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DOOR

WILLEM RHIJNVIS VAN WIJK

GEBOREN TE MAKASSAR

★

Aan mijn Ouders!

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THE NUCLEAR SPIN OF NITROGEN

BY

W. R. VAN WIJK.
UTRECHT – DISSERTATION.

INTRODUCTION. [1]

In most band spectra the intensities of the band lines regarded as a function of the line-number suit to a continuous curve.

Also, however, in some cases it has been found that every other line is weaker or even missing.

As this alternation of intensity has only been observed in the spectra of molecules formed by equal atoms, the explanation has been sought in the symmetry of the molecule. It remained however as also for the other resonance phenomena for the new quantum mechanics to give a satisfactorily explanation of this fact. (1)

Moreover as it was ascribed to a resonance in the atomic nuclei, a new way for investigation of the nucleus was opened. (1)

One of the most interesting cases investigated is undoubtly the alternation of intensities in nitrogen, and in this case the expectation that the study of this phenomenon should lead to most interesting conclusions on the structure of the atomic nucleus seems to have been fulfilled.

From the intensity-measurements (2) the electron has been found to lose its spin (3) in the atomic nucleus whereas from the Raman effect (4) the non applicability of the principle of Pauli in its ordinary form has become clear. (5)

With this deep difference between the properties of the electron out and inside of the atomic nucleus it is doubtfull whether the electron has to be regarded as a particle in the nucleus. (5)

But for a good understanding it may be desirable to explain

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[1] The references relate to the litterature list at the end of the article.

first briefly some properties of diatomic molecular spectrum structure, as have been reveiled during the last years by several investigators. (6)

SOME PROPERTIES OF DIATOMIC MOLECULAR SPECTRUM STRUCTURE. [1]

The energy of a diatomic molecule can be divided into three parts.

There is first the electronic energy due to the movement of the electrons in the field of force of the nuclei, further there is the vibrational energy of the nuclear masses and finally the molecule considered as a top, can have an energy of rotation.

Of course there is no sharp division between these three components possible, but if one neglects the mutual interactions, one can get an approximate idea of the molecular energy-scheme by superposing successively the energy of a molecule possessing only one of these three energy-possibilities at a time.

Consider first the electronic part alone.

On account of the axial symmetry of the field of force in which the electrons are moving, their azimuth in a plane perpendicular to the line joining the nuclei (the molecular axis) is a cyclic variable, wich means that the electronic moment of momentum Λ has a constant integral value along the axis.

There is a degeneration of first order in case $\Lambda > 0$ caused bij the indefiniteness of the sense of rotation, which gives rise to the so called Λ type doubling if that degeneration is removed.

If the resultant spin S of the electrons differs from 0, each term with $\Lambda > 0$ is split up into a multiplet by the different possible orientations of the spin, whereas the multiplicity is, just as in the case of atomic spectra, given by $2S + 1$. These sublevels are characterized by a number Ω where $|\Lambda - S| \leq \Omega \leq |\Lambda + S|$.

The energy of the vibration of the nuclei is mainly that of the harmonic oscillator, but it may be mentioned that if a change of electronic state takes place at the same time as a vibrational change, the selection principles of the harmonic oscillator do

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[1] As the subject of this chapter is not directly connected to the nitrogen problem, no litterature is given here but the reader is referred to ref. 6. The notation for the rotationallevels by n is adopted for obtaining the same notation in both bandspectra. For the negative bandsystem n corresponds to N in the usual notation (Faraday meeting) and for the positive bands to J.

not even approximately apply. So in the case of nitrogen the bands corresponding to a $0-0$ vibration transition are strongest.

Then there is the energy of a rotating rigid molecule, which, if the electronic masses are neglected, becomes the problem of the rigid rotator.

There is a constant moment of momentum N of such value, that $N^2 = n(n+1)\left(\frac{h}{2\pi}\right)^2$ if $n = 0, 1, 2, \ldots$ and the energy of this system is $\frac{N^2}{2I^2}$, wherein I means the inertia-moment of the rotator and h the Planck constant.

Also the component of N in an arbitrary direction is constant, corresponding to a non-appearance of the azimuth Φ of the rotator in the Schrödinger-equation. So the factor of the characteristic functions, depending on Φ takes the form exp. $\pm \{i\, m\, \Phi\}$

In the case of equal nuclei it is clear that by interchanging the nuclei all coördinates are left unchanged except Φ, what passes over into $\Phi \pm \pi$. Thus the characteristic functions are left unchanged for n even and reversed in sign for odd n values (symmetrical or antisymmetrical in the nuclei respectively).

Thus the total energy of the molecule can be written as $E = E\,el. + E\,vibr. + \frac{h^2}{8\pi^2 I}\, n(n+1)$ [1].

The different lines of one band are caused by transitions from rotational energy states to others, the electronic and vibrational part for all initial levels being identical; the same is true for the final levels.

In the case of the third positive nitrogen bands (7) ($^3\Pi - {}^3\Pi$) electronic transition, (this designation means that $\Lambda = 1$ in both initial and final state and the multiplicity is 3) and also in the negative bands (8) ($^2\Sigma - {}^2\Sigma$, $\Lambda = 0$ in final and initial state, $S = 1/2$), only the transitions with $\Delta n = \pm 1$ remain strongly present, moreover in the positive bands Ω does not change, and so the following schemes of the bands are applicable, the arrows indicating the strong transitions.

(arrow to the left: line of the R branch $\Delta n = -1$)
(arrow to the right: line of the P branch $\Delta n = +1$)

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[1] In the case of the negative bandsystem of nitrogen $n = 0, 1, 2, \ldots$ For the third positive group $n = \Omega, \Omega + 1, \Omega + 2, \ldots$

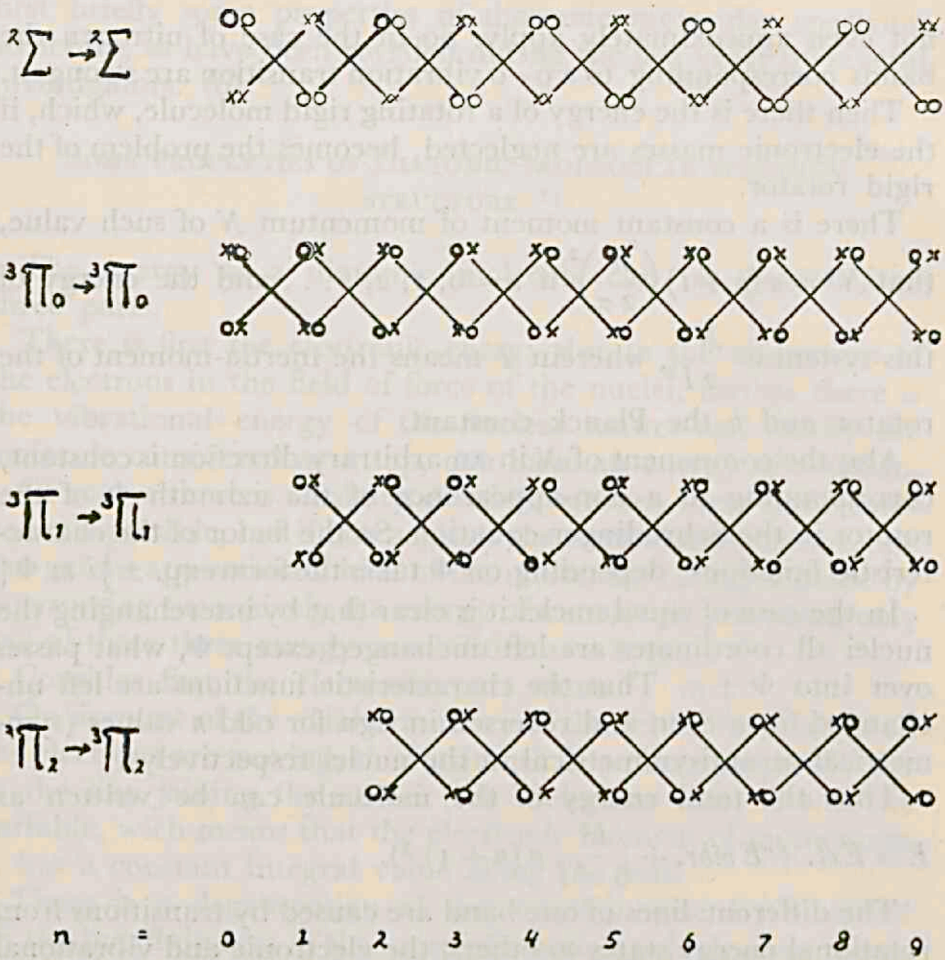

Fig. 1. Showing P & R branch transitions between the rotational-levels.

Now the structure of the bands from which in fig. 2, at the end of the article, there are given photometer traces is easily understood.

Every level in the negative bands is degenerated, by the influence of the electronic spin. [1]

In the positive bands the Λ doubling causes a doubling of the lines of the Π_0–Π_0 series (denoted as *c* series) and a broadening of the Π_1–Π_1 lines (*b* series). In the Π_2–Π_2 lines (*a* series) the effect is undetectable. [2]

In the negative bands for $n > 0$ the degeneracy caused by the two orientations of the electronic spin is removed and so every

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[1] The degeneated levels are designed as double in the fig. 1.

[2] For convenience the old notation is beheld. The designation of the lines is that given by Zeit (ref. 7).

level splits up into two, so that for large quantum numbers the lines appear as double. The plate from which the photometer trace in fig. 2 has been taken was photographed with too low resolving power to show this doubling.

The characteristic function of the molecule in the limiting case of negligeable interaction can be written as a product of factors, depending on the electronic coordinates referred to the fixed nuclei (including the electronic spin) and of the coördinates of the nuclei due to the vibration and rotation.

The electronic factor is either symmetrical in the nuclei (*Sy* N) or antisymmetrical in the nuclei (*An* N) if $\Lambda = 0$ but if $\Lambda > 0$ from the two sublevels one is *Sy* N, the other *An* N. (9).

The function dealing with the nuclear vibration is always *Sy* N, as only the distance of the nuclei comes into it. The symmetry of the rotational part, however, has already been discussed before, and the subsequent functions corresponding to $n = 0, 1, 2$ etc. are alternately *Sy* N and *An* N. The translation of the molecule has been neglected till here. Its characteristic function is always *Sy* N.

The total characteristic function thus is *Sy* N or *An* N according to the even or odd number of *An* N factors figuring in it.

If there is a coupling between the different motions in the molecule, the separation of the characteristic functions into factors is impossible, but the symmetry properties remain unaffected. Now the o and × signs in the fig. 1 can be understood. The × mean *Sy* N, the o *An* N terms.

With these considerations on the symmetry properties the phenomenon of alternating intensities can be understood. For if according to the extended Pauli principle only the functions which are antisymmetrical in both electrons inclusive nuclear electrons and protons correspond to actually existing states, in the case of equal nuclei containing an odd number of particles (electrons + protons) all *Sy* N terms do not occur and so the + terms of fig. 1 should be omitted. On the other hand, if there are an even number of particles in the nucleus the o states should not appear.

It is the case if there are no other coördinates than its space-coördinates required for defining the nucleus.

But if the nucleus possesses a spin, a formerly forbidden state (*Sy* N) can be transformed in a permitted one by the mutual orientation of the nuclear spins. Then this state does appear with a weight, calculable from the different possible spin-vector orientations.

Generally speaking with a nuclear spin of m units, the weights of the different *Sy* N and *An* N spin-orientations are in the ratio $\frac{m+1}{m}$. This can be found by considering the different possible orientations of the spins which give resultants of $2m$, $2m-1$, $2m-2$, ... 1, 0, units. Of these, the states $2m-2k$ (k integral number) are *Sy* N, the others *An* N. Because of the statistical weight of $2(2m-2k)+1$ for the spin $2m-2k$, the sum of the weights for the *Sy* N states is $2m^2+3m+1$ and that for the *An* N states $2m^2+m$, so that their ratio becomes equal to $\frac{m+1}{m}$. The ratio 2,0 : 1 found in the nitrogen bands therefore indicates a spin of 1 unit. A ratio of 3,0 : 1 or 1.66 : 1. corresponds to a spin of $\frac{1}{2}$ or $\frac{3}{2}$ respectively.

From the Kramers–Heisenberg dispersion formula governing the intensities in the light scattered by atoms or molecules it follows that only the frequency-difference of two energy-levels, which both combine with a third, appear as Raman-frequency.

So in the application to diatomic molecules only the combination-frequencies with $\Delta n = 0$ or ± 2 occur in the Raman-light if the transitions between the rotational-levels are governed by the combination rule $\Delta n = \pm 1$.

Actually in the Raman-effect in Nitrogen, the lines resulting from a 2–0, 3–1, etc. and from a 0–2, 1–3 etc. transition have been found. The lines corresponding to $\Delta n = 0$ transitions, however, have been blurred by the exciting line.

Also in these Raman-lines the alternating intensities have been found, although no attempt was made for measuring the ratio.

The lines due to a transition between levels of even rotational number have been found more intense.

This indicates that also without regard to the nuclear spin the characteristic functions for these states are antisymmetrical in all particles.

CONCLUSIONS ON THE NUCLEUS OF NITROGEN.

If one assumes a nucleus as built up from protons and electrons (i.c. they may form α particles), then the nitrogen-nucleus with its atomic weight 14 and atomic number 7 must contain 14 protons and 7 electrons, that is an odd number of particles.

Therefore the states corresponding to *Sy* N functions (*Sy* N neglecting the nuclear spin!) should be less frequent.

Moreover if it is possible for a system of electrons and protons to form a nucleus by an adiabatic transformation, upon which assumption the foregoing rules of symmetry and antisymmetry in the particles are based, the total spin of the nucleus should be formed by addition and subtraction of the individual spins for the free particles. As the protonic and the electronic spins are both $\frac{1}{2}\frac{h}{2\pi}$ ($h=$ Planck constant) one should expect an half-integral amount of nuclear spin. But the measurements in the negative and positive band spectra of nitrogen indicate the integral value one for the nuclear spin (3). Besides in the Raman-effect in nitrogen it has been found that the rotational levels in which n is even are more frequent.

As there seems to be no doubt that the ground level of N_2 is *Sy* N, this result is also not in agreement with the simple point of view which considers the N nucleus as formed by an odd number of particles. (5)

The hypothesis has been made that the electron [1]) loses its spin and its influence on the statistics of the nucleus. In fact this hypothesis explains both discrepancies in this case.

On the other hand, however, there seems to be an indication that the electrons in the nuclei of the atoms of atomic weight $4\,l$ ($l = 1, 2 \ldots$) are grouped in shells containing an even number of them, which may mean that the Pauli-principle does apply and that also the spin remains for the nuclear electrons. (10)

Consequently general conclusions cannot yet be drawn and further experimental research is wanted. In the following a critical discussion and extension of the intensity measurements is given.

EXPERIMENTAL METHODS.

Measurements have been done in the negative band spectrum of Nitrogen, due to the $\overset{+}{N_2}$ molecule and also in the third positive group, which is emitted by the neutral N_2 molecule.

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[1]) In the cases where the number of protons is odd, a half-integral nuclear spin-value has been found.

Of the negative bands the following four have been measured: 3914, 3884, 4278, 4237 Å corresponding to vibrational transitions 0 – 0, 1 – 1, 0 – 1 and 1 – 2. Of the positive bands: 3371, 3805 and 3755 have been investigated (vibr. trans 0 – 0, 0 – 1, 0 – 2). (fig. 2.)

In both cases, the light source was a hollow cathode of sheet nickel about 1 cm high, 2 cm deep and 0,1 to 0,3 cm broad; the dimensions varying from tube to tube. A nickel plate mounted together with the cathode on a lamp-bridge at a distance of about 0,5 cm, served for anode. This apparatus was sealed into a glass sphere of 15 cm diameter provided with a tube, 20 cm long, bearing a (if necessary quartz) window at its end.

When filled with nitrogen at a pressure of about a millimetre and put on high voltage (500–1000 V), a strong light appears only in the inner part of the hollow cathode. If the current is strong (50 to 100 mA) the cathode comes to a red or even yellow heat. The small dimensions of the radiating gas layer and its enclosure by a metallic box are very favorable for securing a constant temperature throughout, which is of importance for the intensity-distribution in the bands. For as the intensity-distribution over the lines in a band depends on the relative concentrations of the upper rotational-levels, (which concentrations in turn are determined by the temperature of the gas), only in the case of a radiating gasmass of uniform temperature, a simple intensity-distribution in a band can be expected. (11)

The nitrogen used, was produced by heating sodium-azide in vacuo. The spectrograph used was the 6 M grating of this institute, of which a description is given elsewhere. (12) Generally a slit-width of 0,003 – 0,004 cm was used for the bandspectra-photographs. Also two plates of band 3914 Å have been made with 0,007 and 0,009 cm slit-width to see if variation of the slit-width did alter the results. For the objection could be made to the way by which the density marks have been taken, that it gives the density curve for broad lines, as the density marks were about 0,008 cm broad (see below).

As the results of the bands taken with the wider sliths were in good agreement with the others, such an effect seems not to have occurred.

The density marks have been taken in the usual way with an incandescent lamp burning on constant voltage and a small prism spectrograph (glass or quartz as was necessary), in which the slit-width has been varied in several (12 or 10) steps. Before use this slit has been tested under a measuring microscope. Also as a check density marks have been put by using a mercury arc on constant current and placing reducers before the slit of the grating. As is mostly the case in working with reducers, the points, giving the density-curve, spread more, due to the inhomogeneities in the reducersteps, but systematic effect were not detected. The difficulties met in finding reliable reducers, especially for the ultraviolet region were the reason for taking the density marks by the method of slith-width variation.

The great difficulty in taking density-marks is in most cases due to the stray light. By the use of a very short slit of only 0,008 cm height this stray light has been removed to a large extent. Moreover by a diaphragm behind in the spectrograph the scattered light from one exposure was kept away from the other density marks.

In the case of the ultraviolet bands 3805, 3755, 3371, also a nickeloxide glass has been put before the slit of the spectrograph to filter out the intense visible spectrum of the quartz band lamp, what also is a source of stray light.

With these precautions the stray light is reduced in any case too much below 1 % of the intensity in the spectral region used.

There was generally a difference in time of exposure between the band spectra and the density marks. In a research on its influence on the density curve, carried out in this institute, the maximum time differences are determinated for which the results are not affected. (13) Care has been taken that these limits were not exceeded. Also for the strong 3914 Å band in the first order an exposure has been made with the same time as used for the density marks, from which plate quite similar results as from the others were obtained.

Bands and density marks have been taken on Ilford special Rapid *H* and *D* 400 plates from the same box, as it was impossible to put them on the same plate on account of the small dimensions (9–6 cm) of the plates to be used in the prism spectograph.

In the beginning the plates of size 12–9 cm have been cut into two and on one half the band-exposure, on the other the density-marks have been taken. Control experiments have shown however that in our case no greater scattering in the density-intensity relation appears for plates from the same box than is obtained from a single plate. Therefore furtheron, for the plates exposed at the same time only once density-marks have been taken. Also for plates from different boxes but treated in the same way, the density curves were fairly parallel to another.

All plates (band spectra photographs in different orders of the grating and density marks) have been put together on a glass strip and fixed on it by rubber bands, so that intense shaking during the development process (8 min. in 5 % Rodinal at 18° C.) was possible without fear for an overlapping of the plates. It is essential for avoiding disturbing developments-effects that the developer should be kept into incessant vehement motion, and if this is done one can get rid of them to a large extent. Also several times two plates with density marks were taken and fixed at different stages on the glass plate. They showed no systematic difference in their density-curve.

Especially in the case of the positive Bands these development effects are very disturbing, for the lines to be compared lie close to one another (distance about 0,004 cm).

It may be mentioned that the development effect (Eberhard effect) is probably due to partial fluctuations in the concentration of the liquid, so that near a strong line the concentration is lower. This causes a dependance of the density-intensity relation on the density in the neighbourhood, which especially in the case of an analysis, where the intensities in regions with a large density-gradient is sought, gives rise to great errors. In the peak of a line this gradient equals zero and so one might suppose a smaller influence of these effects in peak-intensity measurements, what has been checked experimentally in this institute.

For recognising the presence of such developer effects directly, the plates of the band 3371 on which the most accurate measurements have been made (it was the only band which could be taken in the 4th order of our grating), were developped a longer time, 9 or even 10 min., than the others, till a strong chemical fog appeared. The Eberhard effect, if there is any, then shows itself by clearing up of the fog near a strong line. At last plates could be obtained what showed no trace of such an effect.

After the development the plates were washed for several minutes and then fixed in a solution of Sodium hyposulfite.

After drying, the plates were photometred with the Moll-microphotometer of this institute. Whereas an analysis of the curves was necessary, much care was given to obtain correct focussing of the instrument. The instrument was used therefore with a primary slit of 0,025 cm and a slit-width varying from 0,0025 cm to 0,0050 cm on the thermopile. A narrower slit decreased the measuring accuracy, in causing a too small galvanometer deflection, whereas a much wider one (0,008 cm) was not narrow enough with respect to the linewidth. These values were found experimentally, as the same line-doublet was taken several times with different slit-widths.

No longer distance than about 2 cm on the plate was taken in one run, after which the plate was refocussed.

The overlapping of the different parts taken in several runs gave a good check on the adjustment of the instrument.

Immediately before and after each run zero-marks were taken and also an unexposed part of the plate close to the measured lines, was photometred on the same part of the registration. By doing thus any inconstancy of the photometer-lamp could be taken into account and also local fluctuation on the plate could be detected and if so, corrected for.

A registration of line C_{29} in band 3371 is reproduced here as an example.

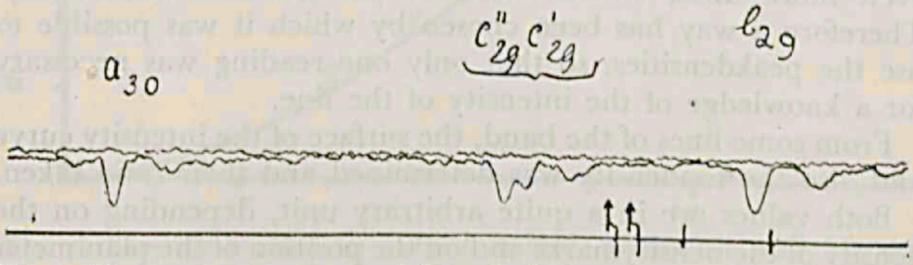

Fig. 3. Photometer trace as used for actual measurement of c_{29} 4th order.

Especially in the case of small densities, an exact knowledge of the galvanometer deflection for the unexposed plate is necessary. Different relations between the distances on the plate and registration on the drum have been used. The registrations were developped with Hydroquinone *A* – *B* or Metolhydroquinone. If an analysis was aimed at, the registration was enlarged by means of an epidiascope, if not they were measured directly with a glass scale.

THE REDUCTION OF THE MEASUREMENTS.

Negative Bands. In $^2\Sigma - {}^2\Sigma$ bands the lines possess a fine structure so that for large quantum numbers, they appear as doublets of nearly equal intense components. Nearer to the origin there are theoretically three components (except for the first P and R branch lines which are really double), and also the intensity of the components should differ greatly; but the fine structure is so narrow that for lines below $n = 10$, even in the third order of our grating no splitting up could be detected. From the intensity measurements there are, however, indications that for the lines near the origin the structure is more complicated (see page 15).

For the purposes of this research, only the total intensity of the line, (without regarding its fine structure) was needed, so that the densities of the plate had only to be reduced to intensities and the intensity-curve of each line integrated by means of a planimeter. Whereas the fine structure pattern becomes wider with the quantum number, the shape of the line alters and therefore it is incorrect to use the density of the peak as a measure for the total intensity.

But the reduction of a line into intensities means a determination of the intensity of about fifty points, so that it is a lasting work to measure one band containing about forty or even more lines, which could be measured with accuracy. Therefore a way has been chosen by which it was possible to use the peakdensities, so that only one reading was necessary for a knowledge of the intensity of the line.

From some lines of the band, the surface of the intensity curve and also the topdensity was determined and their ratio taken.

Both values are in a quite arbitrary unit, depending on the density of the densitymarks and on the position of the planimeter arm, so that for each plate the ratio should differ. Therefore in the case of no overlapping of the intensity curves in the top of the line (wide spaced doublet) this ratio was taken to unity and the values were reduced in proportion. Thus this value of surface intensity divided by peak intensity goes towards 0,50 with narrowing of the fine structure.

Then the values got from the chosen lines were plotted in a diagram with this ratio on the y-axis while on the x-axis the number of the bandlines were plotted.

As an example a reproduction of such a diagram is given for the band 3914 Å taken in the second order. The lines analysed were in this case the numbers 16, 17, 18, 21, 22 of the R branch.

It can be seen from the figure that for lines of higher quantum number than 23 no correction in the peak intensity had to be carried out whereas for all numbers lower than 14 the peak-intensity had to be multiplied by 0,50. For the lines of the third order exposures these numbers were 22 and 3 respectively. Now from the curve drawn through these points the surface intensity could be found by multiplication of the intensity in the top of the line by the ratio corresponding to its number.

In this way the analysis of all lines (about forty) was made superfluous. Moreover the same curve could be used for other plates taken with the same focussing of the grating, so that no analysis was necessary at all for later plates.

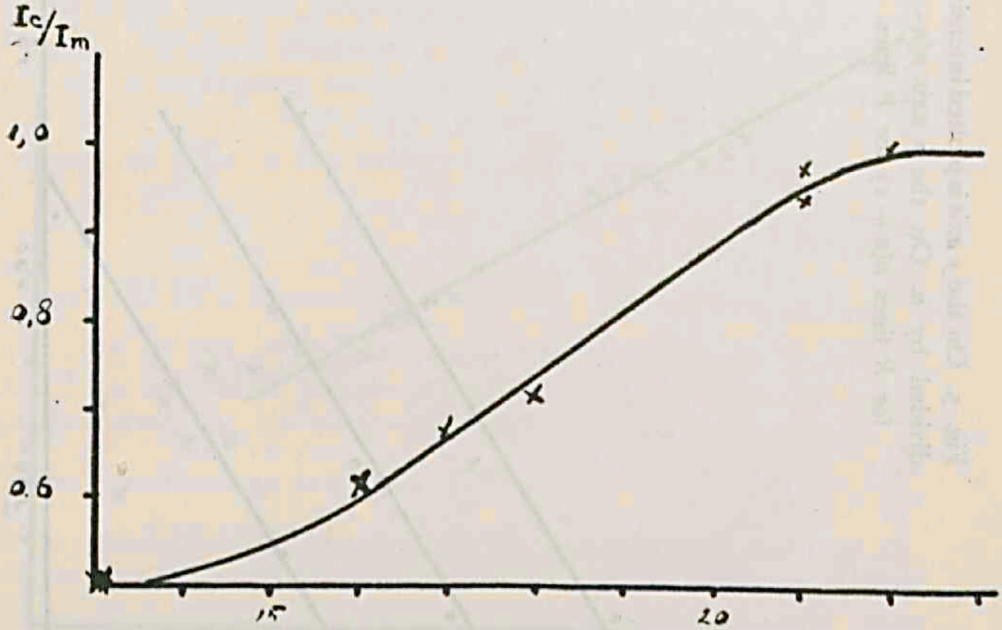

Fig. 4. Showing surface intensity I_c divided by peak-intensity I_m as a function of line number.

Of course a check was made. So in one case the 3914 Å Band photographed in the third order, was worked out fully in both ways by drawing the intensity-curves and planimetring them and also in the way described before.

If the procedure is correct, the intensities got in these two ways should have a constant ratio throughout the whole band. (They do not give the same value because of the arbitrariness in the value mentioned before). That this was the case can be seen from fig. 5, wherein the crosses represent the values got from direct planimetring and the circles those from peakdensities.

The exact parallelism of the two straight lines shows the correctness of the procedure.

In this way for the third and second order a reducing curve has been drawn, but for the first order the topdensity could be used without any correction for a sufficiently large number of lines (about 25 or 30).

A measurement of the doublet separation has also been

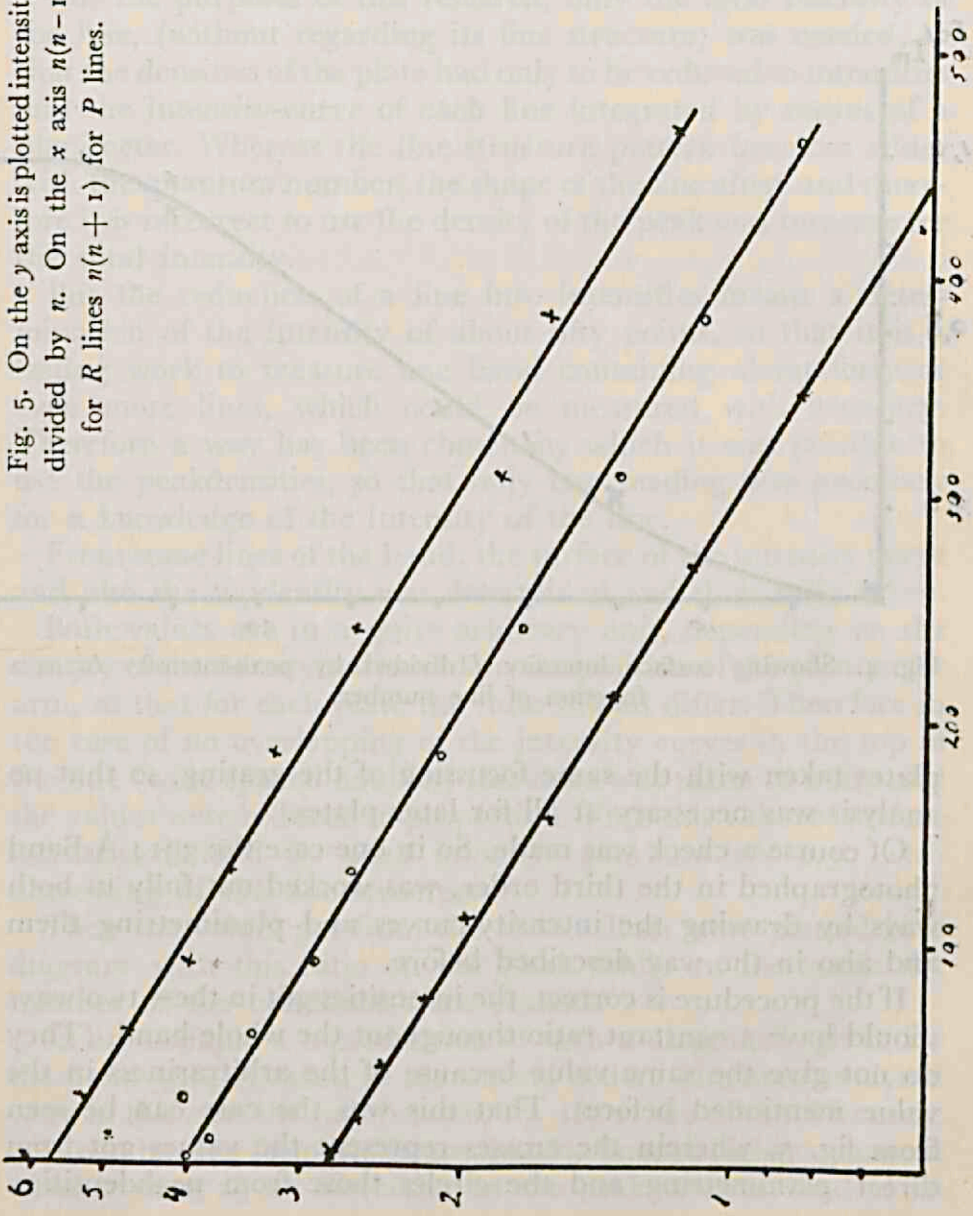

Fig. 5. On the y axis is plotted intensity divided by n. On the x axis $n(n-1)$ for R lines $n(n+1)$ for P lines.

carried out. By analysing the intensity-curve for the widely separated doublets into its components, the curve of a line possessing no fine-structure was obtained. Then an attempt was made to account for the experimentally found ratio of surface- and peakintensity by assuming that the actually line was built up from two equal intense components, whose distance varied with their quantum number n.

In this manner the distance of the components could be measured. For the high numbers $n > 10$ this distance was a linear function of n in concordance with theorie, but for $n = 7,6,43$, the doublets width seemed to become quite irregular, what should indicate for an unequal intensity of the components.

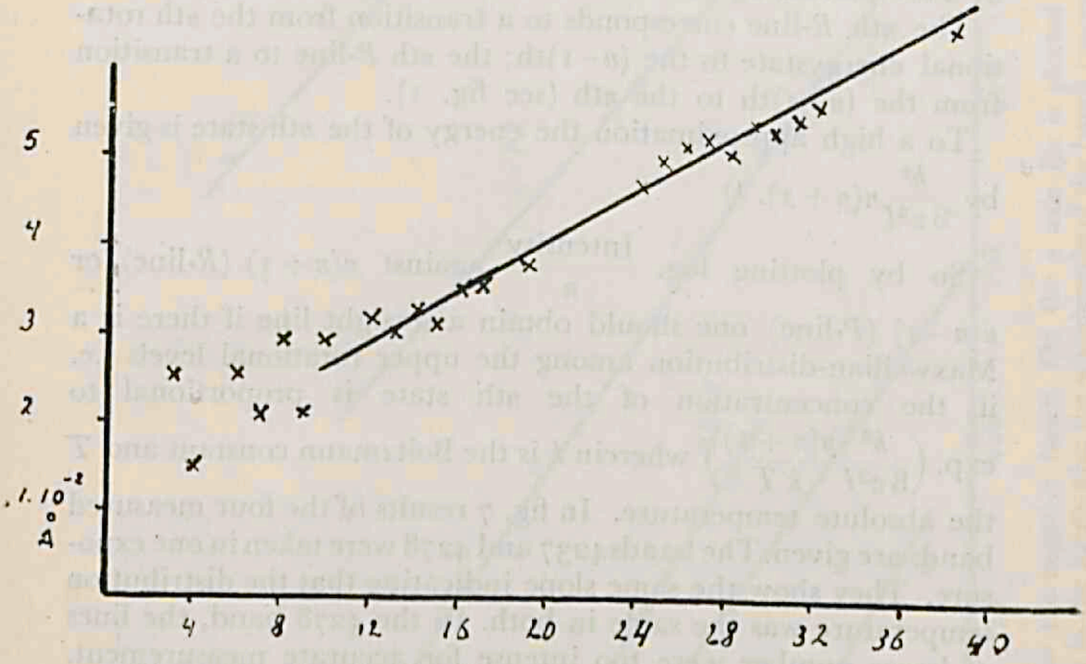

Fig. 6. Showing doublet-distance as a function of line number.

In fig. 6 the distance of the doubletcompoenects is plotted on the axis of Y against on the X-axis, the number of the line.

In total there have been worked out from band 3914 Å 2 third-, 2 second- and 2 firstorder plates; from band 3884 Å 2, from 4278 and 4237 Å 1 plate, all taken in the first order.

The intensity of a spectrumline is proportional to a product of two factors sc. the radiationchance from the upper to the lower level (where from boths their statistical weights have to be taken into account) and the concentration of the upper level. In this case the former factor is proportional to n for both the nth P as well as the nth R line, understanding under a line the total complex of fine structure components.

If one suspects that the latter factor can be described with a Maxwell-Boltzmann distribution function, the following graphical representation of the measurements seems to be preferable.

On the axis of Y the logarithms of the line intensity divided by n are plotted against the rotational energy of the upper level.

The nth R-line corresponds to a transition from the nth rotational energystate to the $(n-1)$th; the nth P-line to a transition from the $(n-1)$th to the nth (see fig. 1).

To a high approximation the energy of the nth state is given by $\frac{h^2}{8\pi^2 I} n(n+1)$. [1]

So by plotting log. $\frac{\text{Intensity}}{n}$ against $n(n+1)$ (R-line) or $n(n-1)$ (P-line) one should obtain a straight line if there is a Maxwellian-distribution among the upper rotational levels i.e. if the concentration of the nth state is proportional to exp. $\left(\frac{h^2}{8\pi^2 I}\frac{n(n+1)}{kT}\right)$ wherein k is the Boltzmann constant and T the absolute temperature. In fig. 7 results of the four measured bands are given. The bands 4237 and 4278 were taken in one exposure. They show the same slope indicating that the distribution temperature was the same in both. In the 4278 band, the lines of lower number were too intense for accurate measurement. Several further lines have been measured; they fall however out of the figure to the right side. As the slopes of the lines are equal to $\frac{h^2}{8\pi^2 I}\frac{1}{kT}$ log e multiplicated by a factor depending on the choice of the units on Y and X axis, the temperature T can be calculated from it.

The calculation of the temperature from the slopes of the lines using the value $1.34.10^{-39}$ for I gave temperatures varying from

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[1] As professor ORNSTEIN remarked to me, perhaps the inertia moment of the lower level should be taken, this depending on the rules for excitation by electron-impact. This should alter the temperature of the distribution bij about 0,5 % but it does not affect the straightness of the lines at all.

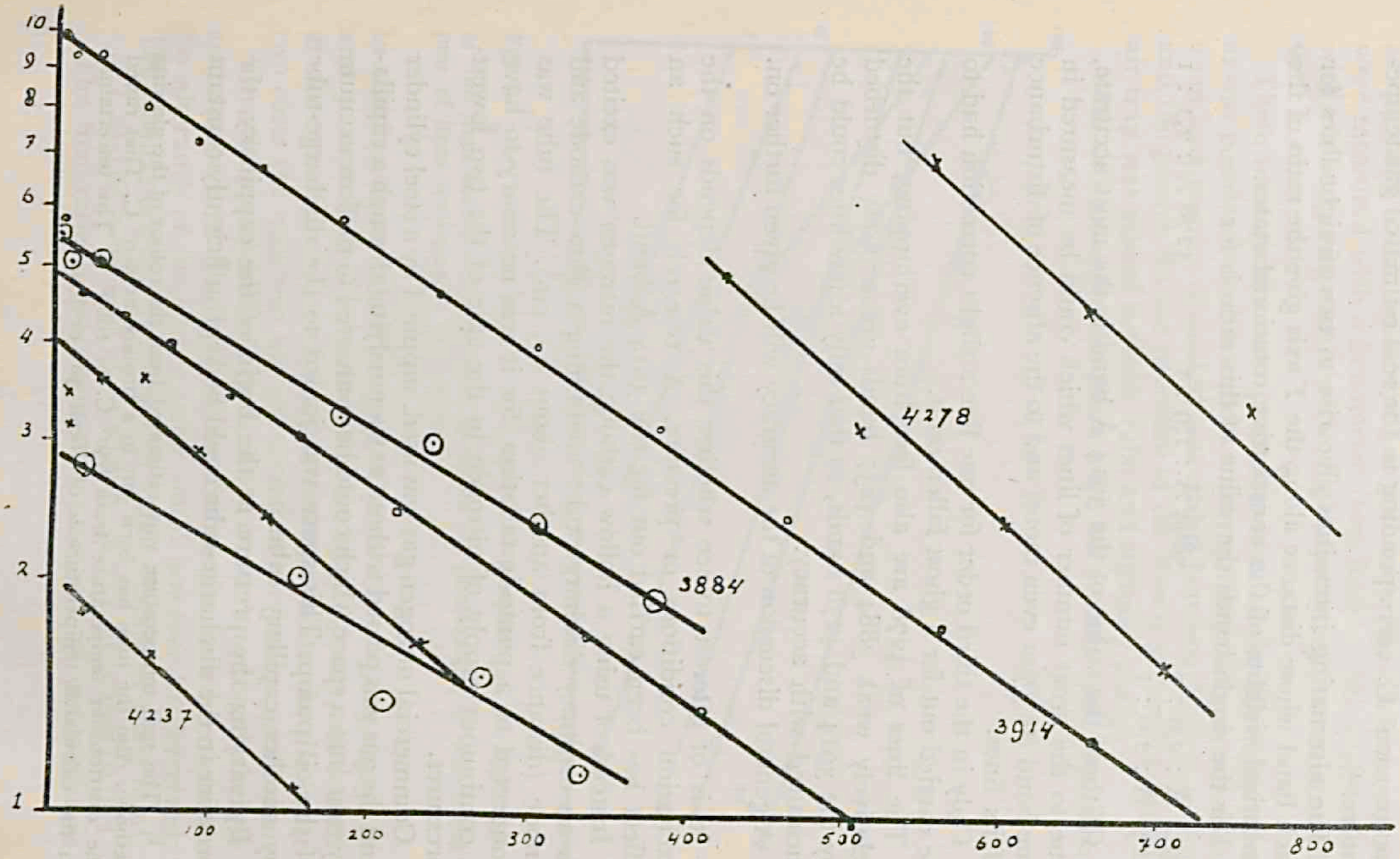

Fig. 7. Specimina of the lines obtained for the negative bands. On the y axis I/n is plotted against $n(n-1)$ or $n(n+1)$ respectively on the x axis. The numbers in the figure give the wavelengths of the bands corresponding to the lines.

500 to 1000° C. corresponding to the real estimated gas temperature. [1])

The alternating intensities give rise to two straight lines for each band whose distance along the *Y* axis give the ratio of the statistical weights of the consecutive rotational states.

For the single bands the value of this ratio is for

3914 Å : 2,0 : 1 3884 Å : 1,9 : 1 4278 Å : 2,1 : 1
4237 Å : 2,0 : 1

Of these the value of the 3914 Å band is the most accurate, due to the great number of lines which could be measured in one band (to 35 or even more) and to the absence of disturbance of its lines.

Only in the third order for one line a slight correction had to be carried out for a ghost fallen on it.

The lines of 4278 are also free from overlapping, but the relatively weak 3884 and 4237 bands are severely disturbed by the 3914 and 4278 bands, so that only a few lines could be measured with accuracy.

A general discussion of the accuracy will be given further on.

It is of interest to see whether this value depends on the excitation conditions or pressure. A research for such an effect has been carried out for the 3914 Å band.

Instead of using a hollow cathode the nitrogen was excited in an ordinary discharge tube containing a glow-cathode and anode (distance from another about 0,5 cm). The tube was connected to a pump-installation, for it was necessary to have a continuous supply of nitrogen in the case of the two lowest pressures.

Commercial nitrogen gas was used, supplied by a steel cylinder and the gas was passed without extra purifying through a capillary tube into a space which could be connected to the low vacuum of the oil pump. This space was joined to the discharge-tube by another capillary tube.

By varying the pressure at the begin of the cappillary, the pressure in the discharge-tube could be kept sufficiently constant

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[1]) The real temperature was estimated from the colour of the glowing cathode. Also the tube has been put in a furnace at 300° C. That raised the distribution temperature from 650° C. to 1000° C. The uncertainty in the calculated temperature is of about 30° or less.

at any value in the region 0,005 to 0,10 mm. The lower pressures were measured with a kenometre (von Reden), the lowest also with a Mac-Leod gauge; the two higher pressures were measured with a manometre.

These instruments were placed as near to the dischargetube as was possible, whereas a short wide capillary tube (dimensions 2 cm long, 0,5 cm diameter) was sealed between the kenometre and the pumps. In the exposure at the lowest pressure a liquid air trap was placed between tube and mercury pump.

With this arrangement there have been taken four exposures

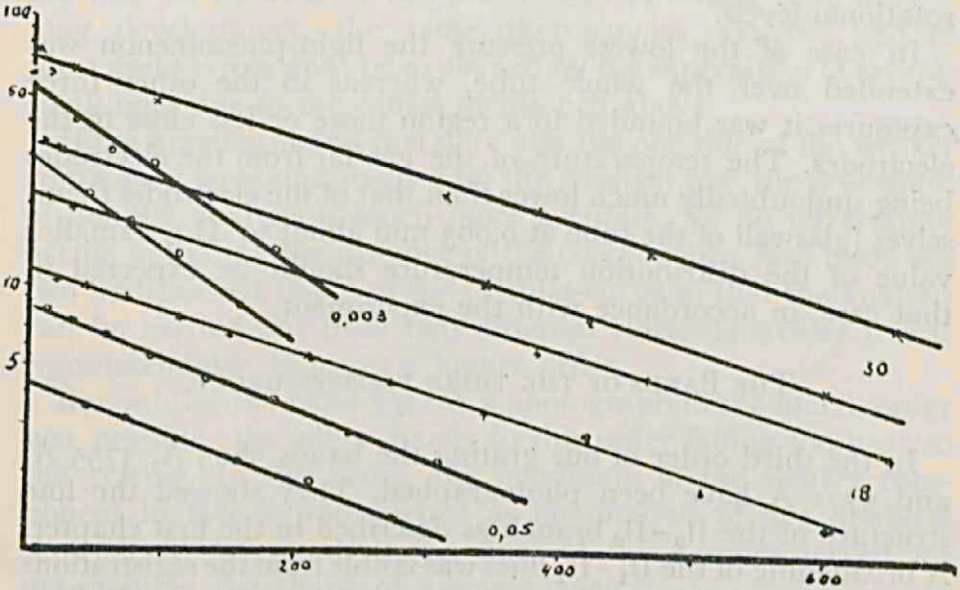

Fig. 8. $\frac{I}{n}$ on a y axis; $n(n-1)$ on x axis. The numbers at the right side of the lines indicate the pressure at which the photograph, to which the pair of lines corresponds, was taken.

in the first order of the grating at pressures of 0,003, 0,05, 18 and 30 mm. The first value is correct at 50 % only, the second at about 20 %, the other two are more accurate. In the latter two cases the tube was not connected to the pump-installation.

Thus the pressure has been varied in a ratio 1 to 10000 whereas at the same time the discharge-conditions also have been varied. No alterance of the 2,0 : 1 value could however be perceived. The results obtained from the four exposures are at 0,003 mm : 1,9 : 1, at 0,05 mm : 1,95 : 1, at 18 mm : 2,0 : 1, at 30 mm : 2,0 : 1.

The first ratio is less accurate on account of the weakness of the photograph so that the densities felt in an unfavourable part of the density-curve.

In fig. 8 the curves are reproduced. In the curves corresponding to 18 and 30 mm there are lacking some points; these have been omitted because they were too strong for accurate measurement or also in one curve because the plate was stained there.

The temperatures calculated from the slopes of the lines were 380, 680, 870 and 870° abs. respectively. These values give strong support to the idea, that it is the real gastemperature, which determines the distribution of the energy among the rotational-levels.

In case of the lowest pressure the light-phenomenon was extended over the whole tube, whereas in the other three exposures it was bounded to a region more or less close to the electrodes. The temperature of the gas far from the electrodes being undoubtedly much lower than that of the electrodes themselves (glaswall of the tube at 0,003 mm about 50° C.), a smaller value of the distribution temperature should be expected in that case, in accordance with the experiment.

THE BANDS OF THE THIRD POSITIVE GROUP.

In the third order of our grating the bands 3805 Å, 3755 Å, and 3771 Å have been photographed. They showed the fine structure of the $\Pi_0 - \Pi_0$ branch as described in the first chapter. A broadening of the $\Pi_1 - \Pi_1$ lines was visible from the registrations for the lines of high quantum numbers, whereas the $\Pi_2 - \Pi_2$ lines appeared quite narrow. The different behaviour of these three multiplet component bands has been explained by theory.

In particular the relatively wide fine structure of the Π_0 levels, which is independent of the rotation has been explained by the fact that the sublevels pass over into a P_1 and P_2 level, if the axiality of the molecular field of force is removed. The great number of lines occurring in one band and the narrowness of the fine structure (0,025 – 0,030 Å) causes great difficulties in the intensity measurement.

Whereas the top of the weaker components was not undisturbed, the intensity-curve of the doublet had to be analysed in any case. On account of the strong overlapping, the disturbance by ghosts and other lines and the unequal intensities of the components, which causes difficulty in finding the true distance of the doublet-components in the registration, the reached accuracy was not great. The intensity-ratio of the components found from planimetring and from the peakintensity of the analysed components, differed mainly 30 % and in several cases even 50 % or more.

The intensity- ratio taken from the surface measurements were systematically higher than those from peakdensities.

At first this errors were ascribed to development effects, but also when the plate was certainly free from such effects in showing no clearing of the plate, close to strong lines after a long development, the same discrepancies occurred and so these great errors must be explained by the supposition of several small ones due to the causes mentioned above.

If the maximum and therefore also the one side of the weaker component were undisturbed by the stronger one, the procedure of doublet analysis is much more accurate, for no uncertainty occurs in finding the distance of the components. Also there are two undisturbed slopes of the doublet-curve so that the analysis can be carried out from two different sides. Therefore it was recommendable to go to a higher order.

But only for the band 3371 Å a photograph in the fourth order was possible, the other bands in this order falling too near to the grating. In this order, the maximum of the weaker component in fact is undisturbed as an analysis of five doublets showed. In this analysis the lines of the *a* series served for determining the shape of a single line.

On the other hand, the ghosts are more intense and also there was a strong continuous background throughout the whole band. In the third order of the positive bands such a background also occurs but relatively much less intense.

But at this background varies slowly in the band, it cannot greatly affect the measurements in one doublet although it mights cause systematic errors in the determination of the intensity-ratio of two different doublets.

First it has been shown that this background was due to light of the same frequency as that of the band, as its structure already leads one to suspect, for the ground has its maximum near the head of the band and decreases slowly towards both sides.

By photographing the line 3341 Å of the mercury spectrum, while keeping away the visible light and the resonance line by a nickel-oxide glass combined with a nitroso-dimethyl-anilinegelatine filter, it was shown that its background was really of the same wavelength as the line.

Beyond a certain distance from the line the ground was found to decrease almost linearly towards zero.

From this plate also the intensities of the ghost were determined. In procents of the intensity of the main line the following values were obtained: 14% for the 1st ghost, 5% for the 2nd, 3 % for the 3rd, 8 % for the 4th whereas the following ghosts were less than 3%.

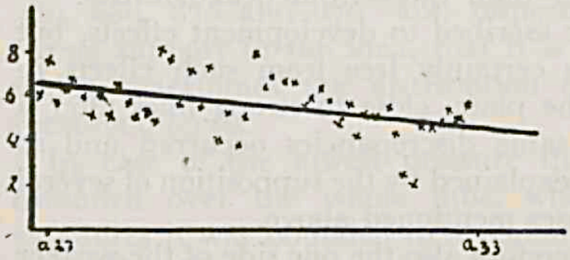

Fig. 9. Intensity of continuous background plotted on y axis against wave-length on x axis.

But once stated that the background was due to light of the same wavelength it is permitted to substract the intensity of the background from the total intensity of line together with ground, for obtaining the real intensity of the line.

That correction has been carried out in this way, that the whole continuous ground between a_{25} and a_{33} was plotted as a function of the wave-length and its mean was taken by drawing a straight line through the points.

In fig. 9 the graph of this background is reproduced.

Commonly the correction for a ground is made by substraction its value close to the lines from the total intensity.

For this procedure the assumption has to be made that the background under the line does not differ appreciably from that close near.

But this assumption is not correct in our case for there are strong variations in the intensity due to the at random-distribution of the ghosts.

For example it is visible from the fig. 3 that it should be quite incorrect to subtract the value of the ground at the right side of the doublet. In any case the mean value over a large extended part has to be taken. This was done in the described manner. The straight line was drawn also with regard to further points and other plates.

Then by measuring the plate under the comparator and also from the wavelengths measurements, the lines which had to be corrected for ghosts were determined.

As the intensity-ratio for mean line and ghost has been determined before, the intensity curve of the line reduced in this ratio could be substituted on the spot, where the ghost was

TABLE I.

Plate 1.

Line	Uncorrected Intensity	Mean of Bcakground	disturbing ghosts	Corrected Intensity	Ratio.
c_{25}	41.6 24.3	6.5	I c'_{28} c''_{28}	34.6 17.8	1.94
c_{26}			strongly disturbed		
c_{27}	34.4 18.4	6.0	IV c'_{29}	25.8 12.4	2.09
c_{28}	33.5 18.3	5.9	I $c'_{27} c''_{27}$ II $c'_{26} c''_{26}$ III b_{31}	2.50 12.3	2.03
c_{29}	25.5 14.0	5.7	IV c'_{25} c''_{25}	19.8 7.5	2.64
c_{30}	26.5 16.2	5.4	III a_{33}	21.1 10.0	2.11
c_{31}	24.0 14.8	5.2		18.8 9.6	1.95
c_{32}	22.7 12.0	5.0	IV a_{29}	15.9 7.0	2.25
c_{33}	20.8 12.0	4.5	I b_{34}	14.3 7.5	1.94
c_{34}	19.3 11.3	4.0		15.3 7.3	2.10
Plate 2.					
c_{28}	13.8 8.4	2.8	Same as in plate 1	9.8 5.6	1.75
c_{29}	12.4 7.6	2.7		9.7 4.6	2.10
c_{30}	11.8 7.1	2.6		9.2 4.0	2.30
c_{31}	10.8 8.7	2.4		8.4 3.9	2.15
c_{32}	9.7 6.7	2.3^5		6.3^5 4.3^5	1.45
c_{33}	9.7 5.4	2.2		6.5 3.2	2.04
c_{34}	8.3 5.6	2.0		6.3 3.6	1.75

expected. Then the intensity which feel under the maxima of the doublet components was corrected.

The latter correction was only a slight one, whereas that for the continuous background could reach a value up to 40 % of the weaker component.

The results of the plates worked out in detail are given in table 1.

In the third column the ghosts for which has been corrected for, are given. [1])

From these measurements a mean value of 2,11 for plate 1 and of 1,93 for plate 2 for the intensity-ratio of the components is obtained. [2]) These both values allow the value 2,0 found for the negative bands within the limits of accuracy, but exclude the values 1,7 and 3,0.

In both band systems the same value of the intensity-ratio thus has been found, although in the latter case the accuracy is not so great as in the former. [3])

OTHER MEASUREMENTS.

A research for hyperfine structure in the atomic spectrum, has also been carried out but, without success. (14) The lines which have been photographed were at 4098 Å and 4104 Å in the spectrum of $\overset{++}{N}$ corresponding to the $3^2S - 2^2P$ (15). They did not appear in the hollow cathode discharge but were strongly present in a condensed spark. Unfortunately, in this light source they appeared broadened. By lowering the pressure to 10^{-2} mm and varying the discharge conditions (series spark-length, capacity and self-induction), it was possible to get them much more narrow, but even in the most favourable conditions used, they possessed a half-width of 0,03 Å in the third order of the grating. In this order the grating possesses a resolving-power of nearly 240.000, which is not very large for hyperfine structure research,

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[1]) The intensities of the ghosts were: 14% 5% 3% 8% for the four first ghosts respectively; the following were less than 3% of the the chief line.

[2]) It is worth to notice that, if one omits the two outfalling values of c_{29} in plate 1 and c_{32} in plate 2, one gets 2,05 and 2,02 for the mean value of the ratio.

[3]) See discussion more below.

but for these unfavourable wave lengths there was no higher resolving-power available.

On the registration a slight broadening towards the longer wavelengths was observed in the density-curve. It is, however, possible that this is an ordinairy spark-broadening.

As the hyperfine structure of these lines, if present, should be probably due to that of the ^2S level, only from the intensities of the components (and not from their number) the amount of nuclear spin could have been determined.

Therefore at the same time a research was undertaken to see wether the intensity-measurements do agree with the other criteria (Multiplicity and Zeeman effect) for asigning a definite value to the nuclear spin.

The line chosen for this research was the resonance line of Thallium at 3776 Å emitted by the transition $2^2S-2^2P\frac{1}{2}$ (15).

This line shows a hyperfine structure of three components and from this structure, together with the Zeeman effect a nuclear spin of $\frac{1}{2}$ has been assigned to the nucleus. (14)

The lightsource in this case was a 440 V Pfund-arc with copper electrodes, on the cathode of which pieces of lead-Thallium-alloy were placed. (16)

In the third order of the grating all three components were sufficiently resolved. In the first order the line appeared as a doublet.

The theoretical intensities calculated on the basis of a spin $\frac{1}{2}$ for the three components are 1 : 2,0 : 1 respectively and for the doublet components in the first order 1 : 3,0.

The results showed that even in the case of an alloy containing only 10^{-5} part of Thallium[1]) and an arc-current of only 0,5 Amp. there must have occurred a strong self-absorption. The intensities found from both, peak and surface measurement, were as 1 : 1,45 : 1 [2]).

An alloy of concentration 10^{-6} gave 1 : 1,64 : 1 by which result the idea of self-absorption is strongly supported.

In the case of a concentration 10^{-7} the emission was too weak

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[1]) The thallium poorer alloys have been made by diluting an alloy with its ten or hundredfold weight of spectroscopically thallium free lead. The new alloy was then assumed to contain a 10^{-1} or 10^{-2} smaller concentration of thallium.

[2]) The self-absorption did not manifest itself in the shape of the intensity-curves, which was the same for all components.

for a third order photograph. The intensity-ratio for the doublet components in this case was 1 : 2,80.

It appears thus that the theoretical intensity-ratio is really reached in the limit for very low concentration.

But the most interesting point of this research is that even in exceedingly small concentrations there is a strong self-absorption. Since in many cases the hyperfine structure is caused by the ground-level (alkalies), great care has to be taken in drawing conclusions as to the nuclear spin from the intensity-ratio of the components and repeated measurements with varied concentration or (and) current density seem to be unavoidable.

Intensity-measurements carried out on the green Thallium line 5350 Å ($^2S \rightarrow {}^2P_{\frac{3}{2}}$) confirm the results got from the ultraviolet line.

In the second order of our grating the green line shows a doublet fine structure arising from the splitting of the S level.

Theoretically with a spin $\frac{1}{2}$ of the Tl. nucleus the intensity ratio should be 3,0 : 1, whereas the value 2,7 has been found experimentally.

As for this measurement an alloy containing 10^{-5} parts of Tl. has been used, this result indicates also a much less selfabsorption for the green line as compared to the ultraviolet line. This should be expected on account of the fact that the latter (3775 Å) is the resonance line.

Discussion of the accuracy.

For the photographic intensity measurement of an undisturbed narrow line, having a density in a not too unfavourable range of the density-curve, the mean error is about 8 %. Of course this value refers only to the kind of plates and development-process used in this research.

So for the lines of the band 3914 Å of the negative system the mean error can be set at this value. This makes a probable error of 2 % for a band measurement consisting of thirty lines.

Here a table is given for the results from the registrograms of the band 3914, all containing at least thirty lines so that the weight for all values is the same.

TABLE II.

Order of the grating	Ratio
1	2.09
1	1.98
2	2.03
2	1.96
3	2.05
3	2.10

Mean Value 2.03^5

In total taking the mean for all bands the probable error in the intensity-ratio comes out to about 1 %.

Of course there must be assumed that the errors occurring in the single results are quite unsystematical.

But the excellent concordance of the results taken from first, second and third-order exposure, which also showed the same temperature for the rotational levels, gives strong support for the correctness of that assumption. So in every case from comparation of the different values for the single plates, the accuracy (probable error) of 2 % seems to be reached.

For the other bands the accuracy is less. The band at 4278 Å is also undisturbed but as the plate was over-exposed, only 10 lines were easily measurable.

As the results from it, both, in regard to the distribution temperature [1]) and to the ratio of stronger and weaker lines, did agree with that of the 3914 Å, it was thought that no new exposure was necessary. So the accuracy for that band is 4 % to 5 %.

The bands 3884 and 4237 are strongly disturbed which reflects itself in the larger spreading of the points.

Also the errors seem to be not quite unsystematical [1]) for in both measurements of 3884 the ratio alters rom about 1,8 to 2,0 through the band. It is then difficult to give a taxation of the mean error, but if one takes the mean of the divergencies of the points, one finds about 12 % in this case for a single measurement. With this value the accuracy reached, is calculated to about 7 % for the 3884 band and 12 % for the 4237 band.

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[1]) The overlapping of the strong 3914 or 4278 band is more serious for the lines of lower quantumnumber.

For the plates taken in the fourth order of the 3371 band the mean divergence from he mean value is of about 7 % for plate one and 10 % for plate two, which should give an accuracy of the same order as in the 3914 Å band of about 3 %.

But on account of the strong corrections carried out, one can feel fear for systematic errors, although there are no indications for them in comparing the results of the two plates, whereas also no systematic discrepancy between surface- and top-intensity was observed. On the other hand if the mean of the continuous ground on the photogram was taken, the ratio for plate one comes out to 1,94.

Now there is no doubt that this latter method of correction, where the mean value over the density instead of over the intensity has been taken, is not the right one but this demonstrates the great systematic influence of the corrections on the result.

In any case the mean value of the plates never had been found to exceed the range 1,90 – 2,11, so that in each case it seems highly improbable that the true value should lie out of this interval.

For the Thallium measurements the mean error is 8 %.

As the theoretical values for the relative intensities of the two components in the first order for a nuclear spin of 1, $\frac{3}{2}$, ... are 2 : 4; 3 : 5, ... there is no doubt that of the theoretical values only that for a spin $\frac{1}{2}$ comes into account, although the value of 1 : 3,0 was not reached on account of the strong self-absorption

SUMMARY.

After an introduction the essential features of bandspectrum structure, including Raman effect, as far as necessary for an understanding of the following pages are exposed (page to). The difficulties in the interpretation of the experimental data are dealt with (page 006 to 006).

The essential points of the experimental methods are given: of the discharge tube (p. 008), of the photographic data (p. 009 to 010), of the photometring of the plates (p. 010). The reduction of the measurements giving a value 2,0 for the ratio of alternating intensities of the negative bands 3914 3884 4278 4237 Å is explained (p. 010 to 018). A Maxwell-Boltzmann distribution among the rotational levels has been found. A description of the arrangement for a research on band 3914

under varying pressure is given where no alterance of the ratio has been found. (p. 018 to 020).

In the band 3371 of the third positiv group the same value was found. (p. 020 to 024).

Measurements caried out on the thalliumline 3776 indicate a strong self-absorption, but notwithstanding the intensities seem to confirm the value $\frac{1}{2}$ for the nuclear spin p. 025.

The accuracy is discussed in p. 026 to 028. For the valuetaken from one plate of band 3914 Å a probable error of 2 % is found. The mean ratio taken from six plates is $2{,}03^5$. For the other negative bands the accuracy is 4 % to 8 %. In the band 3371 the ratio lies in the range 1,90 to 2,10. The thallium measurements are affected with a probable error of 6 %.

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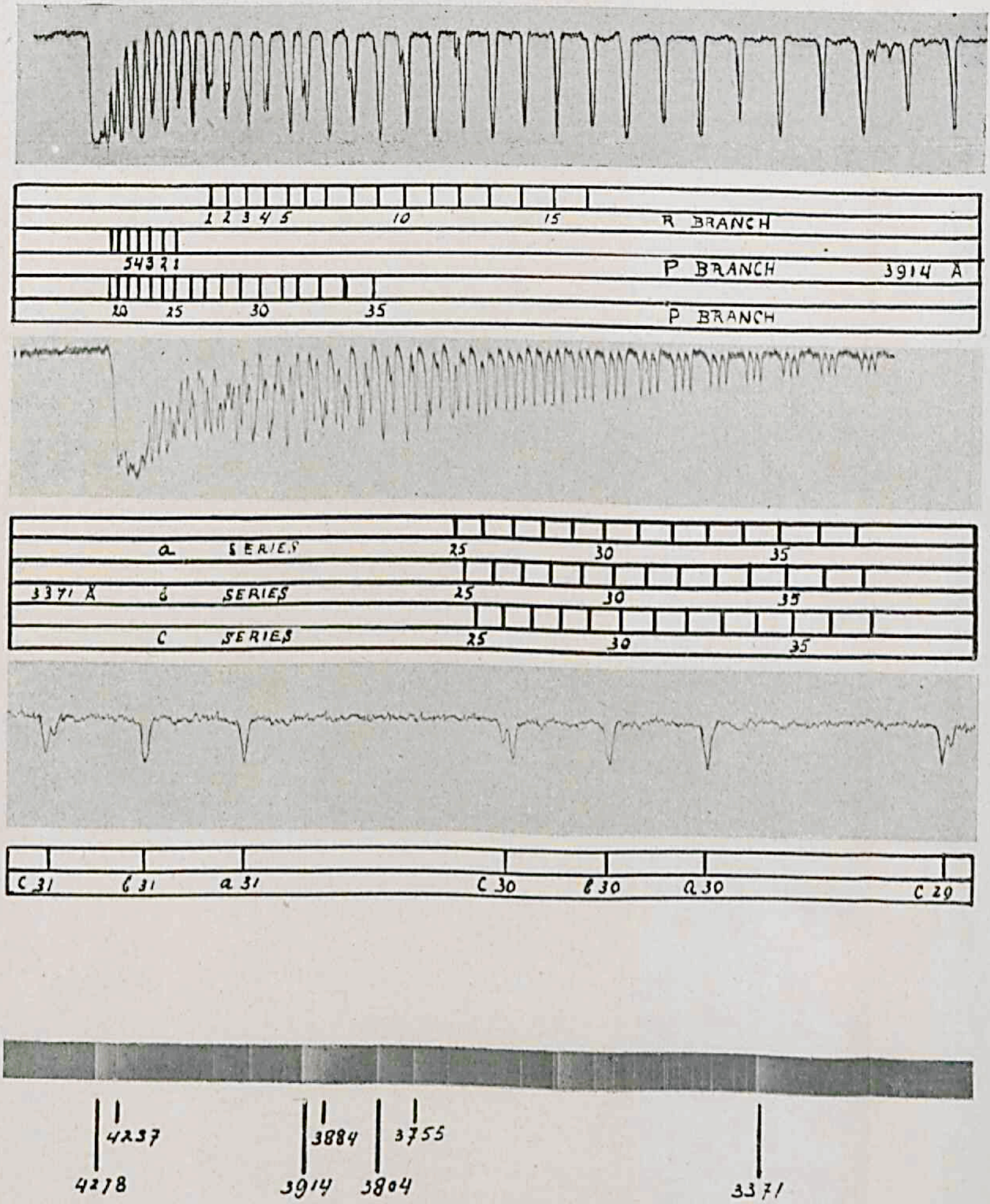

Fig. 2. *a*. photometer-trace of band 3914 Å 1st order.
b. photometer-trace of band 3371 Å 1st order.
c. photometer-trace of band 3371 Å 4th order.
d. Low dispersion-photograph showing the different bandheads.

STELLINGEN

Terecht merkt K. T. Compton op, dat [illegible] van een electrische ontlading [illegible] is.

K. T. Compton, Phys. Rev. [illegible]

[illegible]

De bewering van [illegible]

[illegible]

De door hen gevonden [illegible]

[illegible]

[illegible]

[illegible]

[illegible]

STELLINGEN.

Terecht meent K. T. COMPTON, dat de ionisatie van het gas tusschen de electroden van een electrischen lichtboog overwegend van thermischen oorsprong is.

K. T. COMPTON, Phys. Rev. 21, 266, 1923.

De beperking dat een volume element vele moleculen moet bevatten, zooals in de leerboeken der kinetische gastheorie wordt vooropgesteld, is in de meeste gevallen overbodig.

Bovendien geeft de daarop berustende wijze van behandeling der gaswetten aanleiding tot misverstand.

De bewering van TAMM dat de behandeling volgens het correspondentie principe van de incohaerente lichtverstrooiing niet tot ondubbelzinnige resultaten voert is onjuist.

TAMM, Zt. f. Phys. 62, 545, 1930.

De waarde die JENKINS en HARVEY voor het kernmoment van Lithium (3/2) opgeven is vermoedelijk te groot.

De door hen gebruikte methode van intensiteitsmeting is in het algemeen niet van toepassing op een absorptie spectrum.

HARVEY and JENKINS, Phys. Rev. 35, 789, 1930.

Bij voorkeur zou men hyperfijnstructuur onderzoekingen in een bandenspectrum moeten verrichten.

Zeer ten nadeele van de practische bruikbaarheid hebben de meeste medewerkers aan het „Handbuch der Physik" en het „Handbuch der Experimentalphysik" het idee „leerboek" vooropgesteld.

Tegen de hypothese van ROSENTHAL over den oorsprong der coronalijnen zijn nog andere bezwaren dan de door hem zelf genoemde aan te voeren.

ROSENTHAL, Zt. f. Astrophys. 1, 115, 1930.

Bertrands bezwaar tegen de oorspronkelijke afleiding van de Maxwellsche wet der snelheidsverdeeling berust op misverstand.

Bertrand, Calcul des Probabilités, bl. 29, 1907.

De afwijzende houding, die in de Physica tegen door sommatie van niet convergente reeksen verkregen resultaten wordt aangenomen is niet gerechtvaardigd.

Zie b.v. Szegö in Riemann-Weber Differentialgl. d. Physik. I, bl. 148, 1925.

In het lemma I § 619 bladz. 545–546 Goursat III 1923 zouden de woorden „ainsi que sa dérivée" beter achterwege gelaten kunnen worden.

[illegible] de [illegible] van een [illegible] vertraagd.

[illegible]

[illegible]

[illegible]

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