



The photographic method in quantitative beta ray spectroscopy

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RAY SPECTROSCOPY

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PROEFSCHRIFT

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DRUKKERIJ Fa. SCHOTANUS & JENS, UTRECHT



Aan mijn Ouders

Jegens allen, die gedurende mijn studietijd tot mijn wetenschappelijke vorming hebben bijgedragen, moge ik hierbij uiting geven aan mijn groote dankbaarheid.

Tot U, Hooggeleerde ORNSTEIN, Hooggeachte Promotor, wensch ik mij daarbij in het bijzonder te richten. De ideale sfeer, die Gij op het UTRECHTSCH E LABORATORIUM wist te scheppen, hebben mij het werken, eerst als student en later meer onder Uw directe leiding als assistent, steeds tot een vreugde gemaakt. Het vertrouwen dat Gij in mij steltet, toen Gij mij verzocht een onderzoek ter hand te nemen samenhangend met de voor het Laboratorium nieuwe, kernphysische richting, heb ik steeds op zeer hoogen prijs gesteld, en het is mij dan ook een groot genoegen de eerste te kunnen zijn, die in den vorm van een proefschrift getuigenis aflegt van deze voor het Laboratorium nieuwe richting van physisch onderzoek.

De groote invloed, die Gij, Hooggeleerde UHLENBECK, op mijn wetenschappelijk denken en physisch inzicht hebt gehad, mede door Uw beminlijke persoonlijkheid en Uw groote didactische gaven, zijn nauwelijks onder woorden te brengen. De intieme colloquia, waarbij onder Uw leiding recente experimenteele en theoretische vraagstukken behandeld werden, zullen voor mij onvergetelijk blijven.

Beste MILATZ, voor de wijze, waarop jij mij steeds in alle moeilijkheden, zoowel binnen als buiten het Laboratorium tegemoet gekomen bent, zal ik je steeds dankbaar wezen. Ook aan de totstandkoming van dit proefschrift heb jij een belangrijk aandeel gehad.

De voortdurende medewerking, die ik van jou, TER HORST, bij alle technische moeilijkheden heb mogen ondervinden, zal ik dankbaar gedenken.

Beste BRINKMAN, dat ik onder jouw leiding de eerste schreden op het gladde pad der experimenteele Natuurkunde heb mogen zetten, beschouw ik als een groot voorrecht.

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INTRODUCTION.

The importance of an exact knowledge of the β ray spectra has led to the development of several experimental methods. It seemed therefore interesting to investigate the possibility of applying the photographic plate for quantitative measurements. In this thesis a purely photographic method has been worked out to obtain the true intensity distribution in a β ray spectrum of a radioactive substance.

In order to obtain the intensities from the photographic densities on the plate, the density curves (relation between density and intensity at a certain energy) and the sensitivity curve (relation between the relative photographic activity and the energy) must be known for the plates used. The density curves have been measured by varying the strength of the source. The sensitivity curve for the plate has been obtained by changing artificially the energy of the β particles with the aid of an electric field, enabling to compare the densities, caused by the same number of particles with and without the changed energies.

The greater part of this investigation has been performed with a ThB source, emitting a combined β ray spectrum of ThB, ThC and ThC'. As a strong γ radiation is emitted by this source, which would disturb some of the intended measurements, a part of the investigation has been performed with a RaD source (in equilibrium with RaE), emitting no γ rays.

No attempt has been made to eliminate the errors influencing the β ray spectrum itself, such as e.g. those caused by stray

radiation, γ rays, etc., as the chief aim of this investigation was to develop the photographic method for β ray spectroscopy.

Due to technical difficulties in using high tensions the application of the photographic method has been restricted to a limited region of energy, namely from 20 to 350 keV.

CHAPTER I.

THE BETA RAY SPECTRUM.

§ 1. General Remarks.

This thesis deals with a method of investigating the β ray spectra of radioactive sources. The nature of these rays is well established, the only problem which still arises and which seems rather difficult from experimental standpoint is that of the distribution of the electrons over the energies, which in analogy with the optical spectra is called the „ β ray spectrum”.

The methods for the investigation of the β ray spectra will be treated in the following paragraph. Here a short account of the main features of the spectra will be given.

They generally consist of one or more rays of homogeneous energy and a more or less continuous spectrum (see e.g. a reproduction of such a spectrum in fig. 27) whose energy range extends from zero to about one to two million electron-volts. For a long time it was assumed that the continuous spectrum originated from stray radiation and thus had no essential meaning. Further research, however, has led to a different interpretation. It could be proved that the β particles producing the continuous spectrum come in reality directly from the nucleus of the radioactive atom, whereas the electrons which produce the homogeneous „lines” are of secondary origin; the latter are thrown out of the electronic atomshell by the γ rays also produced by the nucleus. Two experimental arguments for the primary character of the continuous part of the β ray spectrum are the following:

- a. the energetic upper limit of the continuous spectrum seems

to be connected with the decay period of the radioactive substances in the same way as is the case with the energy of the α radiation;

b. the total number of the β ray particles constituting the continuous part of the spectrum equals the number of decaying atoms of the radioactive source (Gurney²⁴).

These results obtained with the β ray spectra are, however, in contradiction with all other known radioactive phenomena. All energy relations in nuclear physics, namely, are generally characterised by a pronounced discontinuity, such as is the case e.g. with all α and γ energies. The reason for this discontinuity is that every nucleus can merely occupy a discrete set of energy levels, and that a radioactive process consists in a transition of one such energy level to another one. But, how could it be explained that there existed primary particles emitted from the nucleus possessing a continuous energy range without supposing either that the theory of radioactivity was wrong or that the principle of energy conservation was violated? Fermi⁽²⁰⁾ has indicated another possibility: together with the emission of a β particle from the nucleus there would be expelled another hypothetical particle, the so-called „neutrino” which should be electrically neutral and possessing a mass between zero and that of the electron. The whole energy liberated should then be distributed according to statistical laws over the electron and the neutrino. Fermi⁽²⁰⁾ could even calculate with this theory the actual form of the β ray spectrum. The first experiments have shown a slight divergence from the theory of Fermi. Thereupon Konopinski and Uhlenbeck⁽²⁸⁾ introduced a slight alteration of the theory which seemed to fit in better with the experiments, although the latest experiments seem again to confirm the first theory of Fermi.

The main subject of all researches on β ray spectra, which are not only performed on the spectra of the known natural radioactive elements, but also on many artificially radioactive elements, is to compare the spectra obtained experimentally

with the theories of Fermi and of Konopinski—Uhlenbeck. The main criteria of the validity of these theories lie in the behaviour of these spectra at the lowest and at the highest energies. But, just these parts offer the greatest difficulties to exact measurements; the reasons of this will be explained here below.

As the intensities of the spectra decrease strongly at both ends sensitive measuring instruments are necessary, not only in order to find the position of the real limits, but also to determine the exact shape in these regions.

Two other difficulties exist for all experiments on β ray spectra. Firstly the great distortion of the spectrum caused by all sorts of secondary rays, which plays an especially prominent part in the energetically lower parts of the spectrum because all secondary electrons will have lower energies than the primary β and γ rays. Secondly the sensitivity of most detecting instruments depends on the energies of the electrons which are detected. Up to now it appeared to be extremely difficult to determine the way in which the sensitivity depends on the energy. For a long time the importance of these two sources of error has not been recognised and so frequently wrong conclusions have been drawn.

This thesis will especially deal with one of the above mentioned points, namely the determination of the sensitivity function for the photographic method. It must, however, be emphasized that the principle of the method can be applied also to other cases.

The following paragraph will give a brief survey of the experimental methods applied at present with the aim of getting an appreciation of the significance of the photographic method compared with the other methods of investigation.

§ 2. Review of experimental Methods.

A. *Methods of detection.*

Here below will be given a brief enumeration of the methods

of detection with their principal characteristics concerning their way of operation, the possibilities of their application, their sensitivity, their precision and their degree of selectivity.

Measurement of Charge.

This method is the most direct one for obtaining the number of electrons and is not at all selective. Gurney⁽²⁴⁾ employed e.g. a Faraday cage in which the electrons are caught. He intended not only to obtain the intensity distribution in continuous β ray spectra, but also to determine the total number of primary electrons which, as explained in the preceding paragraph, is important for the knowledge of the origin of these spectra. The charge is measured by a sensitive electrometer.

Apart from instrumental difficulties the only drawback is the relatively low sensitivity of this method. The electrometer used by Gurney possessed a sensitivity of 3×10^{-4} volt per division. The capacity of the system being mainly defined by that of the cage cannot be lowered arbitrarily. The capacity of the system used by Gurney was 87 cm, so that one division corresponded to about 18×10^4 electrons. It appears from his reproduced curves that it was possible to measure the rate of accumulation of charge with an accuracy of about 1 division per minute (limited e.g. by natural leak and secondary effects). This means that about 10^4 electrons per second are necessary to keep the error under 10%. The best electrometers obtainable nowadays are about ten times more sensitive than that used by Gurney, so that the minimum current for measurements with 10% accuracy can be accepted to be 10^3 electrons per second. This method cannot be improved by other methods of measuring the charge, such as e.g. that using amplifiers.

Recapitulating, the method, sketched above appears to have the great advantage of being a direct method without selec-

tivity effects, but it is necessary to employ relatively strong electronic sources.

Measuring of Ionization in an Ionization Chamber.

This method is based on the fact that electrons, passing through a gas-filled volume, cause an ionisation of the gas the magnitude of which depends on the electronic energies. Introducing an electric field in this gas the positive and negative ions move to the cathode and anode respectively. The total charge of these ions can be measured either with an electrometer or amplifier.

This method has frequently been applied in all domains of nuclear physics, however not for measuring the β ray spectra.

The main disadvantages are the great selectivity and the relatively big disturbances, caused by the effect of other ionizing agents which are of the same order of magnitude as the electronic effect itself. The main reason, however, that this method is scarcely used for this purpose, is the existence of better methods. The ionization chamber has in fact been applied for β ray spectra only by Ellis⁽¹⁴⁾, Chadwick⁽¹²⁾ and Madgwick⁽³³⁾.

Concerning the sensitivity attainable with this method a high-sensitive ionization chamber, recently designed by Barnes⁽³⁾, may be mentioned, enabling the measurement of about one electron per second.

In § 13 an application of the ionization chamber to the measurement of the strengths of radioactive sources will be treated.

The Counters.

The principle of the electric counter was first introduced by Rutherford and Geiger⁽³⁸⁾, further developed by Geiger⁽²³⁾ to the so called „Spitzenzähler“ and finally perfected in 1928 to the „Geiger-Müller“ Counter.

The principle of operation is the following: just as in the case of the ionization-chamber an electric field is applied

between two electrodes in a gas-filled space. The strength of this electric field is, however, chosen near the value where a spontaneous discharge would take place. As soon as an electron enters the field, it is accelerated and produces secondary ions which in their turn produce again new ions etc., so that the primary ionization is automatically amplified many times. The geometrical and electrical device provides a breaking off of the discharge after a sufficiently short time. The advantage of this method will immediately be obvious. The effect of even one electron being greatly amplified all auxiliary apparatus can be very simple. As furthermore the magnitude of the discharge does not depend on the initial cause any selectivity of the method seems to be absent, provided, of course, there exists a primary ionization.

The type of Geiger-Müller counter, which at present is mostly used, consists of a thin wire forming the anode and of a coaxial cylinder forming the cathode.

According to the above given view the counter would represent the ideal measuring instrument, with a sensitivity of counting each entering electron and with a total absence of selectivity. Unfortunately matters are somewhat different. The energetic selectivity begins to play a part at higher and at very low electronic energies. The reason is that the ionization per unit of length is a function of the energy and that it diminishes at higher energies, so that the probability that an electron will not produce one or more pairs of ions in the counter will increase. The probability can be calculated (*viz.* Ornstein, Milatz, ten Kate and Miesowicz⁽³⁴⁾) and the calculation has experimentally been verified by the same authors. Riehl⁽³⁷⁾ and recently de Vries and Sizoo⁽⁴⁵⁾ have also made researches about this point. The probability that an electron will or will not be detected, depends not only on the energy of the entering electron, but also on the pressure of the gas in the counter and on the geometrical circumstances.

As to the sensitivity of the counter, this will already be

diminished by the above explained selectivity effects. But still another cause exists limiting the ideally attainable value of sensitivity, namely the so-called „zero-effect” which is caused by spurious radioactive and cosmic radiations. Even when there is no electronic source in the neighbourhood of the counter, this zero-effect exists depending on the geometrical and other conditions of the counter, and which amounts in general, for the β counters applied for radioactive purposes, to about one impulse per three seconds.

Here also it is possible to calculate the minimum intensity of the electronic source necessary to obtain a certain accuracy, e.g. of 10%. But because of the fact that this accuracy is here determined by purely statistical factors, it can be augmented arbitrarily by increasing the time of observation, because the relative statistical fluctuation of a number of observations diminishes inversely proportional to the square root of this number, contrary to most other sources of errors. Thus we must fix a certain time of observation. With the above given value of the zero-effect it is easily verified that for a time of observation of 1 minute the number must be at least 200 electrons per minute to have an error less than 10%, whereas for a time of observation of three minutes the corresponding number is 30 per minute.

The Geiger-Müller counter is by far the most frequently applied instrument for measurements in the β ray spectra, so that it may suffice to mention only the latest publications referring to investigations on the continuous spectra of Ra D + E and Th B + C + C", performed with counters and in which other literature can be found. These publications are of *Flammersfeld* ⁽³¹⁾ on the continuous spectrum of Ra D + E and of *Alichanian* and *Zavelskij* ⁽¹⁾ on the continuous spectrum of Th B + C + C".

The Scintillation Method.

This method is based on the fact that when particles are falling on screens of phosphorescent zinc sulfide, or some

other substances, the spots which have been hit by such particles show short flashes of light which can be seen by considering the screen with a microscope. This method has often been used for α particles, but the effect caused by electrons is too weak for applying the method in this way to quantitative measurements in β ray spectra. The only cases in which the scintillation screen is used in quantitative electronic researches are for the visual estimation of intensities in cathode-ray tubes, or for amplifying the action of electrons on the photographic plate (only applicable with electrons of low energies, viz. v. Borjes and Knoll⁽⁵⁾).

The Cloud Chamber.

This method which was first introduced by Wilson⁽⁴⁸⁾ in 1911, and which is used very much in the whole domain of nuclear physics is based on the following phenomenon. When a gas volume which is saturated with water vapour, is suddenly expanded, so that the gas becomes over-saturated, the water vapour tends to condensate on charged nuclei. It is possible to adjust the expansion in such a way that the condensation will only take place at such spots where charged atoms are present. This phenomenon furnishes the possibility of studying the tracks of elementary ionizing particles, the secondary ions acting as charged nuclei.

The most characteristic feature of this method compared with all preceding ones, is its possibility to study, or to register photographically individual particles and processes, and to give many-sided informations with each observation; namely it gives the exact form of the track in three dimensions, the nature of the particle and also its energy. However, apart from all these advantages, there exist also disadvantages, and one of the most serious ones is, that this method becomes very laborious when it is necessary to study a great number of such processes, as is required for statistical purposes, and also for the study of β ray spectra. This is probably the reason that this method is very rarely applied to the β ray spectra.

The Photographic Method.

Since the whole following chapter deals exclusively with this method, only some characteristic features will be mentioned which are of importance for comparison.

The photographic plate has some properties analogous with those of the expansion method, and others belonging at the same time to the other methods mentioned above.

It has in common with the Wilson chamber method that it is also able to fix individual processes, though under less favourable circumstances. Yet, this function of the photographic plate has e.g. been used by Blau, Wambacher⁽¹⁴⁾, Taylor⁽⁴⁴⁾, Powell and Fertell⁽³⁶⁾ for the investigation of the ranges of different particles, for the study of nuclear reactions and recently for the determination of neutron energies resp.

On the other hand the density of the photopraphic plate enables, after calibration, to determine the number of particles that have fallen on the plate, so that the plate can also be used as a counting instrument.

This combination of properties, however makes the photographic plate so specially suitable for the intended purpose, as it is possible to obtain with one observation a whole range of intensities as a function of the position on the plate.

But, apart from the advantages of this combination of possibilities, the photographic plate possesses also some of the disadvantages of the preceding methods.

In the first place there exists a pronounced energetic selectivity. The greater part of this paper will deal with the problems in connection with this point.

Furthermore the magnitude of the sensitivity of the photographic plate is relatively small. As this point will also be treated in detail in the following chapters, only some general remarks will be made. As the photographic plate is an accumulative instrument and as moreover its sensitivity depends on the size of the exposed surface, this sensitivity has to be defined

in a specific way; namely by the number of electrons which has to fall on a certain surface of the plate (in an arbitrary time) to enable the measurement of the intensity with an accuracy of e.g. 10⁰%. So for instance a number of 10⁵—10⁶ electrons per mm² with an energy of 100 keV is required for this purpose. A further discussion of the circumstances determining the sensitivity will take place in the last chapter.

B. *Methods of measuring the energy.*

A short enumeration of the methods, used for this purpose, will be given enabling to judge the relative significances of these methods, and to justify the choice made in this paper.

The Absorption Method:

All sorts of radioactive radiations undergo absorption (a diminution of intensity and energy) in matter, the mechanism and the rate and kind of absorption of the radiation being different under different circumstances.

If the radiation is of an energetically homogeneous nature, and consists of a parallel beam, an exponential dependence between the transmitted intensity J and the thickness x of the absorbing material will exist:

$$J = J_0 e^{-\mu x}$$

Here μ is called the absorption-coefficient, which depends on the kind and energy of the radiation and on the absorbing material. It appears that the absorption-coefficient is approximately proportional to the density ρ of the absorber; it is therefore convenient to introduce the mass-absorption-coefficient $\frac{\mu}{\rho}$, which is thus independent of the absorbing material.

It must, however, be emphasized, that the exponential law holds only for absorbing thicknesses, which are small compared with the total range, because at greater absorptions a qualitative alteration of the radiation takes place, as not only the intensity,

but also the energy diminishes and becomes inhomogeneous, and moreover the spatial distribution becomes more divergent.

In reality the notion of absorption is a many-sided one. Firstly there must be made a distinction between an energetic absorption and an absorption in intensity. This latter can be subdivided in a real absorption (when the radiation is partly or wholly captured in the atoms of the absorber) and a scattering and reflection out of the beam.

The application of the absorption method to energy measurements of β rays appears to offer serious difficulties, though it has been applied in the earlier experiments. The reasons why this method is unsuitable for this purpose are that it is difficult to obtain homogeneous and narrow electronic beams of radioactive origin (only in this case there exists a well defined dependence of the absorption-coefficient on the energy) and that relatively large energy losses occur, even at small absorptions.

Application of electric Fields.

This method can be applied in two forms, namely by determining the deviation of the particle or by determining the tension, which must be applied to stop it. Its application is only possible in those cases where the energies of the particles are not too high, because otherwise high electric tensions are necessary. As all radioactive radiations are generally of a rather high energetic nature, this method has seldom been applied in the cases which are considered here.

Application of magnetic Fields.

The application of a magnetic field has two important properties causing that this method is by far superior to all other methods:

1. the action of the magnetic field on a charged particle alters the shape of the path without changing the energy of the particle so that the energy remains well defined during the whole time of a measurement.

2. It is technically rather easy to produce magnetic fields of the required strengths, and no materials except the ferromagnetic ones disturb the field.

The measurement of the energy by this method takes place generally with transversal field (field perpendicular to the velocity of the particle). In this case the particles describe circles, whose radii depend in a simple way on the energy of the particles. A closer discussion of this point will be given in § 10.

The radius can be determined, either by using a small part of the circle and measuring the deviation of the endpoint of the considered part of the path, or by using one half of the circle and measuring directly the radius. The first alternative is sometimes necessary, when the particles are very fast and heavy so that it would be difficult to extend the magnetic field over a sufficiently large area to enclose a greater part of the circular trajectory within the field.

The second alternative, namely of using half circles, was first proposed by Danysz⁽¹⁸⁾. It has also been applied in this thesis due to several more special advantages regarding the application to the photographic method. As, namely, the sensitivity of the photographic plate depends on the size of the area which is exposed to a certain beam of particles, it is of special importance to have the possibility of collecting as much as possible of the beam on a small area of the plate, but this focussing must not take place at the expense of the separation of the different energies. The method of using half circular trajectories meets all these requirements in a satisfactory way.

CHAPTER II.

THE PHOTOGRAPHIC METHOD.

§ 3. The blackening Action of various Radiations.

In this paragraph a brief survey will be given of the main known facts concerning the blackening action of various radiations on the photographic plate.

The photographic plate can be blackened by visible, ultra-violet and infrared light as well as by Röntgen, γ , β and α rays.

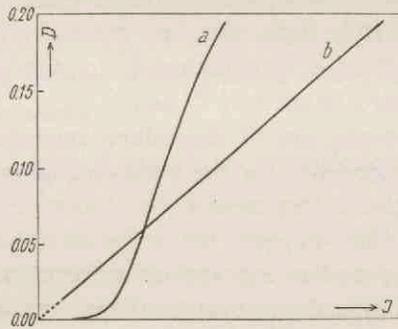


Fig. 1.

- a. Density curve for visible light.
- b. Density curve for Röntgen rays.

In fig. 1, the general shape of the density curves (curves giving the density as a function of the intensity falling on the plate) for visible light and Röntgen rays are given, and

from this figure the difference in the photographic action of these two kinds of radiation can be seen. The curves for the optical radiation always show a superproportional part at low intensities (one speaks often of the existence of a „thresholdvalue“ of the intensity, being the value of the intensity where the linear part of the curve intersects with the abscissa), whilst the curves for Röntgen rays show, already from the beginning, a proportional relation. The γ , β and α rays appear to act on the plate in the same way as the Röntgen rays. The exact shape and the factors determining the shape, will, in the case of the β radiation, be discussed in the paragraphs 5, 16 and 18.

Another difference of the behaviour between the rays giving a density curve of the type *a* (fig. 1) and those giving a density curve of the type *b* exists. Only in the last case the so-called reciprocity law of BUNSEN-ROSCOE⁽⁹⁾ holds, that is to say that the density, caused by a certain total intensity of radiation, is independent of the time during which this intensity has fallen on the plate. In the other case, e.g. for visible light, this law generally does not hold. This point will also be discussed, in the case of the β radiation, in more detail in § 5 and § 16.

All further results are of secondary importance and will be treated subsequently in the following paragraphs, so far as the β radiation is concerned.

Although it lies beyond the scope of this thesis to give an account of the modern conceptions concerning the mechanism of the blackening action, some consequences of the conceptions put forward by Webb⁽⁴⁷⁾, Gurney and Mott⁽²⁶⁾ may still be given here because they are immediately applicable to the electronic action on the photographic emulsion.

The action of those rays which give rise to density curves of the type *a* (fig. 1), appears to be characterised by the fact that on the average about 100 quanta must be absorbed by an AgBr grain of an undeveloped emulsion in order to make this grain developable, whereas for all rays giving a

density curve of the type *b*, already one absorbed quantum may be sufficient for this purpose. These results which are explained by the conceptions of the above mentioned authors, allow some conclusions concerning the action of electrons on the photographic emulsion, assuming that already one electron can make an AgBr grain developable. This assumption seems justifiable by the great ionizing power of the electrons, considered here.

From this assumption it follows that the density curve for the electrons will be of the type *b* in fig 1, and furthermore that the reciprocity law will be valid. Another consequence is that the shape of the density curve is independent of the electronic energy.

One must, however, be warned against the error of thinking that it should already be possible to determine once for all a density curve for electrons being generally valid. As long as this point has not been checked by extensive experimental work, it remains necessary to determine the density curve for each plate and for each electronic energy separately.

§ 4. The Use of the photographic Plate for quantitative Measurements.

The photographic method for quantitative measurement has some properties which procure a particular position for it among all other methods. The plate furnishes a measuring instrument that never fails to work so that the trouble caused by disturbances of instrumental kind do not occur.

The only disturbances playing a part are caused by the faults and fluctuations originating at the manufacturing of the plates. These irregularities necessitate the introduction of a special technique when using the method: every plate has to be calibrated separately.

The calibration of a plate is carried out by determining the relation between the density and the intensity falling on the plate per unit of area for one or more wavelengths or

energies. If this dependence can only be determined relatively (i. e. if only the ratios of the intensities are known), the calibration has to be divided into two parts: a determination of the relative density-intensity curves for the separate wavelengths or energies, and a determination of the relative sensitivity of the plate for these different wavelengths or energies.

The measurement of the relative density curves offers no difficulty. The relative intensity falling on the plate can be varied in a known way: e. g. by variation of distance and of the strength of the source or by using diaphragms*).

The determination of the sensitivity of the plate as a function of the wavelength or the energy is, however, more difficult. For electronic radiation it has not been carried out up to now and also in the case of optics it has taken a longer time to establish a suitable method. In § 6 and § 7 the method for obtaining the sensitivity in the case of β radiation, and the comparison with the method used in optics will be elucidated.

Some attention may be paid to the measurement of the density. Though the definition and the way of measuring this quantity is irrelevant, because the same instrument and definition is used in calibrating and in using the plate, it is, on the other hand, important for the discussion of the accuracy and for the comparison of the results of different authors.

The suitable quantity for expressing the density, is defined in the following way: if J_0 is the intensity of light falling on the plate, J the intensity transmitted, then the density D is given by

$$D = {}^{10}\log \frac{J_0}{J}$$

The value of the density depends, however, on the optics

*) Often variation in time of exposure has been used; this is only justified if the reciprocity law (see § 3) has been tested.

of the applied photometer, and discrepancies between the results of different authors may partly originate from this point (§ 18).

In the experiments described here Moll's microphotometer has been used. For a detailed description see Ornstein Moll and Burger⁽³⁵⁾. This photometer renders possible the measurement and the registration of the density of the plate even on areas as small as 10^{-6} cm². For further discussion of this subject see § 20.

§ 5. Review of the Use of the photographic Plate for quantitative Measurements of β Ray Spectra in the Literature.

Before 1920, the authors who used the photographic plate for the study of the β ray spectrum contented themselves with visual estimations of the intensities (e.g. of the different characteristic lines in a β ray spectrum). In 1921 Geiger and Bothe⁽⁶⁾ calibrated the plate for the first time with the help of a density curve, obtained by variation of the distance. In their experiments they used the whole β radiation of RaE, consequently an energetically heterogeneous radiation. Notwithstanding this fact they obtained some important density laws, namely the proportionality of the density curve at low intensities and the validity of the reciprocity law (see § 3).

Further research on this point had not been done until 1927. Ellis and Wooster⁽¹⁸⁾, who wished to determine the absolute intensities of the characteristic lines (in order to get the internal conversion coefficients), for the first time applied the action of energetically homogeneous rays on the plate. These rays were obtained by the half-circular focussing method, explained in § 2 and discussed more thoroughly in § 10. They were able to confirm the results obtained by Bothe and Geiger in the case of heterogeneous rays, for that of homogeneous rays. They found in addition that the shape of the (logarithmic) density curve is independent of the electronic energy and, within certain limits, of the

conditions of development, and moreover that the reciprocity law is valid.

Concerning the sensitivity of the photographic plate for different electronic energies these authors had to assume a certain dependence of it on the energy without being able to check this assumption experimentally. They assumed namely, that the sensitivity would be proportional to the ionization of the particles in the plate. This ionization was known to be inversely proportional to the square of the velocity.

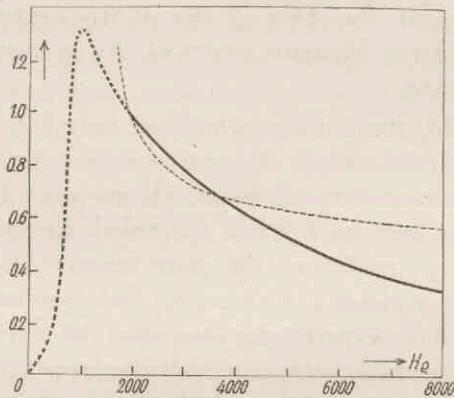


Fig. 2.

Relative photographic action of electrons of different energies, found by Ellis, Aston and Wooster (^{16, 18, 15}). The dotted line gives the sensitivity which they expected from theoretical considerations.

In 1928 Ellis and Aston (¹⁶) published a comparison of the photographic results with those of the measurements of Gurney (³⁴), who had worked with a Faraday-cage on the same spectra (charge-determination, see § 2), and in this way they were able to obtain for the first time a sensitivity curve for the photographic plate. This curve, which is represented in fig. 2 by the solid line, shows that the original assumption of Ellis and Wooster (which is represented by the dotted curve in fig. 2) was not correct.

As an explanation for the failure of this hypothesis, they assume that it is caused by the reflection of the electrons in the emulsion and on the glass. This point will be discussed more quantitatively in § 19.

In 1932 Ellis ⁽¹⁵⁾ published a continuation of the sensitivity curve to lower energies, which was based on the comparison of some characteristic lines whose intensities were measured by others. This proposed continuation is represented by the broken part of the curve in fig. 2. However, the author admits, that this part is somewhat problematic.

In conclusion a few words may be said about a publication by Bories and Knoll ⁽⁵⁾ in 1934. These authors have used the electrons of a cathode-ray tube, and could attain energies up to 80 keV. They give a number of absolute density curves, from which the sensitivity curves can be derived directly. They find a surprisingly great dependence of the shape of the density curves on the energy so that their results seem to be in contradiction with other experiments.

The preceding researches (except the last one), have been performed in order to investigate the relative intensities of the characteristic lines in the β ray spectra. The photographic method has, however, never been applied to the continuous spectrum. In this case, these continuous spectra being the subject of investigation, it is even more important to know the sensitivity of the plate. The purely photographic method applied in this thesis in order to obtain this sensitivity of the plate will be described in the next paragraph.

§ 6. The Method of changing artificially the electronic Energies in a β Ray Spectrum.

The following problem had to be solved: when the photographic density on two spots of the plate caused by two beams of electrons of known energies is given, then the unknown intensities at these two spots must be found.

If the energies of the electrons in the two beams are equal, the intensities can be obtained by means of the density curves.

If, however, the two energies are unequal, the following artifice has been applied: change the electronic energy of one beam by an electric field in such a way that this energy becomes equal to that of the second beam. In this way obtaining two beams of the same energy, the ratio of the intensities can be determined by means of the „monochromatic” density curve and thus the desired ratio of the first and the second beam are obtained.

The way in which this principle can be applied to the β ray spectrum will be explained below.

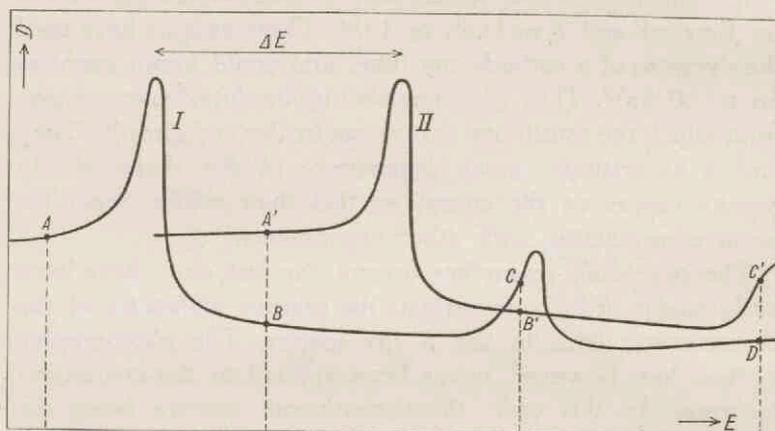


Fig. 3

Illustration of the principle, representing an imaginary photographic spectrum: I with unchanged energy, II with changed energy (changed by ΔE).

In fig. 3 a photographic β ray spectrum (I) is schematically represented (the density D as a function of the energy). It is possible to obtain the relative intensities in the points A, B, C, and D, showing equal distances in energy, by taking one more spectral photograph (supposing that the density curves are known). In this second photograph all energies ought to be changed by an amount ΔE , corresponding to the distance in energy between the points A, B, C and D.

This energy-shift can simply be obtained by a corresponding electric tension, and changing the energy of every electron in this way by the amount ΔE . The shifted spectrum is represented in fig. 3 by curve II, and the points, corresponding with A, B, C and D are A', B', C' and D'. The intensities at A', B', etc. are, however, equal to those at A, B, etc. respectively.

These relations can be written analytically as follows:

$$\frac{J_{A'}}{J_A} = 1 \quad \frac{J_{B'}}{J_B} = 1 \quad \frac{J_{C'}}{J_C} = 1$$

whereas the ratios

$$\frac{J_B}{J_{A'}} = p_1 \quad \frac{J_C}{J_{B'}} = p_2 \quad \frac{J_D}{J_{C'}} = p_3$$

can immediately be obtained by means of the density curves, the energies of the electrons, falling on the plate at the points A and B', B and C', C and D' respectively, being equal. In this way the desired ratios are obtained in the following form

$$\frac{J_B}{J_A} = p_1 \quad \frac{J_C}{J_B} = p_2 \quad \frac{J_D}{J_C} = p_3$$

The result therefore is, that with two photographic spectra, one unshifted and the other one shifted, the real intensities in a number of points of the spectrum, can be obtained.

It is, however, possible to obtain the intensities not only in these points, but over the whole spectrum. For this purpose it is necessary to derive the sensitivity curve of the plate. This is possible with the aid of the same pair of spectral exposures. The energies corresponding to the points

A, B, C and D will be written $E(A)$, $E(B)$, $E(C)$ and $E(D)$, respectively.

In order to deduce the value of the sensitivity for the plate e.g. at the energy $E(B)$, expressed in that of the energy $E(A)$ (being the ratio of the intensities with energy $E(A)$ and $E(B)$ resp. giving the same density), it is sufficient to determine the densities, caused by the same electronic intensities, but having energies $E(A)$ and $E(B)$ resp. These densities can directly be read from fig. 3, and are simply D_A and $D_{A'}$ (the densities in the points A and A'). The relative sensitivity at the energies $E(C)$ and $E(B)$ is determined in the same way; the reduction to the first energy $E(A)$ is accomplished by means of the density curve. In this way the sensitivity for the energies belonging to the points A, B, C and D can be derived. In this case, however, the values at intermediate points can also be obtained owing to the fact that the sensitivity curve of the photographic plate is a smooth function, so that it is permitted to interpolate between these points.

The way in which the real intensities in arbitrary points of the spectrum can be determined with the aid of the density- and the sensitivity curves of the plate, is thus evident.

The experimental details and results, obtained with the principle explained in this paragraph will be the subject of the following chapters.

§ 7. Comparison with the photographic Method used in Optics.

Before proceeding to the treatment of the experiments and their results a comparative survey of the differences and resemblances between the method, given in the foregoing paragraphs and that used in optics will be given. The technique of quantitative spectroscopic measurement has been developed especially for the purpose of optical research in the Utrecht laboratory and so it was possible to profit by the experience acquired.

In the two cases of electronic and optical spectroscopy the circumstances are nearly the same: in the first case the deduction of the real intensity from the density is wanted, when the energy is given, and in the latter case the deduction of the intensity from the density is wanted at given wavelength.

This analogy can be emphasized by considering the light as a stream of photons in the same way as the electronic radiation consists of a stream of electrons.

In both cases it is necessary to measure the density-intensity curves of the plates, and the determination of them takes place essentially in the same way, namely, by weakening the intensity of radiation falling on the plate in a known way.

The second alternative, namely variation in the time of exposure is incorrect except in those cases where the reciprocity law holds.

Up to the present the circumstances are, however, entirely different regarding the determination of the sensitivity of the plate. In optics a calibrated „standard lamp” is used. The calibration refers to the relative or absolute intensities emitted by the lamp as a function of the wavelength, and are carried out with other, absolute measuring instruments (see e.g. Ornstein, Moll and Burger⁽³⁵⁾).

The application of a standard source to electronic spectroscopy could not yet be taken into consideration, because no reliable calibrated sources were available. But even if a standard source would exist, several difficulties would still have to be overcome. In principle, however, the use of a standard source should be recommended for the future.

In the preceding paragraph a method has been explained to obtain the sensitivity of the plate in electronic spectroscopy, without the use of a standard source, and which is based on the possibility of changing the energies without altering the intensities. The application of the same method in optics would require the possibility of changing the wavelength of the light without changing the intensity, and is therefore not yet possible.

CHAPTER III.

THE EXPERIMENTAL ARRANGEMENT.

§ 8. The magnetic β Ray Spectrograph.

The photographic β ray spectra were obtained by bending the beam of electrons, coming from a radioactive source S , into half circles with the aid of a homogeneous magnetic field and intercepting them by a photographic plate P (see fig. 4).

All exposures were taken in high vacuum.

The spectrograph itself consisted of a metal, entirely air-tight box. It was constructed of aluminium in order to avoid as much as possible stray electrons and secondary radiations. Only the plateholder Ph and the air-tight joints J for this plateholder and for the screen were made of brass.

The dimensions were chosen in such a way that the radii of the half-circular trajectories of the electrons ranged from 2.5 cm to 5.5 cm, and that the distance of the magnetic pole-pieces was 4.5 cm.

The radioactive sources consisted of a thin wire or strip, fixed on an ebonite holder, which could be mounted by two plugs in a fixed position. The length of the source was 0.8 cm and it was fixed at a distance of 1.6 cm from the limiting slit which had a width of 0.4 cm. The other slits E served for the distribution of the electric tension, as will be explained in the next paragraph.

The photographic plates were cut to a size of 2×7 cm², and rested on a brass plateholder Ph . The plate and the

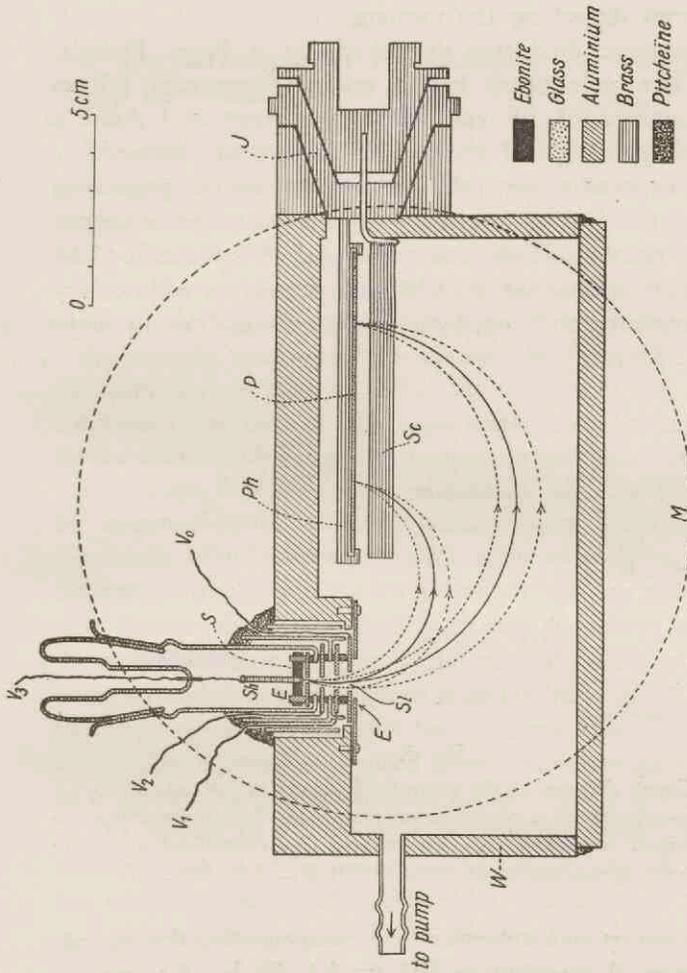


Fig. 4. The magnetic β ray spectrograph.
 S = radioactive source; Sh = sourceholder; Sl = limiting slit; E = electrodesystem for energy-change (see § 9); W = walls of the spectrograph (made of aluminium); P = photographic plate; Ph = plateholder; Sc = screen (screening half of the plate); J = 2 ground joints for plateholder and screen; M = contour of the magnetic pole-pieces; V₀, V₁, V₂, V₃ = electric tensions for changing the electronic energies.

plateholder could easily be removed and re-set with the aid of an air-tight ground joint *J*.

In this joint another joint had been constructed permitting to rotate the screen *Sc* and to expose alternately half of the plate without disturbing the vacuum.

The magnetic field was furnished by a large Dubois-magnet. The pole-pieces had a mutual distance of 4.5 cm and a diameter of 16 cm. With a current of 1 Amp. a magnetic fieldstrength of about 600 Gauss was obtained.

As the exposures generally extended over a period of 6 to 10 hours, it was necessary to maintain the magnetic field fairly constant. This was accomplished by feeding the magnet with a constant accu-battery *B*, and in addition by applying current stabilizing, hydrogen filled iron-filament lamps *L* (see fig. 5).

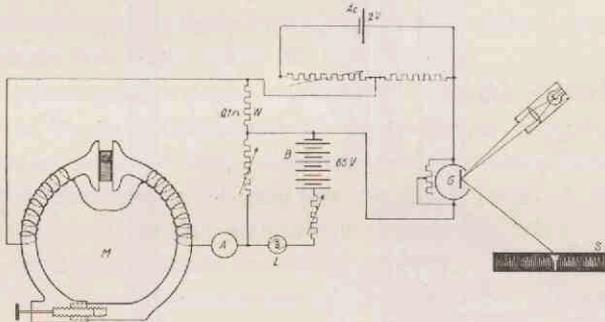


Fig. 5.

Circuit diagram of the magnetic field and its control.

M = Dubois-magnet; *W* = compensating resistance; *L* = hydrogenic iron-lamps for current stabilizing; *A* = amp-meter; *G* = control-galvanometer; *Ac* = compensating accumulator (2 V); *B* = Accu-battery.

The constancy was supervised by compensating the magnet current over the resistance W ($\approx 0.1 \Omega$) by the current from a storage battery *Ac* of 2 V and observing the zero-deflection of the galvanometer *G*. In this way variations of less than $1 \frac{0}{00}$ in the magnet current could easily be observed.

In view of the required long exposures, it was necessary to make sure that no light could penetrate to the photographic plate. Special care was therefore taken to protect these plates from light.

§ 9. The Arrangement for changing the Energy of the β Particles.

In order to change the energy of the β particles, two possibilities were available, namely, the change could either be performed in the beginning of the path (near the source), or at the moment, when they fall on the plate. Both possibilities possessed certain advantages, but technical reasons confined the choice to the first one.

As already mentioned the change in energy is obtained by applying an electric field. The choice of this change is confined, on account of the experimental requirements, between rather narrow limits: if the change is too small, the effects which are a function of its magnitude, become too small to be measured with sufficient accuracy (see § 20) and if it is too large, other sources of error come to the fore, e. g. large deformations of the trajectories (regarding this point see the next paragraph). As a result a change in energy of 25 % of the original energy appears to be suitable. The measurement being extended over a range of energies from 20 to 300 keV, the applied electric tensions had in some cases to be rather high. The arrangement, used here, permitted an application of a tension of maximum 32 kV.

The system of electrodes E which has been applied for this purpose is represented in fig. 4. It consisted of four flat circular equidistancial electrodes, consisting of aluminium in order to avoid stray-effects. Each electrode was provided with a rectangular slit, whose dimensions were chosen in such a way that no electron is stopped which would be able to pass through the last, limiting slit Sl . The outer edges of these electrodes are entirely embedded between glass (fixed

on the glass with pitcheine), as can be seen from fig. 4, in order to be sure that no discharge could take place from these edges. The electrical connections to the outside also pass exclusively between glass-walls.

This construction resulted from a great number of trials necessary to overcome several difficulties. On account of geometrical considerations (see the next paragraph) it was advantageous to have the distance over which the field was applied, as small as possible. On the other hand a definite limit for this distance existed determined by the fact that a tension of the given magnitude had to be maintained. By using the system of electrodes, described above, a well known artifice has been applied: the electric tension has been divided in three equal parts by the two intermediate electrodes in order to reduce the probability of ionization in remaining rests of the gas (the tension is only applied under high-vacuum conditions: the pressure being less than 10^{-6} mm mercury).

Yet, even in the case, that the tension seemed to be well maintained, small discharges appeared to exist and were still able to blacken the plate, due to the long durations of the exposures. The construction described here had the same inconvenience when the source and the electrodes had a positive potential with respect to the rest of the spectrograph. However, in all experiments a negative potential could be used. This means that the electrons have always been retarded*).

The high-tension was obtained by means of a transformer of 0.1 kW transforming 125 Volts up to 10.000 Volts. By rectifying both phases of the secondary output, a continuous tension of 28 kV is obtained.

The circuit diagram used in these experiments is represented in fig. 6, and requires no further explanation. A galvanometer

*) The proper cause of this asymmetry has never been recognised. It was proved by an exposure without the source, that this asymmetry was not caused by the radioactive source.

G again furnishes the possibility of controlling the value of the tension during the exposures.

From the electrical constants given in the diagram it can be calculated that the rectified tension is smoothed out to less than $\frac{1}{2}\%$.

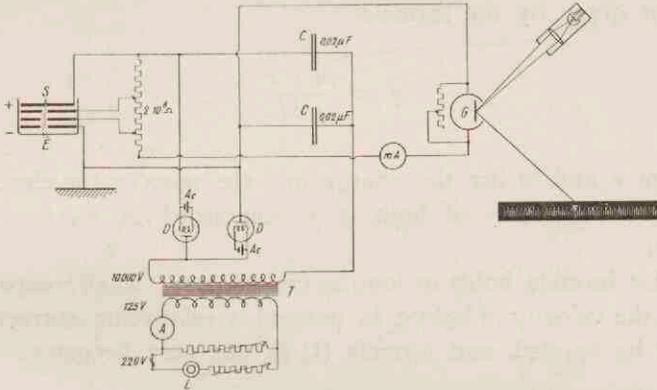


Fig. 6.

High tension circuit diagram.

E = electrode system; L = current stabilizing lamps; C = condensers of $0,02 \mu F$ and for max. 60 kV; D = diode rectifiers for max. 40 kV; W = resistance of $18 \times 10^7 \Omega$ and $18 W$; G = galvanometer for controlling the tension.

The distribution of the tension over the different electrodes was accomplished by means of a high resistance of $18 \times 10^7 \Omega$, from which the required tensions could be taken.

In order to ensure a sufficiently constant tension stabilizing lamps L have been applied in the primary circuit of the transformer. The required constancy is not so high as in the case of the magnetic field, because the applied electric tension corresponds only to a fraction of the whole energy. A variation of 1% in the tension e.g. corresponds with a variation of about 2% in the electronic energies.

§ 10. Geometrical and Kinematical Considerations.

A. Homogeneous magnetic field.

If the direction of the velocity v of an electron is perpendicular to the direction of a homogeneous magnetic field of strength H , this electron describes a circular path situated in a plane perpendicular to the magnetic field, and with a radius given by the formula

$$(1) \quad \varrho = \frac{m v c}{e H}$$

where e and m are the charge and the mass of the electron and c the velocity of light (e is expressed in electrostatic units).

This formula holds as long as the velocity is small compared with the velocity of light c . In general a relativistic correction must be applied, and formula (1) in this case becomes:

$$(2) \quad \varrho = \frac{m c^3}{e H} \frac{\beta}{\sqrt{1 - \beta^2}}$$

where β is written for $\frac{v}{c}$. The expression for the kinetic energy of the electron is:

$$(3) \quad E = m c^2 \left(\frac{\beta}{\sqrt{1 - \beta^2}} - 1 \right)$$

A few remarks must be made concerning the way in which the electronic energy is expressed. The usual unit of energy is the erg. In atomic and nuclear physics, another unit, namely the electron-volt is applied:

$$1 \text{ eV} = 1.59 \times 10^{-19} \text{ ergs.}$$

In electronic research, it is sometimes advantageous to

express the energy in another way, namely by the product $H\rho$, ρ being the radius of the circular path described by an electron in a homogeneous magnetic field of strength H . This product is a function of the energy. This function can be obtained by eliminating β from (2) and (3), and is represented in fig. 7.

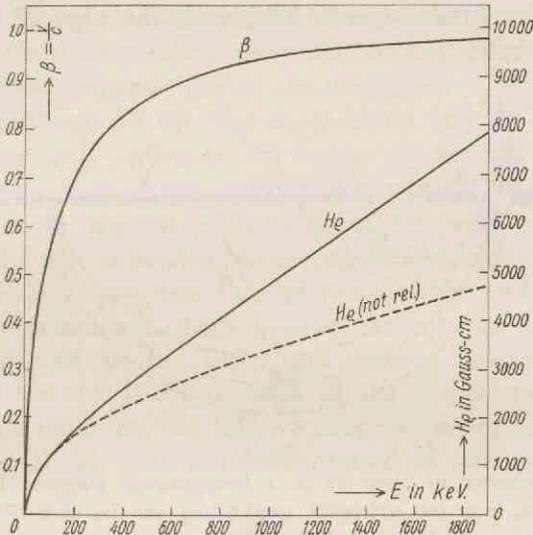


Fig. 7.

$H\rho$ (in Gauss-cm) and β as a function of the energy (in keV).

Dotted curve gives the dependance of $H\rho$ without applying the relativistic correction.

The same figure indicates at which energy the relativistic correction begins to play a part.

If the directions of velocity and magnetic field are not mutually perpendicular the electron describes a spiral, the velocity showing a constant angle with the field, whilst the enveloping cylinder has a radius, given by (2), multiplied by the sinus of the mentioned angle.

Applying these elementary laws to the case discussed in the foregoing paragraphs, it is possible to study the realization of the photographic β ray spectrum in more details.

Suppose that a beam of electrons of *one given velocity* is emitted by the source S (fig. 8) and passes through the limiting slit Sl . The figure shows that the electrons describing three arbitrary trajectories a , b and c , lying in a plane perpendicular to the magnetic field, meet the plate P at nearly the same spot.

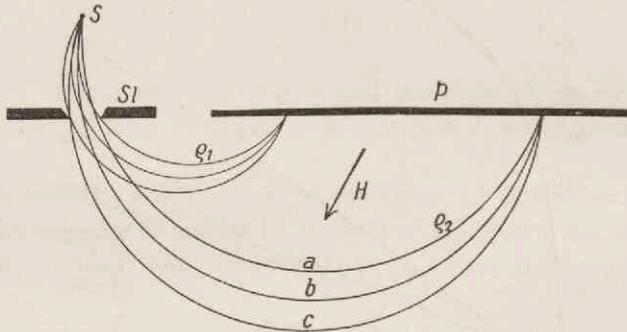


Fig. 8.

Trajectories of electrons in a homogeneous magnetic field, arising from the source S , and falling on the plate P : a , b and c having the same radius.

This property of focussing the electrons makes this method so specially useful. This focussing, however, occurs only in the direction of the dispersion, and not perpendicular to it. If, namely, the same number of electrons describes in one case circles with radius r_1 , in another case circles with radius r_2 , then the intensity falling on the plate per unit of area is in these two cases inversely proportional to the two radii.

The real intensity distribution of such a homogeneous beam on the plate, however, does not consist of a sharp line, because the focussing is not exact and also, because the shape of the source has some influence. Calculations of the real intensity-distribution on the plate, caused by a homogeneous beam,

as a function of the slitwidth, the distance and the shape of the source have been performed by Wooster⁽⁴⁹⁾ and Li⁽⁵²⁾.

If the source emits a beam of electrons, *continuously* distributed over the energies, an analogous reasoning can be applied to this case. Suppose that the distribution of the electrons over the energies is uniform (in a H_Q -scale), then the intensity falling on the plate decreases proportionally to the radius of the corresponding trajectories, owing to the divergence of the beam perpendicular to the dispersion.

The width of the slit, and the position and the shape of the source only influence the range of the energy falling on a given spot of the photographic plate. The magnitude of this range e.g. as a function of the width of the slit can easily be calculated from geometrical considerations, namely in this way, that the difference of the radii of two circles is determined, both passing through the source and the given spot of the plate, and passing near the boundaries of the slit. If the distance of the source to the slit is a , the slitwidth $2b$, the relative range of energy falling on one point of the plate can be expressed as follows:

$$(4) \quad \frac{\Delta(H_Q)}{H_Q} = \frac{b^2}{a^2}$$

With the applied values $a = 1,6$ cm and $b = 0,2$ cm this range is about 1.5% of the proper energy expressed in H_Q .

The indefiniteness of the energy under the influence of the breadth of the source can approximately be expressed by:

$$(5) \quad \frac{\Delta(H_Q)}{H_Q} = \frac{d}{\varrho}$$

where $2d$ is the breadth of the source. For a source of 0.02 cm breadth this gives a range of $\frac{1}{4}\%$ and for a source of 0.15 cm breadth, which was also used, it would give 2% variation of the energy at a ϱ of 4 cm.

Hartree⁽³⁷⁾ has calculated the influence of slight deviations of the homogeneity of the magnetic field on the spectrum. This influence could be neglected in the experiments described here.

B. Homogeneous magnetic and electric field.

A further investigation is necessary in the case that a homogeneous electric field is combined with the magnetic field. The electric field in question is only applied in a small region of the trajectories of the electrons, namely between the source S and the slit Sl (see fig. 9). In a first approximation the influence of the electric field on the shape of the trajectories may be neglected. Then the problem can be treated as if the electrons are coming from the source already with the changed energy.

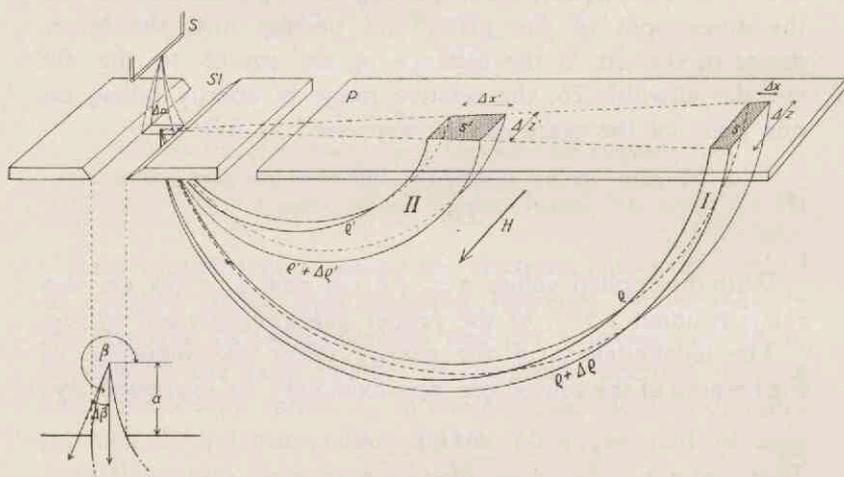


Fig. 9.

Illustration of geometrical circumstances when applying a homogeneous magnetic field perpendicular to the plane of drawing and a homogeneous electric field between S and Sl .

I = beam of original energy.

II = beam of changed energy.

In fig. 9 two electronic beams I and II are represented. It is supposed that they emerge from the source in the same solid angle $\Delta\Omega$, one with the original energy E , the other one changed to an energy E' . They constitute a part of a continuous spectrum. Suppose further as is shown in fig. 9 that the first beam hits the plate P on a certain area s , the other one on an area s' . The density of the plate at the spots s and s' does not only depend on the intensities of the two beams (which are supposed to be the same, see above) but also on the areas which they cover.

The size of the two areas can easily be calculated. In part A of this paragraph it is already shown, that the dimensions Δz and $\Delta z'$ are proportional to the radii, thus

$$(6) \quad \frac{\Delta z'}{\Delta z} = \frac{\varrho'}{\varrho}$$

where ϱ and ϱ' are the average radii belonging to I and II, resp. The dimensions Δx and $\Delta x'$ depend on the intervals of energy of the two beams. Approximately the following expression holds:

$$(7) \quad \frac{\Delta x'}{\Delta x} = \frac{\Delta\varrho'}{\Delta\varrho}$$

where $\Delta\varrho$ and $\Delta\varrho'$ are the maximum differences of the radii in each beam, corresponding to their intervals in energy. The latter, however, are equal, because every electron of the first beam undergoes the same change in energy. From the formulae (2) and (3) the relation follows:

$$(8) \quad \frac{\varrho' \Delta\varrho'}{\varrho \Delta\varrho} = \frac{\frac{E'}{mc^2} + 1}{\frac{E}{mc^2} + 1}$$

On the other hand the ratio of the areas s and s' is given by:

$$(9) \quad \frac{s'}{s} = \frac{\Delta x' \cdot \Delta z'}{\Delta x \cdot \Delta z}$$

so that, from (6), (7) and (8), it follows that:

$$(10) \quad \frac{s'}{s} = \frac{\frac{E'}{mc^2} + 1}{\frac{E}{mc^2} + 1}$$

The following table gives some values of this ratio, which have occurred in the experiments:

E in keV.	25	25	50	100	150	200	300
E' in keV.	20	16	35	70	120	170	270
$\frac{s'}{s}$	0.99	0.98	0.97	0.95	0.95	0.95	0.96

The deformation of the circular trajectories by the electric field has been neglected until now. This neglect should be justified.

The main possible influence is a change of the solid angle $\Delta \Omega$ of the beams, which would cause a change of their intensities.

It is possible to find an approximate expression for this change by calculating the variation of the angle $\Delta \beta$ (see fig. 9) existing at the source between the tangents of the limiting trajectories situated in a plane perpendicular to the magnetic field. In this calculation the equations of motion of an electron in a combined electric and magnetic field have been

used. The result, which is only deduced for low (not relativistic) energies, can simply be written:

$$(11) \quad \frac{\Delta \beta}{\Delta \beta'} = 1 - \frac{\Delta E}{4E}$$

where ΔE means, with a positive sign, an increase of the energy.

Therefore a variation in energy of 20% only causes a variation in the solid angle of 5%. Furthermore the variations given by the formulae (10) and (11) act just in an opposite direction, regarding their photographic effects.

Any deformation of the trajectories, caused by slight deviations of the homogeneity of the electric field (e.g. on account of the slits in the electrodes), could be neglected.

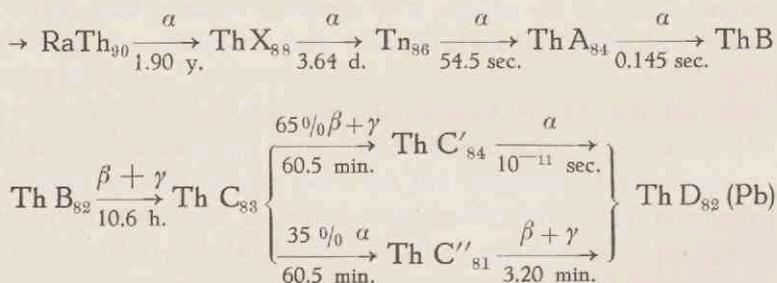
CHAPTER IV.

THE RADIOACTIVE SOURCES.

§ 11. Thorium B, C and C''.

The measurements described in this paper have been done for the greater part with the β ray emitting substances ThB, ThC and ThC''. Before dealing with the properties of these radioactive elements, a survey of the way in which they are obtained from other active substances, and of the relations existing between the different products, will be given.

Starting from the radioactive element Thorium as mother-substance a great number of active elements arises in succession, emitting α , β or γ rays (sometimes a combination of these radiations). One of these decay products is called Radio-Thorium (RaTh). A list of the decay products, arising from RaTh, with their atomic number, the kind of radiation they emit and their half-live periods is given below:



The first β emitting body of this series is ThB, showing a half-period of 10.6 hours. This substance must therefore be obtained from a longer living product. Some RaTh was available for this purpose, possessing a radioactive strength of about 0.2 mC (see § 13).

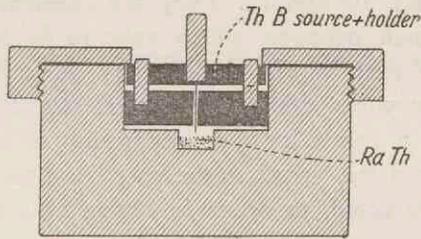


Fig. 10.

Exposure vessel for obtaining ThB, suitable for one source.

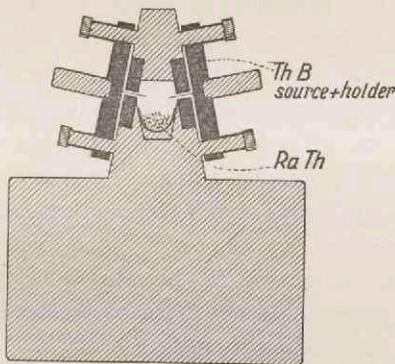


Fig. 11.

Exposure vessel for obtaining ThB, suitable for two sources.

The way in which ThB was obtained from RaTh was the so-called *recoil*-method. In order to activate an object with ThB, it is exposed in a space containing Tn-gas. This

gas escapes continuously from the RaTh, being one of its decay products. The atoms of the decay product of Tn, ThA, attach themselves on the object to be activated. As the half-period of ThA is very short (0.1 sec.), these atoms decay nearly immediately to the following product ThB.

The vessels, used for obtaining ThB in this way, are represented in fig. 10 and 11. They are constructed of brass, except the small grooves, which have to be insulated from the remaining parts of the vessels, and serve for the introduction of the wires, which had to be activated. The first figure shows a vessel for the preparation of a single ThB-source, whereas the vessel, represented in the second figure, has been constructed, in order to obtain two sources at the same time.

The ThB sources were obtained on thin platinum wires fixed on ebonite holders. These wires could be inserted in the small grooves of the described vessels. During exposure to the Tn-gas emanating from the RaTh, they were as near as possible to this substance in order to have a greater yield of activity. During the exposures, the wires always had a potential of -220 volt with respect to the vessel. Reference may be made to the investigations of Gabler⁽²²⁾, regarding the influence of different circumstances, as volume, tension, concentration etc., on the obtained activity.

The RaTh itself was available in a so-called high-emanating form, indicated by Hahn⁽²⁶⁾, and consisting mainly of iron-hydroxide, with which the RaTh is mixed. According to Hahn, it is possible to obtain with these high-emanating preparations an emanating power of 80 0/0, but when these are preserved under dry conditions, a certain ageing sets in, causing after some years a diminution of this power to about 35 0/0.

The activity of the exposed wire does not only depend on the conditions of exposure mentioned above, but also on the length of exposure. It can be assumed, that the number of ThA-atoms (which arises directly from the decaying Tn-

atoms), adhering to the wire, will be proportional to the time of exposure. The number of ThB-atoms originating from the ThA, will, however, not increase unlimitedly, because it decays at the same time according to the exponential law

$$(1) \quad n = n_0 e^{-\lambda t}$$

where λ is the decay-constant of the active substance, related with the half-period T by the formula

$$(2) \quad \lambda = \frac{\ln 2}{T}$$

The β ray activity of the wire as a function of the time t of exposure, is proportional to the number of ThB-atoms, being on the wire. This number can now easily be determined as a function of the time from the differential equation:

$$(3) \quad dn = \nu dt - n \lambda dt$$

ν being the number of ThA-atoms, attaching themselves on the wire per unit of time. The solution of (3) is:

$$(4) \quad n = n_\infty \left(1 - e^{-\lambda t} \right) = n_\infty \left(1 - 2^{-\frac{t}{T}} \right)$$

n_∞ being the number of ThB-atoms present on the wire after an infinite time of exposure.

This relation (4) also gives the activity of the source as a function of the time of exposure. It is represented graphically in fig. 12.

Among the decay products of ThB, two elements exist which also emit β particles. These are ThC and ThC''. ThC is a branching substance, decaying into two separate elements ThC' and ThC'', respectively by β - and α ray emission.

Of the decaying ThC atoms, 65% emit β particles, the other 35% emit α particles. All β emitting products, mentioned above, moreover emit a strong γ radiation.

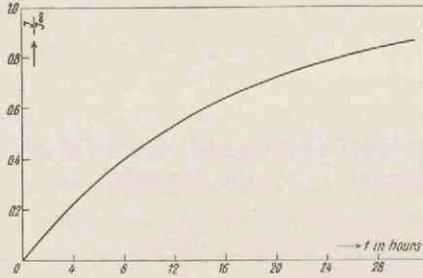


Fig. 12.

The activity J of the ThB source as a function of the time t of exposure.

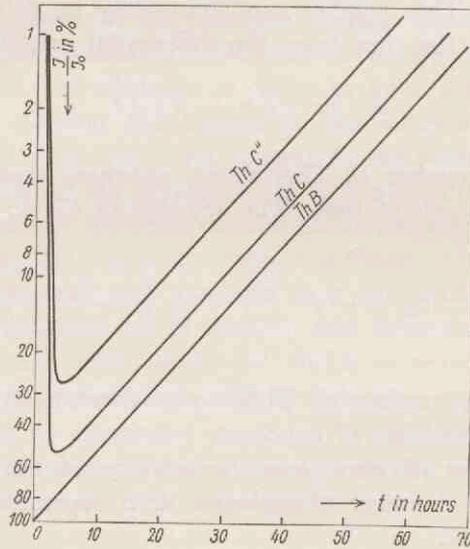


Fig. 13.

Relative radioactive intensities of ThB, C and C'' (in logarithmic scale) as a function of the time t , assuming that at $t = 0$ only ThB is present (with intensity J_0).

Taking the β ray spectrum of a ThB source, in reality a combined spectrum of ThB, ThC and ThC'' is obtained. The ThB source can be considered to give a determined β ray spectrum, if the ratio of the radioactive intensities of the different decay products does not vary with the time.

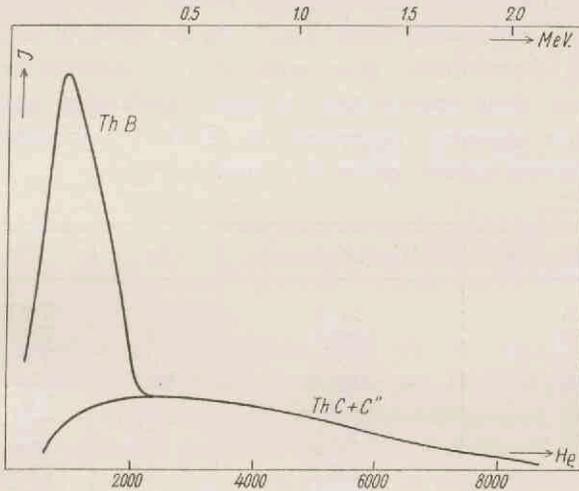


Fig. 14.

Continuous β ray spectra of ThB and ThC + C'', obtained by Gurney (24).

This appears to be the case in consequence of the short half-periods of all these decay products. In fig. 13 these relative intensities are given as a function of the time under the extreme condition that at $t = 0$ ThB should be present without any of the decay products. Even in this extreme case this figure shows, that after about three hours already an equilibrium between the different products exists. As the ThB sources are obtained by exposing the wire to the Tn during several times this period, this equilibrium is already reached during this exposure.

Before proceeding to the experimental results a reproduction

may be given of the continuous spectra of ThB and ThC + C'', obtained by Gurney⁽²⁴⁾ with the charge measuring method, already mentioned. This reproduction shows that in the region of 0—2000 H_Q , the main part of the total radiation originates from ThB (fig. 14).

The characteristic lines are superposed on the continuous spectra. These lines also originate partly from ThB, partly from ThC and partly from ThC''. In the table, given below, the main characteristic lines with their nomenclature, their H_Q -values, their approximate, relative intensities and their origin are inserted, according to Ellis⁽¹⁵⁾ (compare also the improved values, given by Surugue⁽⁴³⁾):

Main char. lines	H_Q (gauss. cm)	Int.	Origin
A	541.0	3	ThC → C''
B	660.8	2	ThC → C''
E	1106.8	3	ThB → C
F	1385.8	200	ThB → C
G	1593.8	3.6	ThC'' → D
H	1691.0	6.6	ThB → C
I	1751.0	25.0	ThB → C
J	1807.7	6.2	ThB → C
L	2603.1	1.4	ThC'' → D
M	2886.6	1.5	ThC'' → D

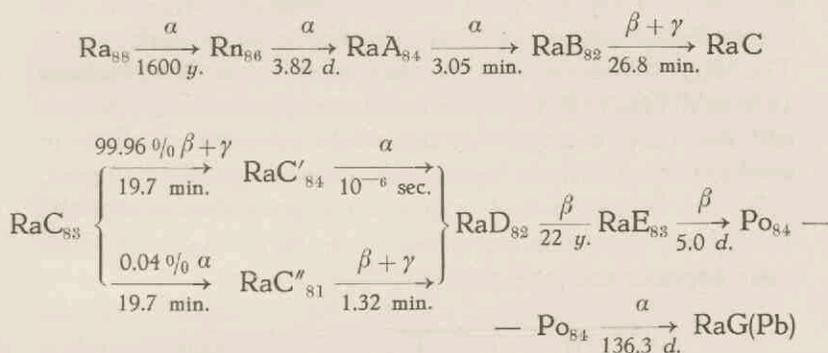
In the reproduction of a photographic β ray spectrum obtained with ThB + C + C'' (fig. 27) five of these lines, namely F, G, H, I, and J can be observed.

§ 12. Radium D and E.

For some measurements a Ra D + E source has been used, because no γ rays are emitted by these bodies and because their β ray spectrum is stronger in certain energetic regions, which were of importance for the intended experiments.

RaD and RaE are members of the Ra-family, which, in its turn, comes from the Uranium-family. Uranium itself is the ultimate mother-substance for this whole sequence. Here

below a list of decay products, starting from Ra, will be given, with their atomic numbers, their half-life periods and the kind of radiation emitted by them:



The usual way of obtaining RaD and its decay products is by using tubes, filled with Rn-gas. The diagram given above shows, that such a tube contains after some years only RaD with its decay products RaE, Po and Pb. Erbacher and Philipp⁽¹⁰⁾ have worked out an electrolytical method for obtaining RaD, RaE and Po in a pure state from these tubes.

Although some Rn-tubes were available in the laboratory, their radioactive strength was not sufficient for the intended experiments. Owing, however, to the kindness of Prof. E. Stahel at Brussels, who lent a preparation of the required strength, it was possible to perform these experiments*).

The preparation, received from Prof. Stahel, has been obtained by him as a cathode-deposit (metallic RaD + Pb) by performing an electrolysis in a HCl- solution.

Here again a few words must be said about the properties of the radiations emitted by RaD and its decay products. RaD

*) The writer wishes to express his gratitude to Prof. Stahel for lending this preparation.

itself emits a β radiation of very low energy namely a continuous one with an energetic upper limit of about 10 keV, and furthermore five characteristic lines which are given in the table at the end of this paragraph. The decay product, RaE, possesses a β ray spectrum of higher energies being only of a continuous nature (no characteristic lines of RaE exist). The upper limit of this continuous spectrum lies at about 1170 keV. Finally RaF must be mentioned, also called Polonium, emitting only α rays. An important property of all these products is, as already mentioned, the absence of γ radiation.

In the following table the five characteristic lines of the RaD β ray spectrum are given with their H_Q -values and their approximate intensities:

H_Q :	600	606	628	714	738
Int.	50	2	0.5	20	10

§ 13. Calibration of the Radioactive Sources.

A. *Relative measurements.*

This paragraph gives a short account of the calibrations of the different radioactive sources, carried out in three ways: by γ -, β - and α ray measurements.

The measurement of the γ radiation, and also in some cases of the β radiation, was performed by determining the ionization current through a condenser, this current being measured by the rate of decrease of the potential difference at this condenser by means of a string electrometer (E d e l m a n n-type).

In this way only relative measurements referring to a standard source could be performed. And even using a standard source certain precautions ought to be taken in consequence of possible differences in ionizing power of the γ - or β rays, even if the sources have the same radioactive strength.

This method has been applied to measure the strength of the RaTh-source, present in the laboratory, and serving for the furnishing of the ThB sources. As a standard source a Radium source of known strength was available. The measurements were performed in each case with $\frac{1}{2}$ cm lead between the source and the condenser, in order to screen off the very soft components of the γ radiation. The distances of the Ra and the RaTh sources to the condenser were adjusted in such a way that the rate of ionization was the same. From the ratio of the distances, the strength of the RaTh-source could be deduced and was found to be 0,2 mC (see § 11). It was necessary to put the RaTh source under an air-tight glass cover in order to avoid infection of the apparatus by the escaping Tn-gas.

Also comparative measurements on the ThB sources themselves were performed. In this case the β rays as well as the γ rays, emitted from these sources, were measured with this method at the same time. These measurements were mainly performed in order to investigate the influence exerted on the activity of these sources by applying different conditions of exposure to the Tn-gas.

B. *Absolute measurements.*

Absolute measurements have been performed on some of the ThB sources, in order to obtain the number of emitted β particles (necessary for absolute data about the photographic plate (see § 20)).

Two methods have been applied for these measurements. Firstly the absolute intensity of the β rays, and secondly that of the α rays have been determined.

The β ray measurements have been performed by means of an experimental arrangement, described by Ornstein, Milatz, ten Kate and Miesowicz⁽³⁴⁾ consisting essentially of an electromagnet without iron, in which could be placed the radioactive source and a Geiger-Müller-

counter, enabling the measurement of the β ray spectra in the usual way. Because these same authors had investigated the efficiency of the applied counter as a function of the energy of the particles falling on this counter, it was possible to deduce from the counted number of particles the real one. In order to obtain the whole number of emitted particles it was necessary to integrate over the whole β ray spectrum. For this integration the measurements of Gurney⁽²⁴⁾ have been used. At two or three energies the numbers of electrons have been determined in the way mentioned, and combining these values with the knowledge of the form of the spectrum given by Gurney (see fig. 14), the total number can be obtained taking account of the solid angle (determined by some limiting slits) and of the dispersion (determined by the width of the slit in the counter). The ThB-sources being exposed only during a finite time to the Tn-gas, the value found for the total number of emitted particles ought to be corrected, with the help of fig. 12, in order to obtain this value for an infinite time of exposure. The final result was that the maximum attainable strength for these ThB-sources, exposed in the vessel indicated in fig. 11, amounted to

$$9.3 \times 10^6 \text{ electrons per second,}$$

being equivalent to about 0.12 mC of ThB alone.

Secondly the absolute measurement of the α radiation was performed by Mr. C. v. Heerden and Mr. K. J. Keller with an experimental arrangement, consisting of an ionization-chamber with amplifier. The number of particles was determined by recording the deflections of a galvanometer. In this way it was even possible to distinguish between the particles, emitted by ThC, and those emitted by ThC', by their difference in energy. The result was that the total number of α particles of a ThB-source, again corrected to an infinite time of exposure amounted to

$$2.5 \times 10^4 \alpha \text{ particles per second}$$

being equivalent to about 0.07 mC of ThB alone (the number

of α particles emitted by a ThB-source had to be one half of the number of β particles, as can be seen from the representation of the genetic dependence of the decay products, given in the beginning of § 11).

Taking into consideration the strength of the RaTh-source which was found to be 0,2 mC, the preceding results give an emanating power of about 50 % being in good agreement with the expected emanating power of this source (see § 11).

CHAPTER V.

MEASUREMENTS.

§ 14. Experimental Procedure.

When working with ThB sources, it was necessary to replace the source before every new exposure by one freshly exposed to the RaTh. The adjustment in the apparatus was obtained by two pins, which fitted in the sourceholder. The source had to be connected electrically to the outside to make possible the application of electric tensions. Working with RaD the source could be adjusted once for all.

As photographic plates *Ilford* „Special Rapid, extra sensitive, 400 H & D” were applied, which, compared with other plates, showed to be most suited for the intended experiments.

The applied plates had to be cut into the required shape. Special care was taken to avoid any possibility of fogging, e.g. by pressure or by light, during the cutting. In the room only a feeble red lamp was present when the plates were introduced into the spectrograph. The apparatus was then shielded off from light by black cloths. It was exhausted by a mercury diffusion pump to a pressure of less than 10^{-6} mm. When the vacuum was sufficient, the magnetic field was switched on and the exposure could begin.

In order to obtain the density curve of a plate, the intensity of the radiation falling on the plate must be varied in a known way. In the case of the ThB sources the decay

of these sources could be used for this purpose. Exposing the plate during a certain time to the source, and doing the same after 10.6 hours with another part of the plate the ratio of the intensities on these two exposures would just have been one half. Taking several exposures with constant periods of exposure, the intensity is always a known fraction of that of the first exposure.

In the case of the RaD source, the variation in density has been obtained by varying the length of exposure. In this case the validity of the reciprocity-law must be assumed. If now in both cases the density curves are identical, the validity of this reciprocity law is proved.

In the described way a series of photographic β ray spectra is obtained with different known intensities, from which the density curves at every desired energy of the spectrum can be obtained.

As the density curves are obtained from a series of different photographs it was necessary to use neighbouring pieces of the same photographic plate and further all the plates ought to be developed under identical circumstances.

As is explained in the preceding chapters, the sensitivity curve of the plate is obtained by taking two photographic spectra with the same intensity: one in the usual way, the other by applying a retarding electric potential. These requirements have been fulfilled by alternately switching the tension on and off, so that the total intensities at the two exposures are equal.

This procedure is simple in the case of RaD; the lengths of the exposures with and without the tension have only to be taken the same. For the ThB-sources, however, the decay of the source has to be taken into account. With the aid of a calculated correction curve (fig. 15), one can determine for each time t_1 of exposure without tension, the corresponding time t_2 of subsequent exposure with tension, giving the same intensity.

Taking a photographic spectrum with and without changed energies merely the primary circuit of the transformer must be switched on and off and, at the same time, the screen Sc (fig. 4) must be turned, in order to expose the other half of the plate.

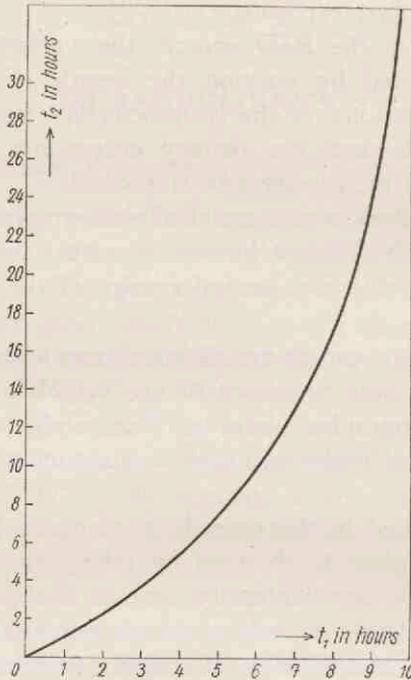


Fig. 15.

Correction curve for the decay of ThB giving at each period t_1 the subsequent period t_2 of exposure to a ThB source, corresponding to equal intensities (see text).

During each exposure the magnetic and electric field were regularly controlled by observing the deflection of the galvanometer.

As developer metholborax has been used, the development

taking place during about $5\frac{1}{2}$ minutes at room temperature. Plates which had to be mutually referred to, were fixed together on one glass plate and then treated as a single photographic plate.

The densities of the plates have been determined by Moll's microphotometer and were compared to an unexposed part of the plate (which had not even been in the spectrograph). A special device was constructed enabling to adjust at the same time the unexposed and the exposed plate in the photometer.

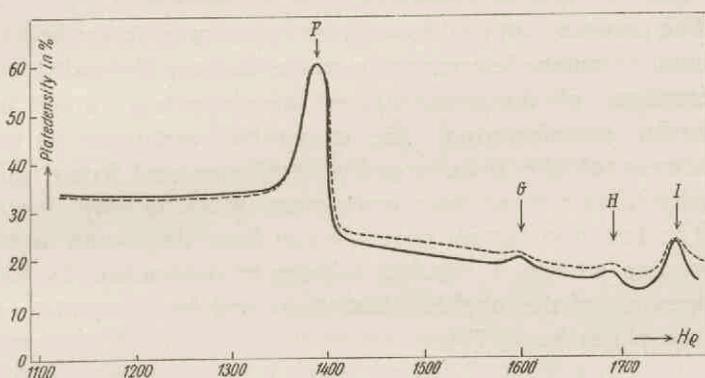


Fig. 16.

Example of working out the photograms. Solid line indicates the direct photographic spectrum, the broken line the retarded spectrum of ThB, C and C'.

The photograms, obtained with the photometer, were measured out and drawn in a diagram, as indicated in fig. 16. The corresponding values of the energies are deduced by comparing the energies of the characteristic lines with those, given by Ellis⁽¹⁵⁾ (see § 11, resp. § 12). The retarded spectrum which comes on the plate in a shifted position, is represented in these diagrams with the original energies as abscissae. The two spectra are in this way directly comparable. The further elaboration has already been dealt with in the second chapter.

§ 15. Sources of Error and their Elimination.

In this paragraph a brief enumeration of the causes of errors of the method under consideration will be given and the possibilities of eliminating them. As has already been remarked in the introduction the possible causes of deformation in the spectra themselves have not been investigated. A discussion of the accuracy of the results will be found in § 20.

A. *Errors inherent to the photographic method.*

The accuracy, attainable with the photographic method in general, is mainly determined by three factors: the individual fluctuations of the properties of the plates (e.g. caused by irregular manufacturing), the unavoidable variations in the treatment of the plates e.g. by developing and fixing; and thirdly the errors of the measurement of the density.

The first two factors have already been dealt with in the preceding paragraph, the third factor is determined by the properties of the applied photometer and by the graininess of the plates (see § 20).

B. *Errors caused by the applied apparatus.*

These errors are partly caused by fluctuations in the electric tensions, partly by geometric conditions. Their main influence has already been discussed in the third chapter. Only one possible source of errors might still be emphasized, namely that of partly shielding-off the β rays by the intermediate slits. This shielding would cause an alteration of the distribution of the intensity over the spectrum. The electrical conditions did not allow to choose these slits very wide. In order to be as free as possible from this error the slits have been adjusted with special care.

C. *Errors caused by stray- and secondary radiation.*

The influence of errors of this type only plays a part, when the real form of the β ray spectra is required. It has,

however, no effect on the applicability of the method given here, at least, as long as the intensity of the stray radiation is small in comparison with that of the radiation to be measured.

The density curves being automatically taken under the same circumstances, in which the spectra themselves are photographed, the true relative numbers of electrons are directly obtained from the densities.

The determination of the sensitivity curve is also independent of stray-, or secondary- or γ radiation, because it may be assumed that by taking a spectrum with- and without electric tension the intensity of these spurious radiations will not be altered. It is therefore allowed to compare the densities of the plate in these cases.

These arguments are only true when the intensity of the investigated electronic radiation is large in comparison with these effects. For this reason when using ThB sources, the method could only be applied with the described apparatus in the energy region between 60 and 200 keV. It was, however, desirable to extend the measurements by means of a RaD source giving a much weaker γ radiation.

D. Errors caused by absorption and reflexion in the sources.

These errors too effect only the spectrum itself. The investigations of Flammersfeld⁽²¹⁾ on the β ray spectrum of RaE have shown the great importance of the properties of the bearer of the source on the value of this type of errors.

§ 16. Experimental Results concerning the Density Curves.

In fig. 17 the curves obtained with ThB, C and C'' are reproduced. They are obtained, as has already been remarked, with one constant period of exposure of about 10 hours. This figure shows that the shape of the curves is the same for different energies of the electrons between 90 and 240 keV.

The density curve in fig. 17 has been drawn on a logarithmic intensity scale. In a numerical scale (see fig 18) the part of

the curve near the origin has a proportional character (compare e.g. § 3).

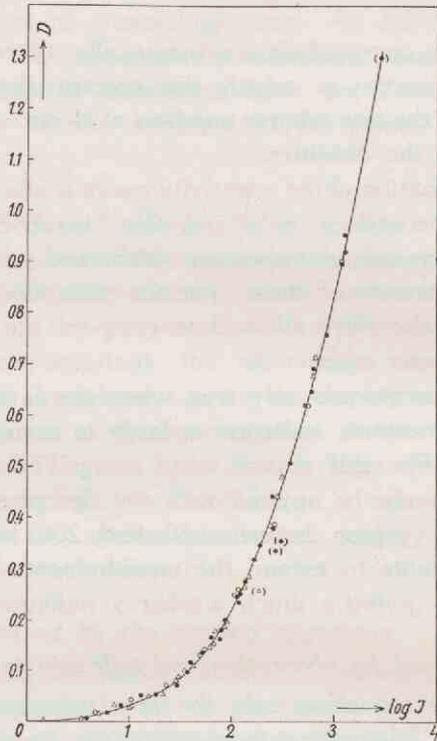


Fig. 17.

Density curves obtained with ThB, C and C'' for different electronic energies:

○ 90–115 keV. ● 115–170 keV. △ 170–240 keV.

Finally density curves have been obtained with the RaD source, for energies from 20 to 170 keV (varying the time of exposure). In fig. 19 the solid line has been copied from fig. 17, and the open circles indicate the points, belonging to the density curves of RaD + E, which appear again to be independent of the energies. This figure shows moreover

the important result, that the curves coincide with those obtained with ThB.

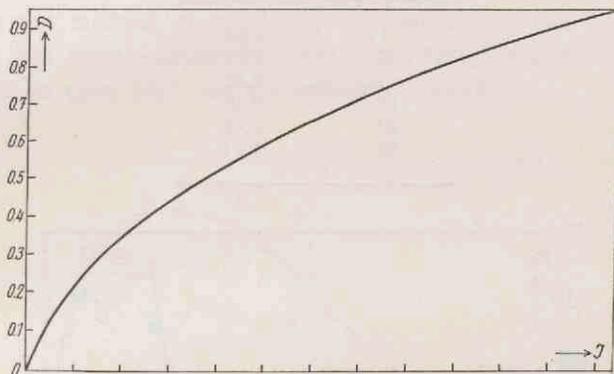


Fig. 18

The same density curve as on fig. 17 but on a numerical scale in the relative intensity.

From the latter fact something can be deduced about the validity of the reciprocity-law (see § 3). A deviation of the reciprocity-law is often characterized by a constant p , the so-called Schwarzschild-exponent, assuming that the density is not a unique function of $i \cdot t$, but of $i \cdot t^p$.

From fig. 19 a limit for the value of p can be deduced by calculating the deviation between the density curves of ThB and RaD for some values of p (in fig. 19 some points of the curves for $p = 1.1$ and $p = 1.2$ are indicated by black circles). From the position of the measured points (open circles) it is seen that the value of p lies certainly between 1.0 and 1.1 *).

In conclusion a short numerical extract of the density curves, obtained in the reported experiments is given in the following table:

*) Recent experiments performed by Mr. T. Tol with a cathode ray tube, show, when they are compared with those mentioned above, that the value of p is equal to one with a possible error much less than 10%.

Rel. Int.	Density
1	0.10
2	0.17
5	0.32
10	0.50
20	0.72
50	1.13

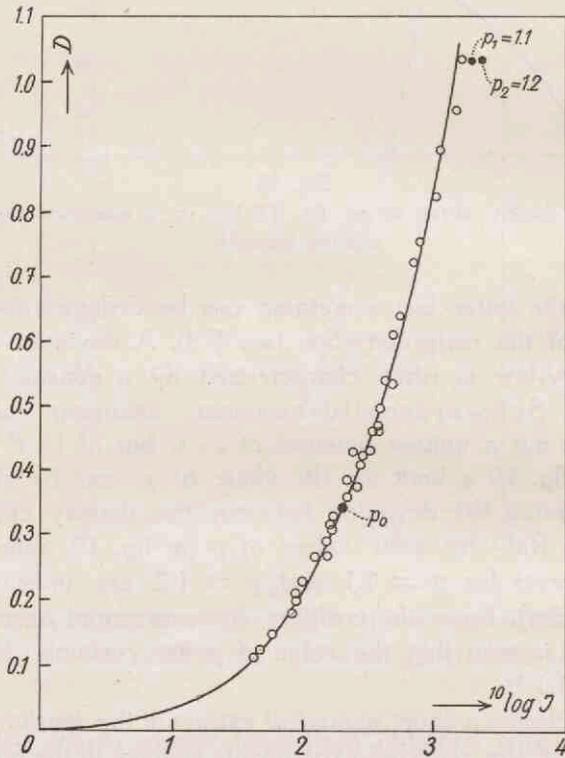


Fig. 19.

- Density curve obtained with ThB (see fig. 17).
- Measured points of the density curve, obtained with RaD, for energies ranging from 20–170 keV.
- Indicating the position of the density curve for RaD, which would be found if $p = 1.1$ or 1.2.

§ 17. Results concerning the Sensitivity of the Plate at different Energies; the β Ray Spectrum.

As the method of obtaining the sensitivity curve of the plate has been explained in the foregoing paragraphs, it may suffice to give here the experimental results.

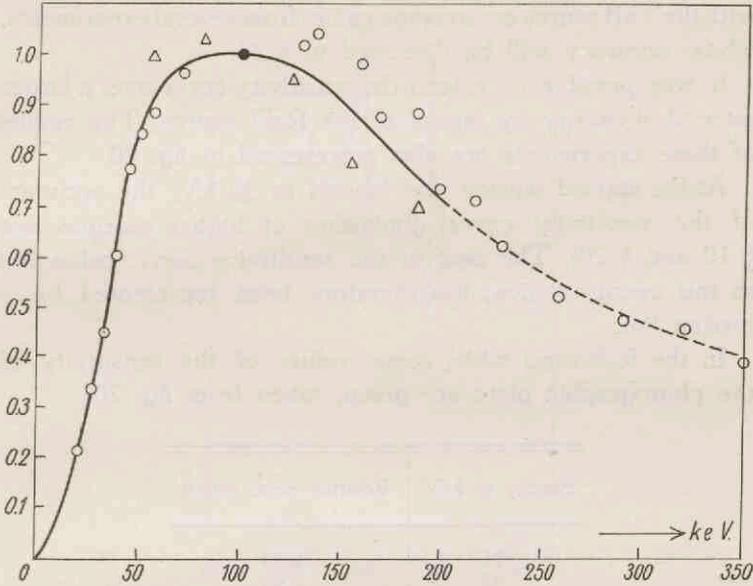


Fig. 20.

Sensitivity curve for the photographic plate, giving the relative photographic action of electrons as a function of the energy.

○ Obtained with RaD + E. △ Obtained with ThB + C + C'' (average values).

In fig. 20 this sensitivity curve is represented. The value belonging to a certain ordinate at a certain energy E in this figure indicates the ratio of the intensities J_1 (at 100 keV) and J_2 (at the energy E) causing both the same density on the plate.

The first experiments have been done with ThB-sources, but as fig. 20 shows, only a relatively small energy interval could be used (from 60 to 200 keV) because the intensity of the ThB spectrum decreases rapidly at both sides of this interval, and, moreover, because the effect of the γ radiation on the plate becomes comparable with that of the electrons themselves (compare § 15). The points indicating the measurements with the ThB source are average values from several experiments, whose accuracy will be discussed in § 20.

It was possible to extend the sensitivity curve over a larger interval of energy by means of the RaD source. The results of these experiments are also represented in fig. 20.

As the applied tension was limited to 30 kV, the accuracy of the sensitivity curve diminishes at higher energies (see § 10 and § 20). The part of the sensitivity curve, belonging to this energy region, has therefore been represented by a broken line.

In the following table, some values of the sensitivity of the photographic plate are given, taken from fig. 20:

Energy in keV	Relative phot. action
25	0.30
37	0.56
50	0.84
75	0.99
100	1.00
125	0.98
150	0.91
200	0.71
250	(0.57)
300	(0.47)

Using the properties of the plate, given in this chapter, the relative intensity distribution of the spectrum of ThB + C + C'' has been obtained and is represented in Fig. 21.

This spectrum is given in a H_0 -scale (each ordinate giving

the intensity in a certain constant H_Q -interval). This scale is obtained from the relative intensities on the plate by multiplying each intensity by the radius of the corresponding trajectory (see § 10).

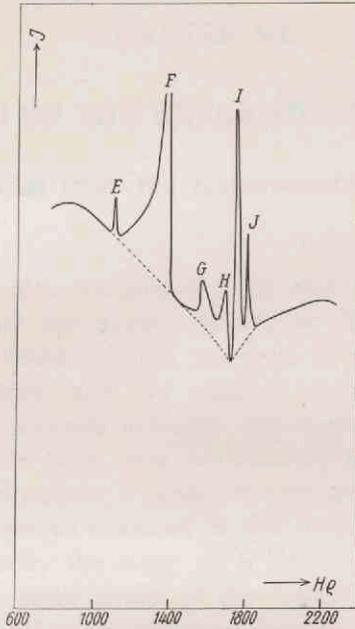


Fig. 21.

Relative intensity distribution of the ThB + C + C'' spectrum (in a H_Q -scale), measured between 800 and 2300 H_Q .

The spectrum of RaD + E is not represented because the source used for the experiments consisted of a rather broad strip (0.15 cm), and therefore a distortion of the obtained spectrum was to be feared. On the other hand there is no objection to use this source for the investigation of the reciprocity law and of the sensitivity curve.

In this connection it may be remarked that the photographic

properties of the plate can be obtained independently of the measuring of the spectra, so that, e.g. it is possible to use different spectrographs or different sources for the calibration of the plate and for the measurement of the spectra, provided that pieces of the same plate are used.

CHAPTER VI.

DISCUSSION OF THE EXPERIMENTAL RESULTS.

§ 18. Conclusions from the Experiments on the Density Curves.

The main results obtained in § 16 are: the independence of the shape of the density curves of the energy of the electrons, the linear relation between density and intensity for low intensities, and the validity of the reciprocity law within the experimental accuracy (see figs. 17, 18 and 19). Analogous results have been obtained by other investigators (compare § 5). Moreover it appeared that generally the shape of the density curves obtained in this thesis and that of other authors are exactly the same, as is shown in fig 22.

In this figure some points of the density curves published by Ellis and Wooster⁽¹⁸⁾ for electrons having energies from 150 to 1000 keV are given, obtained with photographic X ray Ilford plates. According to these authors the curves obtained under different conditions of development and at different energies of the electrons can be represented by the well known formula, already given by Silberstein⁽⁴³⁾ and Busé⁽¹⁰⁾:

$$D = C \log\left(\frac{J}{\tau} + 1\right)$$

J being the total intensity, and C and τ parameters. The equality of the shape of the density curves under the mentioned conditions implies the constancy of C in this formula.

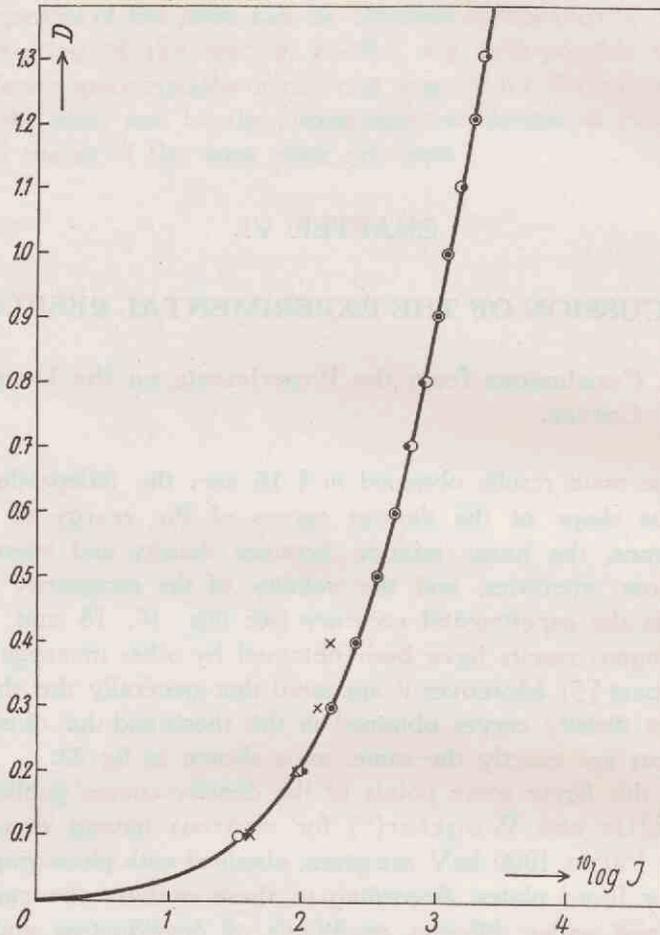


Fig. 22.

- The density curve obtained in § 16.
- Density curve obtained by Bouwers⁽⁷⁾ with Röntgen rays.
- Density curve obtained by Ellis and Wooster⁽¹⁸⁾ with electrons of 150 — 1000 keV and with Ilford X-ray Plates.
- × Density curve obtained by Salbach(1922) with electrons of heterogeneous energy.

Reasons exist (compare § 3) to suppose, that not only electrons of different energies, but also other kinds of radiation

will act in the same way. To check this assumption, the density curve obtained by Bouwers⁽⁷⁾ for Röntgen rays of about 1 \AA with Ilford Special Rapid plates has been compared with that, obtained in § 16. The data of Bouwers are also represented in fig 22. Even in the case of Röntgen rays a perfect similarity between the different density curves exists.

In fig. 22, also some points of a curve obtained by Salbach (1922) for electrons of heterogeneous energies of radioactive origin are indicated, showing a deviation from the other curves. The cause of this deviation might be the use of other types of photographic plates or the different way of measuring the density [Callier⁽¹¹⁾].

In § 5 it has been remarked that the results of the experiments of Bories and Knoll⁽⁶⁾ do not agree with those of others. They find e.g. a strong dependence of the shape of the density curves on the energy. It has therefore no sense to compare numerically their curves with those obtained in this thesis. One may, however, point to the fact that the density curves obtained by Mr. T. Tol with a cathode ray tube in this laboratory (serving for electronic diffraction experiments), agree very well with those in fig. 22 (for the same type of plates), although the periods of exposure are much shorter, being of the order of $\frac{1}{2}$ sec. (In the experiments of Bories and Knoll, these periods ranged from 10^{-2} to 10^{-8} sec.).

§ 19. Discussion of the Sensitivity Curve of the Photographic Plate.

To begin with a qualitative interpretation of the shape of the sensitivity curve, represented in fig. 20, will be given. This curve consists of an ascending and a descending part.

The ascending part, corresponding with electronic energies below 100 keV, can be explained by the fact, that their range is less than the thickness of the photographic emulsion. So long as the range of the electrons is of the given order the

effect on the plate will increase with increasing energy, the energy, absorbed by the plate, being equal to the whole energy of the electrons themselves.

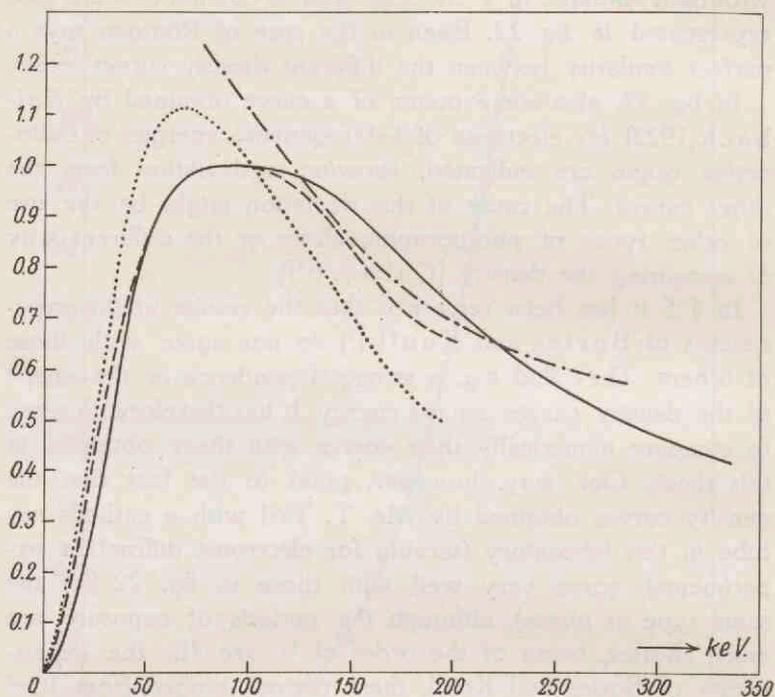


Fig. 23.

- Sensitivity curve experimentally obtained and represented in fig. 20
- . - . - . Curve, giving the proportional dependance with $1/\beta^2$
- Curve, obtained from absorption measurements of Lenard⁽³¹⁾.
- - - - - Curve, deduced from the same measurements of Lenard, taking into account the reflection on the glass.

The descending part of the sensitivity curve, corresponding to the energies of those electrons, which are able to pass through the photographic emulsion, can be explained by the fact that the loss of energy of the electrons in the plate

decreases with increasing energy. An approximate expression for this decrease (already used by Ellis and Wooster⁽¹⁸⁾), is obtained assuming that the ionization of an electron on a certain length of its path is inversely proportional to the square of its velocity. In fig. 23 the sensitivity curve calculated on this basis is given together with the experimental one. The calculated curve has only sense, when the electrons pass through the emulsion, therefore only for the energies corresponding to the descending part of the experimental curve.

In this connection the measurements of Lenard⁽³¹⁾ are of importance, concerning the determination of the „practical absorption” of the electrons in thin sheets of material, which is defined by the apparent decrease of the number of electrons in passing through matter. In fig. 24 the mass-absorption coefficients $\frac{\mu}{\rho}$ obtained by Lenard, are plotted as a function of the energy.

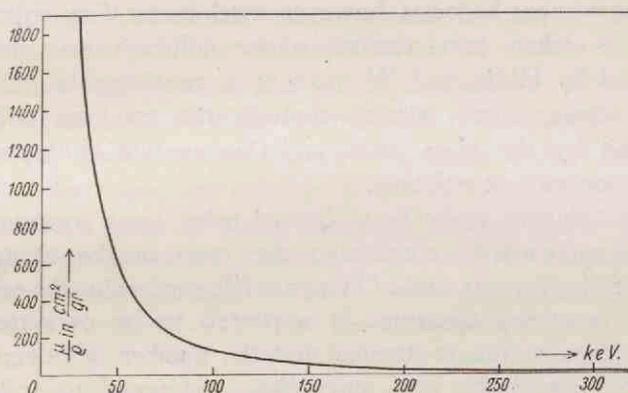


Fig. 24.

Practical mass-absorption coefficient μ/ρ of the electrons as a function of their energy, measured by Lenard⁽³¹⁾.

From this curve, the number of electrons absorbed in the photographic emulsion can be calculated if the thickness and

the specific weight of the emulsion are known. In this way the average energy absorbed by the photographic plate per electron of given primary energy, is obtained. The electronic energy being E , the absorption coefficient at this energy being $\mu(E)$, and the thickness of the emulsion d , the average energy F absorbed in the plate per electron amounts to:

$$(1) \quad F(E) = E(1 - e^{-d\mu(E)})$$

Using fig. 24, the quantity F can be calculated as a function of the energy. Assuming that the photographic sensitivity will be proportional to the absorbed energy, the function F gives at the same time the sensitivity function. It is represented in fig. 23 by the dotted curve. The following numerical data concerning the photographic plate have been used for this calculation:

Measured thickness of the emulsion $d = 15 \times 10^{-4}$ cm.

Spec. weight, corresponding to 40% AgBr $\rho = 3.2$ gr./cm³.

The curves show that only a qualitative agreement exists. This agreement becomes, however, much better if an important factor is taken into account whose influence was already surmised by Ellis and Wooster: a considerable part of the electrons after passing through the emulsion will be reflected by the glass plate, and thus exert a photographic action for the second time.

This influence could be estimated using some experiments of Schonland⁽⁴⁰⁾, concerning the ratio of the electrons, scattered backwards, and of Wagner⁽⁴⁶⁾, concerning the energy of the scattered electrons. It appeared to be permitted in first approximation to assume that the number of electrons, scattered back from the glass plate, amounted to 16% of the electrons, falling on the glass, and that their average energy amounted to 80% of the primary energy (in both cases these percentages appear to be approximately independent of the energies of the electrons). In order to take into account that the scattered electrons re-penetrate the emulsion in all possible directions, it was assumed, that they had to

penetrate in this case an emulsion of the double thickness (corresponding to a cosine distribution of the scattered electrons). Moreover, the primary loss in energy of the electrons, penetrating the emulsion (which was not taken into account in the expression (1)) has been assumed to be 5×10^6 eV/cm (Lenard⁽³¹⁾ *).

A possible photographic action of secondary Röntgen rays could completely be neglected.

The result of all these considerations is given by the broken curve in fig. 23, which is in striking agreement with the experimental results, at least for the lower energies.

§ 20. The Accuracy of the Method.

The principal sources of error have been treated in § 15. In the present paragraph the value of some of the errors, and of their effect on the accuracy of the method will be given.

The accuracy is mainly determined by that of the photographic procedure, and regarding the sensitivity curve, also by some geometrical and constructional factors.

The accuracy of the measurement of the density depends on the microphotometer and on the „graininess” of the plate. A distinction must be made between the determination of the „absolute” density and of relative differences in the densities. In the first case, the adjustment of the microphotometer is the essential cause of error (the image of the illuminated spot of the plate has to be adjusted on the thermo-element). By repeating an adjustment several times, it appeared that a maximum variation of 2% in the experimental value of the absolute density can occur. As each point of the density curves corresponds to separate pieces of a plate and thus to separate adjustments, an error of the given magnitude can therefore be expected.

If, however, only differences in densities on the same plate have to be determined, as is the case in obtaining the sen-

*) Compare e.g. Landolt-Börnstein Hptw. II, page 890.

sitivity curve (see e.g. fig 16), the error in the adjustment is eliminated.

Besides the error, caused by the adjustment of the photometer, the accuracy is determined by the graininess of the plate. A photograph of a part of a plate obtained with the microphotometer does not consist of a smooth line, but of a zigzag line, as is illustrated by fig. 25. This zigzag line is caused by the fact that the number of developed grains, present at the spot where the density is measured, shows statistical fluctuations.

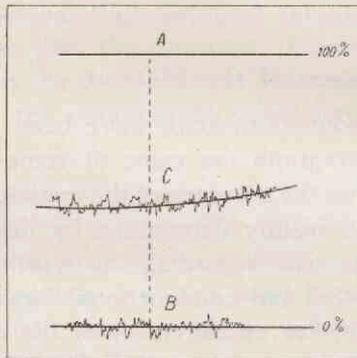


Fig. 25.

Part of a photogram illustrating the influence of the graininess of the plate.

Investigations about the graininess of the photographic plate have been performed by v. Kreveld⁽²⁹⁻³⁰⁾. This author defines a quantity ΔD , the fluctuation of the density, as follows: if the total area of the fluctuations of the zigzag line, divided by the length of that line, is δ (average deviation), and the density D being

$$(1) \quad D = {}^{10}\log \frac{AC}{AB}$$

(see fig. 25), then

$$(2) \quad \Delta D = \delta \frac{0.434}{AC}$$

This quantity ΔD depends on the area O of the image of the first slit formed by the microphotometer on the plate. It could be shown by the mentioned author that the quantity

$$(3) \quad \Delta K = \Delta D \times \sqrt{O}$$

is independent of O . It is called the „absolute graininess“. This quantity has the dimension of a length, and its value is under the circumstances considered here:

$$(4) \quad \Delta K = 7 \times 10^{-5} \text{ cm} \quad \text{at } D = 0.5$$

In order to calculate the accuracy in the real intensities, it is necessary to take into account the density curves. This is accomplished by the mentioned author by defining a quantity γ_d (differential gradation):

$$(5) \quad \gamma_d = \frac{dD}{d \log J}$$

being the differential quotient of the density to the logarithmic intensity (e.g. the slope of the curve in an arbitrary point of fig. 17). Now the expression

$$(6) \quad \frac{\Delta D}{\gamma_d} = 0.43 \frac{\Delta J}{J}$$

gives the relative accuracy corresponding to a given uncertainty in the density. In the same way, the quantity

$$(7) \quad \frac{\Delta K}{\gamma_d} = 0.43 \frac{\Delta J}{J} \sqrt{O}$$

appears to be useful. This quantity is not only approximately constant for different areas O of the image in the microphotometer, but also for all densities ranging from 0.2–1.0 (ΔK and γ_d depend approximately in the same way on the density in this region). It depends, however, on the type of the plate, on the conditions of development and on the kind and energy

of the radiation. The value of $\frac{\Delta K}{\gamma_d}$ appears to amount approximately to

$$(8) \quad \frac{\Delta K}{\gamma_d} = 10^{-4} \text{ cm}$$

Using the value (8), (7) can be written:

$$(9) \quad \frac{\Delta J}{J} = \frac{2.3 \times 10^{-4}}{\sqrt{O}} \quad (O \text{ in cm}^2)$$

This formula shows that the accuracy can be augmented by using a larger image of the slit of the microphotometer on the plate. The efficient size of this image was limited by the photometer and by the condition that the plate-density inside this image ought to be homogeneous.

The absolute number of electrons, necessary to effect a given density, has been determined with the aid of the apparatus designed by Ornstein, Milatz, ten Kate and Miesowicz^(8,4), already mentioned in § 13. This determination was possible by the known efficiency of the counter used, this efficiency having been investigated by the mentioned authors. The number N of electrons falling on an area of 1 cm^2 of the plate, and causing a density of 0.5, was found to be: *)

$$(10) \quad N = 6 \times 10^7 \text{ e/cm}^2 \quad \text{for } D = 0.5 \text{ and for electrons of } 100 \text{ keV.}$$

The statistical fluctuation ΔN in the number N of electrons is given by the formula:

$$(11) \quad \frac{\Delta N}{N} = \frac{1}{\sqrt{N}}$$

*) This result appeared to be in accordance with a determination of this number by Mr. T. Tol in this laboratory, using a cathode-ray tube.

This fluctuation becomes for the value of N , given in (10):

$$(12) \quad \frac{\Delta N}{N} = 1.6 \times 10^{-4} \quad \text{at } D = 0.5$$

If an area O of the plate is used for the determination of the density, the number of electrons, falling on this area is

$$(13) \quad n = NO$$

The relative statistical fluctuation in this number is, using (12):

$$(14) \quad \frac{\Delta n}{n} = \frac{1.6 \times 10^{-4}}{\sqrt{O}} \quad \begin{array}{l} \text{at } D = 0.5 \\ (O \text{ in cm}^2) \end{array}$$

Comparing (9) and (14), it appears that *the fluctuations in the photograms are not caused exclusively by the photometer or by the photographic errors in general but that they are for a considerable part of an essential character, connected with the radiation to be measured.* These fluctuations cannot therefore be eliminated by improving the photometer or the photographic plate. It would e.g. be possible to use plates with smaller grains. These plates being necessarily less sensitive, no advantage is reached. This highest accuracy is only obtained for electrons of the most favourable energies, that are energies between 70 and 150 keV for the Ilford plates of the type Special Rapid, Extra Sensitive (400 H & D).

Continuing the discussion regarding the accuracy of the sensitivity curve, a remark of a more geometrical kind must be made. The sensitivity curve for a given range of energies is generally obtained by a number of steps in energy, as described in the second chapter. The magnitude of the steps giving subsequent points of the sensitivity curve, is limited because the geometrical corrections become of importance for larger steps (see § 10) and these cannot yet be calculated exactly. Now the error made with one step influences all points of the sensitivity curve corresponding to the following steps, so that the error will augment proportionally to the

number of steps. For a future development of this method it will therefore be necessary either to find out a method of obtaining these corrections more exactly, so that the number of steps can be diminished, or to diminish the error for each separate step.

The error made for each step is exclusively determined by the graininess of the plate. This error depends on the average area O on the plate, suitable for the measurement of the density at an arbitrary point. The area used in the experiments, reported here, amounted to $75 \times 10^{-6} \text{ cm}^2$. From formula (9), the relative accuracy in the intensity corresponding to this area, appears to be

$$(15) \quad \frac{\Delta J}{J} = 0.027$$

The accuracy of every point in the sensitivity curve can be calculated, using (15). This accuracy is not yet very high because the relative intensity differences between two successive points are generally rather small (see fig. 20), so that the error in these differences will be larger. Furthermore the part belonging to the higher energies (indicated by a broken line in fig. 20) is still less reliable owing to the great number of steps in this region.

The given accuracy can be augmented by using a larger area O , necessitating an improvement of the photometer and of the photographic β ray spectra.

CONCLUSION.

Surveying the results obtained in the preceding chapters it can be concluded that the photographic method proposed is practically applicable and moreover appears to be rather simple. This simplicity is partly due to the fact that the density curves coincide for all energies investigated.

There exist, however, still some difficulties, partly of a technical kind (e.g. the application of high tensions) and partly of a geometrical character (the exact calculation of the deformations of the trajectories, etc.). Further investigations will surely show that it is possible to overcome these difficulties.

For strong preparations the photographic method appears to be of the same sensitivity as that of a counter. The method has then, however, the advantage that large parts of a spectrum can be investigated simultaneously.

For weak preparations, however, the sensitivity of the photographic plate is rather low which is then a serious disadvantage of the photographic method. An effect being easily detectable by a counter (e.g. one electron per second per cm^2) would require a photographic exposure of about 1000 hours.

It may still be pointed out that the application of electric fields, as used in this thesis, offers some more general prospects. For instance the investigation of the part of lowest energy in the β ray spectrum could be performed with more profit by applying an accelerating tension on the β particles.

Not only a calibration of the photographic plate, but in principle the sensitivity of any detecting instrument can be obtained with the method described in this thesis.

May the given method contribute to the furtherance of some problems in atomic and nuclear physics.

SUMMARY.

First a review of existing experimental methods for the investigation of the β ray spectrum is given. A photographic method for the measurement of β ray spectra is then described. In this method the density curves are obtained by variation of the strength of the source and the sensitivity curve by changing the energy of the β particles. A description of the experimental arrangement and of the properties of the radioactive sources used is given. The density and sensitivity curves for the type of plate used are discussed. The density curves appear to be independent of the energies, and the Schwarzschild-exponent appears to be equal to one, within the experimental accuracy. A further discussion showed, that the density curves show a perfect similarity with those obtained by other investigators not only for electrons, but also for Röntgen rays. The shape of the sensitivity curve can be explained with the aid of the absorption measurements of Lenard, and the reflection measurements of Schonland and Wagner. A β ray spectrum of ThB + C + C", obtained with the method described, is reproduced. Finally a discussion of the accuracy of the method is given.

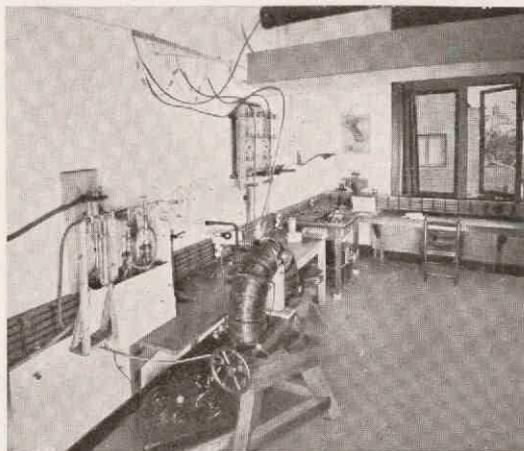


Fig. 26.

The experimental Arrangement.

On the left the Dubois-magnet with the β spectrograph and the mercury diffusion pump. Near the window the electrical and controlling device. At the top the condensers from which the high-tensions are conducted to the spectrograph. The transformer and rectifiers are not seen on the figure.

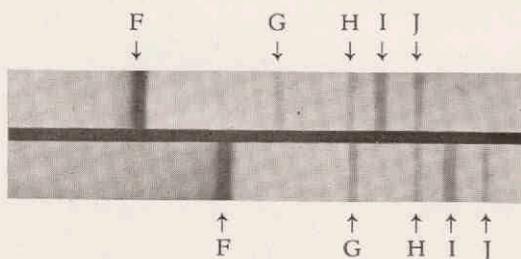


Fig. 27.

Reproduction of a photographic β ray spectrum, obtained with $\text{ThB} + \text{C} + \text{C}''$. The upper spectrum is retarded by a tension of 28 kV, the lower spectrum is unretarded.

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STELLINGEN.

I.

Bij fotografische intensiteitsmetingen aan electronen is het zinvol, van het bestaan van een „nul-effect” bij de fotografische methode te spreken.

II.

Het is gewenscht, dat in wetenschappelijke publicaties op het gebied der Natuurwetenschappen, de experimenteele resultaten niet alleen in de vorm van grafieken, maar daarnaast eveneens steeds in de vorm van tabellen weergegeven worden.

III.

Het zou ten zeerste toe te juichen zijn, wanneer de mogelijkheid van tijdelijke uitwisseling tusschen Nederlandsche en buitenlandsche studenten of assistenten van overheidswege aangemoedigd werd.

IV.

Zij $f_n(x)$ een oneindige rij van reële, continue functies van x met periode 1, die in elk punt gelijkmatig begrensd is, en zij voorts nog gegeven:

$$1. \quad \overline{f(x)} \equiv 0$$

(de streep boven het functie-teeken beteekent de limiet voor $n \rightarrow \infty$ van het gemiddelde over de eerste n functies in een punt x).

2. $\overline{\{f(x)\}^2}$ onafhankelijk van x .

$$3. \quad f_n(x) = \sum_{\nu=0}^{\infty} A_n(\nu) e^{2\pi i \nu x}$$

in elk punt x (bedoeld is het reële deel).

Zij verder het „ ξ -gemiddelde” van de functie $f_n(x)$ gedefinieerd door:

$$g_n(\xi, x) = \frac{1}{\xi} \int_x^{x+\xi} f_n(t) dt$$

dan gelden de volgende benaderde betrekkingen voor het quadratisch gemiddelde van deze grootheid:

$$a. \quad \lim_{\xi \rightarrow 0} \overline{\{g(\xi, x)\}^2} = \overline{\{f(x)\}^2}$$

$$b. \quad \overline{\{g(\xi, x)\}^2} = \frac{1}{2\xi} |A(0)|^2$$

indien $\xi \ll 1$ en indien $|A(\nu)|$ een langzaam veranderlijke grootheid is in het gebied $0 < \nu < \frac{1}{\xi}$.

$$c. \quad \overline{\{g(\xi, x)\}^2} = |A(0)|^2 \text{ indien } \xi \sim 1.$$

V.

Nog steeds laat de experimenteele verwezenlijking van de „Uraanexplosie” op zich wachten.

S. Flüggge, Naturwiss. 27, 402, '39.

VI.

De ingewikkelde meetmethode, die door Joliot en Zlotowski aangegeven wordt, om de kromming in verschillende punten van de baan van een elementair deeltje in de Wilsonkamer te bepalen, kan door een zeer veel eenvoudigere en ten minste even nauwkeurige methode vervangen worden.

F. Joliot, I. Zlotowski, Journ. d. Phys. 9, 393, '38.

VII.

Bij de vergelijking van de vorm van experimenteel verkregen β -spectra met de theorie, dient men er rekening mede te houden, dat de corresponderende energie overgangen eventueel verboden kunnen zijn, hetgeen tot nu toe veelal nagelaten werd.

VIII.

Door Bethe, Hoyle en Peierls is de veronderstelling geopperd, dat de experimenteel verkregen β -spectra samengesteld zouden zijn uit meerdere enkelvoudige spectra van verschillende intensiteiten en met verschillende bovenste grenzen, die elk aan de theorie van Fermi zouden voldoen. Zij meenen echter, dat deze onderstelling in strijd is met het feit, dat de spectra beschreven kunnen worden door de theorie van Konopinsky-Uhlenbeck. Het tegendeel is echter waar: de goede overeenstemming tusschen de theorie van K-U en het experiment is een sterke steun voor de onderstelling van Bethe, Hoyle en Peierls.

H. A. Bethe, F. Hoyle, R. Peierls, Nature
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