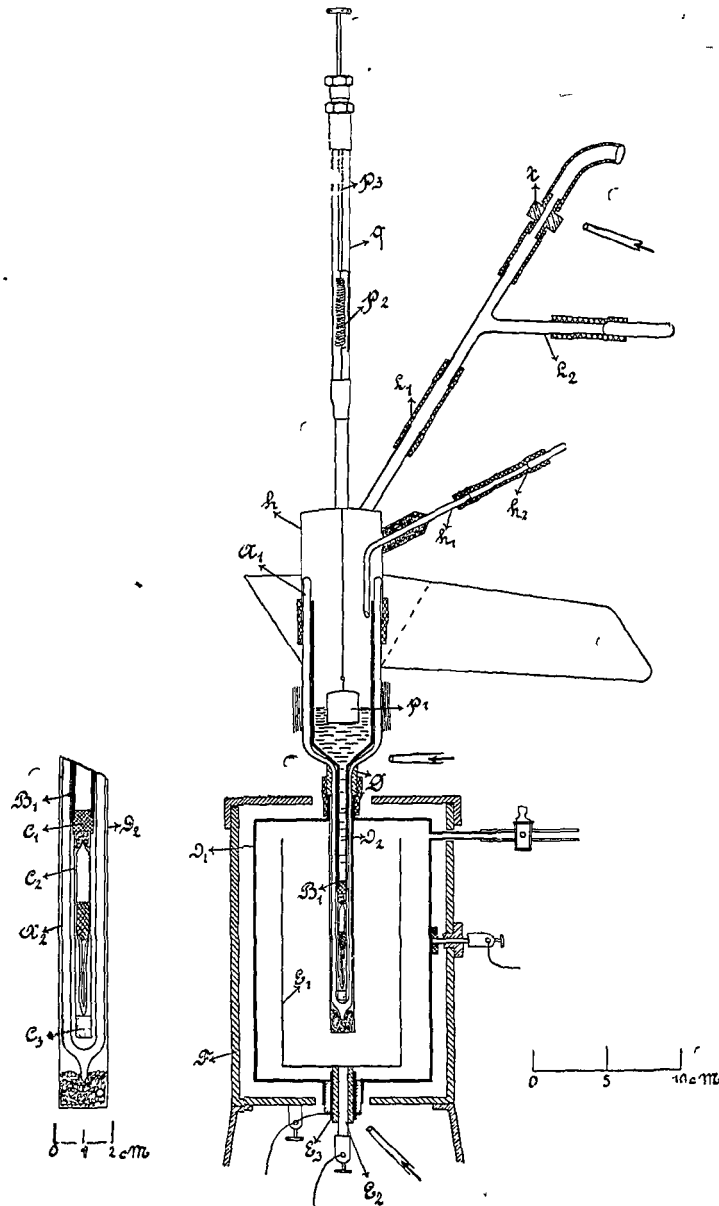


Fig. 1.

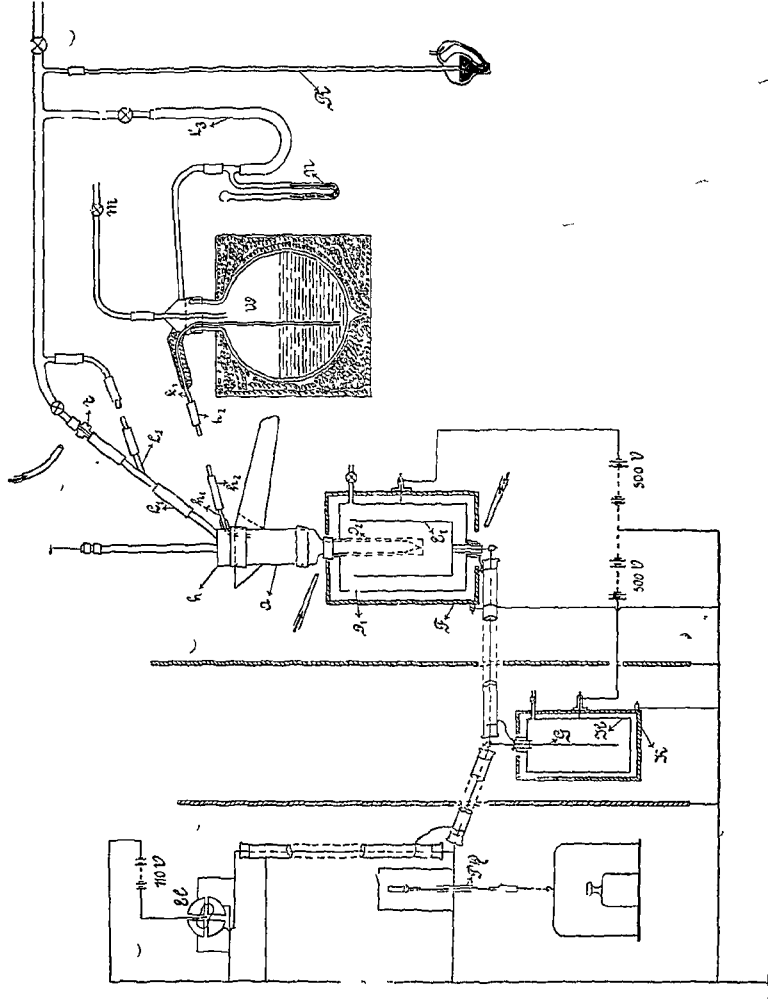
method of cooling seemed to us to be preferable to placing the tube itself into the liquid gas, which is always a little dangerous.

The rays that the radium in the tube sends out are partly of a penetrating nature. They go through the walls of the aluminium tube and those of the vacuum glass, and penetrate through a metal wall into the ionisation space. This consists of a cylindrical box  $D_1$ , which is connected to a battery; in the middle of the lid of this space a tube is soldered, which is closed at the lower end. The insulated electrode  $E_1$  which is a hollow cylinder, is connected with the electrometer. The metal case  $F$ , which is connected to earth, serves for electrostatic protection. When the apparatus is mounted the tube-shaped portion of the vacuum glass is inside the tube  $D_2$ , which is placed centrally in the box  $D_1$ , while it is closed by a thick piece of india-rubber tubing round a piece of amber  $G$  which is sealed to the vacuum glass. When the tube containing the radium is in its place, ions are formed on both sides of the electrode  $E_1$  in the air that fills the box  $D_1$ . The current that is taken up by this electrode is measured by an electrometer and a plate of piezoquartz.

The experiment consists in measuring the ionisation current generated by the rays of the radium: 1. when the radium is at the temperature of the room, and 2. when the radium is cooled to the temperature of liquid hydrogen. The ionisation chamber, which is outside the vacuum glass remains at about the temperature of the room. The chamber is airtight, and the quantity of gas that it contains does not alter during the experiments.

The accuracy of the measurements is greatly increased if instead of measuring the total current, a compensation method is used. This consists in compensating the current to be measured by a current in the opposite direction, which is generated in a second ionisation chamber by a tube containing radium, which is kept at constant temperature during the experiments. This current compensator is of a type which is greatly made use of in radioactive measurements. The insulated electrode  $G$  is in the form of a tube which is closed at the bottom; it is connected by means of copper wires (electrically protected in brass tubes filled with paraffin wax) with the electrometer and with the electrode  $E_1$ . This tube reaches into a cylindrical box  $H_1$  which is connected to a battery and which forms an ionisation chamber. The outside case  $K$  serves for electrostatic protection. The electrodes  $G$  and  $E_1$  are protected in the usual way by a protecting ring connected to earth. The tube  $G$  contains a sealed glass tube with radium salt. The boxes  $D_1$  and  $H$  are kept

at high potentials of opposite sign. Under these circumstances the difference of the two ionisation currents is measured which are generated in the two chambers. With sufficiently strong currents great accuracy can be attained in this way.



It is worth noticing, that the various small imperfections in the method of measuring, which are usually unnoticed, become apparent when the method described above is followed. E. g. when each current is measured separately, the saturation appears to be complete at a potential of about 500 volts. But when the difference between the currents was measured, which was usually under 5% of each current separately, it was found that the current under these circumstances increased with the voltage. When the potential

increases from 500 to 800 volts, the current increases by 2 to 3 thousandths. Constant potentials must therefore be used.

The accuracy is limited by the stability of the apparatus and by the oscillations in the radio-active radiation.

The investigations were made with radium salts in the solid state, contained in sealed glass tubes; the salt was finely granular, and the tubes were not quite filled. When they are shaken the grains can move to a certain extent, which causes a slight change in the distribution of the radiation inside the ionisation chamber. The danger of this is lessened by giving the grains a definite arrangement beforehand by tapping the tube. But in spite of this, small perturbations of this nature remained in our experiments of not more than 1 in 1000. The very greatest care is, therefore, necessary in the manipulations which must be made during the experiments.

The radio-active oscillations of the ionisation current become apparent when the sensitivity of the measurements is raised sufficiently. They cause irregular deviations which can only be eliminated by a great number of measurements. They are least to be feared when gamma rays are used, as was the case in our experiments. In our case they could not do any harm to the determinations.

It is important that the ionisation chambers should contain an unchangeable quantity of air. When working with penetrating rays, the current is approximately proportional to the amount of ionised air. If one wishes to keep the current constant with great accuracy, we must, therefore, take care that the ionisation chambers are properly closed. Each chamber is supplied with a tap. By changing the amount of air in the compensation chamber, the current in the chamber could be so regulated as to get a compensation of the amount required. Both the compensation chambers are filled with dry air by a tube filled with cotton wool, which can be connected to the tap of the chambers by a ground joint, and to an air pump and a manometer to regulate the supply.

We had to take very great precautions to prevent the cryogenic operations from causing insulation errors in consequence of the precipitation of moisture from the surrounding air on the strongly cooled parts of the apparatus. The cryogenic apparatus used by us enabled us to avoid all difficulties of this sort. This instrument, which was arranged for working easily and safely with liquid hydrogen, had moreover the advantage that the radium tube could only come into contact with the gaseous phase of the liquified gas, so that when this was hydrogen there was no fear of solid air being deposited on the tube.

The cryogene apparatus is completely closed. The vacuum glass has a lid  $h$  of thin new silver, which is fastened air tight to the glass by means of an indiarubber ring, so that when the radium tube is in its place, the apparatus can be evacuated, and can be filled beforehand with pure, dry gaseous hydrogen (by  $L$ ). A small hole in the stopper  $C_2$ , upon which the radium tube in the aluminium tube rests, ensures the pressure equilibrium, which establishes itself easily during these operations, so that the radium tube is not exposed to any danger.

The liquid hydrogen is poured into the vessel  $B$  through the new silver tube  $h_1$  and through the india-rubber tube  $h_2$ . For this purpose the glass stopper is removed which closes the india-rubber tube, after the tube with the stopper  $K_2$  has also been taken away, and the india-rubber tube is connected to the syphon  $K_1$  of the large vacuum glass  $W$ , containing the liquid hydrogen that has been previously prepared. Before the syphon and the india-rubber tube are connected, the apparatus and the vacuum glass are connected to a gasometer with pure hydrogen, by the tubes  $L_1$  and  $L_2$ . When the first mentioned connection has been made, the connection of the vacuum glass with the gasometer is broken, and the liquid hydrogen is poured into the apparatus by means of pressure from a cylinder with compressed hydrogen, admitted by the cork  $m$ , and controlled by the mercury manometer  $n$ . The supply-glass and the gasometer are then again connected. The syphon is taken off the inlet tube after the connection tube has been warmed, and this latter tube is immediately closed by a glass stopper.

To prevent these manipulations from shaking the apparatus, we made the indiarubber tube  $h_2$ , which is usually as short as possible, rather long; but as the great cold makes the india-rubber very brittle, and the breaking of it might cause great inconvenience, we used only a length of 7 c.m. In this way the shaking remained below the limits of stability in the apparatus which we used for these experiments. In a larger apparatus, intended for experiments that take longer, more than 24 hours, but with which we have only been able to make preliminary determinations so far, we were able to attain a greater amount of stability, and we were more independent of the shaking caused by the manipulations.

Care must be taken in filling the copper vessel  $B$ , that the liquid gas does not overflow, as it might penetrate into the cooling chamber, which would give rise to irregularities, and might injure the radium tube. On the other hand it is necessary to know when the liquid gas has evaporated, otherwise the experiments might be



continued without our being certain of the temperature. The height of the surface of the liquid gas can be read by means of a float. This consists of a new-silver box  $p_1$ , suspended from a weak spring  $p_2$ , which spring is attached to a rod  $p_3$ . This rod is movable in a packing tube, which is fastened to the upper end of a glass tube  $q$ , carried by the lid  $h$ . Beside the spring and also hanging from the rod, is a flat rod which is provided with a scale at its lower end. In consideration of the very small density of the liquid hydrogen ( $1/14$ ), the float is made very light. When the float reaches the surface of the liquid by the moving down of the spring, this is indicated by the shortening of the spring, and the height of the liquid can be read on the scale and on the rod.

Before pouring in the liquid gas, the float is regulated to the height to which the vessel is to be filled. Before beginning the measurements, the spring is pressed down as far as is necessary to make the lengthening of it show when the liquid is so far evaporated that the measurements must be stopped.

The evaporated hydrogen is carried off by  $L_1$ . The tube  $R$ , the extremity of which is placed in mercury, serves as a safety.

In order to be certain of the insulation of the vacuum glass, and to avoid currents which might be injurious to the constancy of the tension of the battery, a piece of amber is interposed in the tube  $L_1$ . To prevent the amber from being cooled too much by the filling, the cold vapours are carried off by a supplementary tube  $L_2$ , which is coupled off as soon as the filling is completed. When the evaporation of the bath has become stationary, a current of air a little warmer than that of the room directed upon the amber is sufficient to maintain the insulation. This current of air is given by a reservoir of compressed air, the air flows through a long tube, part of which is warmed by hot water.

The connection of the piece of amber,  $g$ , which is sealed to the vacuum glass, with the tube  $D_2$  of the principal ionisation chamber, is very carefully made, to insure an airtight closing, and thereby to prevent the possibility of moisture penetrating to the space between the tube and the vacuum glass. The currents of cold air that come down are kept away by a paper screen. The water that runs down the glass from the lid must also be disposed of. The very low temperature of the vapours inside the lid causes frost to settle on it during the filling, which thaws afterwards. After the filling is finished, the condensation of water vapour out of the air continues; the water thus formed, is absorbed by cotton wool above the paper screen we mentioned, and below it by filter-paper. A current of dry slightly

warmed air is directed upon the amber, which at the same time dries the lower part of the vacuum glass.

Finally, the cooling of the parts of the connection of the main electrode  $E_1$  with the electrometer must be prevented. To attain this a current of dry and slightly warmed air is also directed upon the amber stopper between the stem  $E_1$ , and the protecting ring  $E_3$  at the bottom of the main ionisation chamber.

The cold currents of air, which come down from the tubes that lead off the gases, are diverted from the apparatus by suitable screens, and large currents of air in the room are avoided as far as possible, so as to prevent the ionised air around the contacts from being displaced; these contacts were further protected by various lead protecting mantles (in the figure diagrammatically represented), by tin foil, etc. The influence of the warm currents of air already mentioned was tested at the temperature of the room: they did not cause any electrostatic phenomena.

#### *Preparatory Experiments.*

The experimental method was first studied in Paris, using liquid air as cooling bath.

The current in the main ionisation chamber was procured by using a tube with about 0.1 gr. of radium chloride. In the compensation chamber a tube with about 25 mgr. of radium chloride was used.

In the first experiments the first tube was contained in an aluminium tube with walls of 0.3 mm. thickness; the central tube  $D_2$  in the chamber  $D_1$  was also of aluminium, with walls of 0.5 mm. thickness. The rays, before penetrating into the ionisation chamber, passed through a layer of aluminium of about 0.8 mm. and moreover a glass layer about 2.5 mm. (wall of the radium tube and both walls of the vacuum glass).

During the cooling a diminution of the current in the main chamber could be observed. It was not very regular, and amounted to about 2%, it was perceptible immediately after the liquid air was poured into the copper vessel, and reached its maximum in about half an hour.

When, however, the liquid air was quickly taken out of the vessel, and the temperature of the radium tube was followed with a thermoelement, it could be observed that while the temperature of the radium tube was still constant, the strength of current already began to rise, and reached about its original value, by the time the whole apparatus had returned to ordinary temperature. From this it was evident that the decrease of strength of current which we observed

was not attributable to a change of radiation in the radium tube, but to some other cause.

Various test experiments seemed to show that it was caused by change in the power of absorption of the screens, due to their contraction at low temperatures. It was therefore necessary to make use of heavier and thicker screens, to make sure that we only worked with the most penetrating rays, which are less susceptible to phenomena of this kind. After the radium tube had been inclosed in a copper tube of 1 mm. thickness, we found that the decrease of current when the liquid air was poured in was reduced to 0.1%. The decrease was completed in 10 minutes. Three successive experiments gave this result.

We found that we could make the circumstances even more favourable, by changing the arrangement of the apparatus in such a way that the screens in which the absorption of the rays took place were not cooled at all. In order to do this, the radium tube was once more put into the aluminium tube of 0.3 mm., while the central tube  $D_2$  of the chamber  $D_1$  was replaced by a brass tube of 2 mm. wall thickness. The decrease of the current became by this means less than 1 in 1000. This arrangement was used in the final experiments.

#### *Final Experiments.*

The experiments were made in Leiden from July 20th to 25th 1911.

The ionisation current in the main ionisation chamber was 1100, expressed in arbitrary units (about 10 electrostatic units). The strength of the compensation current was so regulated that it was a little larger. The difference was at most 20 units, about 2% therefore. The rays used for the experiments were gamma rays.

We were able to make two experiments with liquid hydrogen. In the experiments the cold ionisation chamber, as we said above, was filled with dry gaseous hydrogen, and by this we made sure that no deposit could come on to the radium tube.

In the first experiment the current of originally 10.9 units, attained the value of 14.7 units after the pouring in of the liquid hydrogen, which took 15 minutes. This change corresponds to a change in the main current of 0.34%. In the second experiment the current measured had a strength of 18.3 units, and was very constant, the irregularities measured during an hour were less than 1/10000 of the main current. After the liquid hydrogen had been poured into the apparatus, measurements which agreed very well with each other gave for the value of the current during half an hour 18.5

units, and after an hour 18.2 units. We can thus assert that in this experiment, which was evidently conducted under very favourable circumstances the cooling had not caused a change in the main current of as much as 1 in 5000.

We made another experiment at the temperature of liquid oxygen. The current measured had a strength of 1.8 units. Measurements made during an hour at the temperature of liquid oxygen gave a value of 2.6 units for the current measured, which corresponds to a decrease of 0.7 in 1000 in the main current.

It would have been desirable to have made a greater number of experiments and to continue these during a greater length of time; nevertheless it would appear to be justifiable even now to state, that *cooling of radium down to the temperature of liquid hydrogen (about 20° 3 absolute) during a period of not more than 1½ hours does not cause a change in the gamma radiation of 1 in 1000 and probably not even of 1 in 5000.*

It is thus probable paying due regard to the degree of accuracy attained, that this decrease of temperature has no immediate or quickly discernable influence upon the emanation or the active deposits of short period (radium *A*, *B* and *C*). But in these experiments there was no opportunity for detecting an eventual effect upon the radium itself, or a slowly developing effect upon its evolution products.

#### *Experiments with polonium.*

A few preliminary experiments on the influence of low temperatures upon the radiation of polonium have been made in Paris. The experiment which was made only with liquid air, gives rise to some difficulties. A plate on which was some deposit of polonium was placed at the bottom of a long glass tube, which could be immersed in liquid air. This plate radiated through a thin aluminium plate that closed the tube, into an airtight ionisation chamber, where the polonium rays were absorbed by the air. The polonium tube was as far as possible exhausted; and the vacuum was further improved by immersing a side tube containing a little charcoal in liquid air. The radiation was measured at ordinary temperature, and later, when the bottom of the tube was immersed in liquid air. In these experiments changes of current of inconstant amount were observed when cooling was applied. These changes were smaller in proportion as the vacuum was made more complete and kept more constant. It is thus highly probable that they

were entirely due to the influence upon the polonium of the condensation of gases still present in the apparatus.

Experiments made in Leiden in liquid hydrogen with a provisional apparatus have convinced us that one might get rid of the condensations completely, even with liquid hydrogen, by using a ionisation chamber filled with pure gaseous hydrogen and a side tube with charcoal, immersed in liquid hydrogen.

*Conclusions.*

All these experiments which unfortunately are not so complete as we could have wished, confirm the independence of the radiation from the temperature, over a larger range of temperatures than had heretofore been done. Moreover these experiments have brought to light sources of error which must be taken into account, if one wants to make very accurate measurements at low temperatures.

**Astronomy.** — *“The periodic change in the sea level at Helder, in connection with the periodic change in the latitude”*. By Prof. H. G. V. D. SANDE BAKHUYZEN.

At the meeting of the Academy in February 1894 I read a paper about the variation of the latitude, deduced from astronomical observations, and added to this a determination of the change in the mean water level in consequence of the variation of the latitude.

Roughly speaking, one may regard the variation of latitude, as consisting of two parts, a periodic variation which takes place in one year, probably due to meteorological influences, and a periodic variation which takes place in about 431 days, which depends amongst other things upon the coefficient of elasticity of the earth, its resistance to change of shape. As a consequence of these changes of position of the axis of the earth oscillations of the same periods must take place in the mean sea level and if we eliminate the annual oscillation, the periodic variation of 431 days remains.

- For the determination of the latter variation, I had made use of the mean sea level during the different months of the years 1855—1892, taken by the tide gauge at Helder. The results attained then for the amplitude and the phase of the periodic variation confirmed the opinion that such variations actually existed in the water, but as the changes in question are very small, it was desirable to extend the investigation in order to increase the accuracy of the results. I resolved therefore to submit to the calculations all the tidal observations made