SPECTROMETRIC PLASMA DIAGNOSTICS

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ABSTRACT

Some plasma chemistry work done in France during the past three years is described: measurements of transition probabilities and line profiles by J. Chapelle; calculations on non-LTE hydrogen and helium plasmas by H. W. Drawin; and application of N_2^+ and N_2 bands in the diagnosis of nitrogen plasma jets as treated by P. Fauchais.

ABSOLUTE INTENSITY OF A LINE

The measurement of absolute line intensity is a useful tool in plasma diagnostics because we may choose the line and the emitting atom. The emissivity ε_v (power radiated per unit volume and unit solid angle in a line of frequency v) cannot be measured directly, but it can be obtained from the luminance of the plasma I_v .

Let us recall the well known relations for a homogeneous and optically thin plasma of thickness d: $I_v = \varepsilon_v \times d$. For a cylindrical plasma I_v is related to ε_v by the Abel integral

$$I_{\nu}(y) = 2\int_{0}^{x} \varepsilon_{\nu}(r) \times dx = 2\int_{0}^{R} \varepsilon_{\nu}(r) \times r \times (r^{2} - y^{2})^{-\frac{1}{2}} \times dr$$

with the usual notation¹.

When ε_v is known we can calculate the population density N_u of the level u (energy E_u) of the emitting atom if the transition probability A_{ul} of the line is known.

There are large discrepancies (80 per cent) amongst the experimental values of A_{ul} found in the literature; furthermore they sometimes disagree with the theoretical values proposed. The most reliable values were selected by Wiese in 1969². J. Chapelle and co-workers have used an argon plasma jet produced by a d.c. arc for measuring transition probabilities of 41 ArI lines and 23 ArII lines^{3, 4}.

The relative values of transition probabilities of ArI lines agree with values selected by Wiese; but the absolute values found are smaller by 30 per cent on the average for $5p \rightarrow 4s$ transitions, discrepancies being smaller for other transitions with + and - signs⁴. A relative error of 30 per cent in A_{ul} gives an error smaller than three per cent in the temperature.

Values obtained from our measurements on the ArII lines differ from Wiese's values by less than 20 per cent, except for three lines³, for which discrepancies reach 50 per cent.

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From transition probabilities P, Ranson calculated the lifetime of levels 4p of ArI (2p₉ and 2p₆ in Paschen notation); these values were compared with values measured by the Hanle effect, and it was found that Wiese's values are rather high (factor 1.1) whereas Ranson's are rather low (factor 0.9)⁴.

The accuracy of A_{ul} measurements strongly depends on the plasma used. The plasma must have a good stability and cylindrical symmetry for applying the Abel integral, the N_e and T distributions must be known exactly, and the lines must not have any self-absorption. Plasma jets used in Orleans are appropriate for measuring A_{ul} ; however, we might think that it would be better to use a wall stabilized Maecker's arc, in which the plasma is very homogeneous along the axis. By observing the plasma in the axial direction we would avoid the Abel integral which brings out inaccuracy⁵. This advantage is disappointing, however, because the refraction of light at small axis angles can lead to errors in the luminance measurements. The refraction is so constraining that we could not make absorption measurements with a Maecker's arc, and self-absorption could not be verified.

PROFILES AND WIDTHS OF LINES

Using a profile of a line or measuring its width offers two advantages: only relative intensity measurements are needed, and the electronic density N_e is characterized even in non-LTE plasmas. Of course, there are some precautions to be taken with optical thickness in the centre of the line, homogeneity of the plasma, and the resolution of the spectrometer.

In the laboratory, plasma lines are broadened by the statistical Doppler effect and by the Stark effect; the latter is predominant as long as the ionization rate is higher than one per cent. For such measurements hydrogen lines are often used, mainly H_{β} which presents several advantages: calculations which relate its profiles to N_{e} are reliable and allow one to determine N_{e} with an error smaller than five per cent. H_{β} is very intense and much broadened so that it can be used over a wide range of N_{e} : 10¹⁴ to 10¹⁷ cm⁻³ if the spectrometer has normal resolution, 150000 in the fifth order.

 N_e is related to the line width $\Delta\lambda_{\downarrow}$ by $N_e = C(N_e, T) \times \Delta\lambda_{\downarrow}^{\frac{3}{4}}$, the coefficient C depending only slightly on T. C has been tabulated by Griem⁶. Results of such measurements give N_e , whether the plasma is in LTE or not, and whatever its temperature or chemical composition may be.

Other HI lines may be used, as well as HeII lines for $5000 \le T \le 80000$ K and $10^{15} \le N_e \le 10^{19}$ cm⁻³.

Griem has also calculated profiles of 'isolated' lines of a large number of atoms and ions; but for atoms which have complex electronic structures, several approximations are made, and a comparison between theory and experiment is required. Therefore, in Orleans, numerous studies of profiles of ArI and ArII lines, and some HeI, MgII and CaII lines, have been made.

Agreement between values calculated by Griem in 1964⁶ and ours is not too bad (20 per cent); HeI line profiles we observed⁷ fitted Griem's profiles corrected in 1971⁸; but there are large discrepancies between values found for some ArII, MgII and CaII lines (factor two to five on widths). J. Chapelle compared his experimental result with profiles calculated by Brechot-Sahal and Van Regemorter in Meudon⁹, and found that their more refined theory[†] is better.

Allowance must be made for plasma inhomogeneity; for this purpose, the apparent profile is calculated with theoretical profiles corresponding to each point of the N_e and T known distributions, then this profile is compared with the measured one. Indeed the observed profile of a line is the superposition of all the profiles emitted by regions having different electron densities; therefore the observed profile is strongly influenced by emission from the region of highest electronic density, so that the N_e value obtained from the observed profile agrees well with the highest N_e value measured along the axis of the plasma jet.

Our conclusions about the lines we observed are as follows: the line widths calculated by Griem in 1964 or in 1971 are good enough for neutral atoms, even if more refined theories give better results. But for ions the calculated line widths are to be used with caution.

BOLTZMANN DIAGRAM AND LTE DEVIATIONS

Absolute intensities of lines give us population densities N_u of the atom, and line widths give the electronic density N_e . Then applying Boltzmann and Saha laws, we can calculate the excitation temperature T_{exc} from N_u values and an ionization temperature from N_e :

$$N_{u} = g_{u} \frac{N(T)}{Z(T)} \exp\left(-E_{u}/k_{B}T_{exc}\right)$$

$$N_{e} = 2 \frac{g_{z+1,1}}{g_{z,j}} \frac{N_{z,j}}{N_{z+1,1}} (2mk_{B}T/h^{2})^{\frac{3}{2}} \exp\left(-\frac{E_{z,\infty} - E_{z,j} - \Delta E_{z,\infty}}{k_{B}T_{Saha}}\right)$$

(in the usual notation¹.)

In LTE plasma: $T_{exc} = T_{Saha} = T_e$, and it is often easier to get T_e from the intensity ratio of two lines, or better with the Boltzmann plot of log $(\varepsilon_v/A_u \times g_u)$ versus E_u .

It can be shown that use of the intensity ratio of two lines may lead to serious error and that the Boltzmann plot must be used with caution, as shown by the work of H. W. Drawin on LTE deviations in hydrogen and helium plasmas.

First let us consider as an example the plasma we observed 4 cm away from the anode of an argon plasma jet generator¹⁰. A Boltzmann plot was made covering a wide range of energy E_u starting from the 4p levels located just above the ArI metastable levels; the plot is straight and gives a temperature (called T_p) of 4800 K, very near the temperature T_n of neutral Ar atoms, the latter being measured by interferometry. Since T_e is normally almost equal to T_n , we are tempted to define LTE at 4800 K; unhappily having a straight Boltzmann diagram is insufficient for concluding that a LTE plasma exists. Absolute intensities of ArI give us population densities N_u , and by extension of the Boltzmann law down to metastable levels, we get

[†] They take account of the Coulomb attraction (hyperbolic trajectories of electrons).

 $N_{\rm m} = 6.5 \times 10^{14} {\rm cm}^{-3}$, a value which corresponds to $T_{\rm exc} = 10^4 {\rm K}$, or twice $T_{\rm B}$. (In a LTE plasma at 4800 K, $N_{\rm m} = 2.2 \times 10^4 {\rm cm}^{-3}$). The plasma jet behaves like a tank of metastable Ar atoms in partial

The plasma jet behaves like a tank of metastable Ar atoms in partial LTE with all upper excited levels, free as well as bound levels, as $N_{\rm e}$ measurement shows; but the ground level is not in equilibrium with the others, because the plasma is a recombination plasma in a non-stationary state.

Let us note that for studying LTE conditions thoroughly the Boltzmann plot must be extended down to the ground level, which requires absorption line intensity measurements. Such measurements in the visible range allowed P. Ranson to obtain N_m directly by experiment, and corroborate previous conclusions¹¹.

LTE is realized if all transitions (bound-bound, bound-free, etc.) are due mainly to electronic collisions and not to radiative processes or diffusion of atoms. In homogeneous plasmas the LTE criterion is $N_e > N_e^*$ where the critical value N_e^* may be calculated theoretically. In this calculation one must take account of resonance absorption, which is defined by an optical escape factor Λ^{\dagger} .

Griem has given a first evaluation of N_{e}^{*} , supposing $\Lambda_{12} = 0.1$, and has found $\simeq 10^{16}$ cm⁻³ in Ar plasma; by studying an argon plasma jet at low pressure ($\simeq 4$ torr), we observed LTE at an electronic density as low as 5×10^{14} cm^{-3 12}. Our result is explained if we suppose $\Lambda_{1j} = 0$ for the first six resonance Ar lines. Drawin treated LTE deviations in hydrogen and helium plasmas in homogeneous stationary states¹³⁻¹⁴ by making several choices of Λ_{1j} values. His results are shown in *Figure 1* in the case of a hydrogen plasma at 12000 K; we see in it the ratio $b_1 = N_1/N_{1 \text{ LTE}}$ versus N_e and we note how N_e^{*} varies.¹⁵

For the case of a helium plasma, Drawin calculated optical escape factors and then treated LTE deviation with less arbitrary choice of Λ values; three cases were treated: optically thin plasma, plasma slightly optically thick and strongly absorbing plasma. The ratio $b_e = N_e/N_{eLTE}$ is given versus N_1 at several temperatures in *Figure 2*, and in *Figure 3* N_e^* values are given with a parameter x which is the relative deviation of N_1 from LTE.

More details about calculation of optical escape factors made by Drawim¹⁶ cannot be given here. For example some of these results are shown in *Figure 4* for $N_e = 10^{15}$ cm⁻³, $T_n = 2 \times 10^4$ K; Λ_{1j} depends on optical thickness $\hat{\tau}_{12}$ at the centre of the first resonance line $(2^{1}p \rightarrow 1^{1}s)$. $\hat{\tau}_{12}$ may be calculated if N_1 , N_e and T_n are known. At atmospheric pressure and 2×10^4 K; $\hat{\tau}_{12} = 10^4$ to 10^5 when the plasma thickness is 1 to 10 cm, and it is found that the first seven or eight resonance lines are absorbed.

Drawin's results show that it is possible to make a self-consistent calculation of HeI population densities in homogeneous stationary plasmas when the temperature and total pressure are given. At low pressure such a plasma is in a non-LTE state due to escaping radiation; only the first resonance lines are absorbed, so that the ground level and eventually the first excited levels are overpopulated.

In a non-stationary plasma, LTE can be perturbed by other causes, as

[†] Λ_{1j} is the part of the radiation in the $j \rightarrow 1$ resonance line which escapes from the plasma. If the plasma is optically thin for all lines, all Λ are 1.

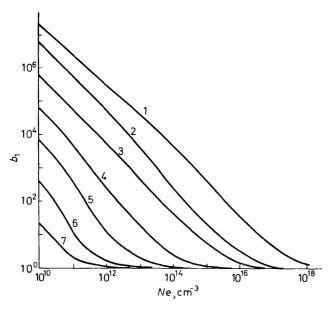


Figure 1. Homogeneous stationary hydrogen plasma at 12000 K. Ratio of ground level density in homogeneous stationary state and in LTE. After H. W. Drawin¹⁵.

- 1: all transitions optically thin
- 2: Lyman α optically opaque ($\Lambda_{12} = 0$)
- 3: Lyman series optically opaque ($\Lambda_{ij} = 0$)
- 4: Lyman series, Lyman continuum, and H_a line optically opaque $(\Lambda_1 = \Lambda_{1i} = \Lambda_{23} = 0)$

5: $A_1 = A_{1j} = A_2 = A_{2j} = 0$ 6: $A_1 = A_{1j} = A_2 = A_{2j} = A_3 = A_{3j} = 0$ 7: $A_1 = A_{1j} = A_2 = A_{2j} = A_3 = A_{3j} = A_4 = A_{4j} = 0$

we have just seen in the case of a recombination plasma. Drawin has also shown that diffusion processes disturb LTE¹⁷. His results seem important, because they indicate modification of the Boltzmann diagram, confirming that the intensity ratio of two lines can give wrong values of $T_{\rm p}$.

For hydrogen and a large number of other elements, impact cross sections increase and transition probabilities for spontaneous emission decrease as higher excited levels are approached, so that one gets LTE above some excited level h for a given N_e value. Drawin calculated population densities of the HI atom taking account of a diffusion flow of atoms in the ground state; such a flow overpopulates not only the ground level but also the first excited levels. Partial LTE is driven off to a higher h level. At low electron density the slope of the Boltzmann diagram no longer yields T_{e} .

A Boltzmann plot is shown in *Figures 5* and 6 for two optically thin plasmas: $N_e = 10^{12} \text{ cm}^{-3}$, $T_e = 6 \times 10^3 \text{ K}$ and $N_e = 10^{14} \text{ cm}^{-3}$, $T_e = 9 \times 10^3 \text{ K}$. In the first case, for instance, partial LTE would be seen at h = 5, but if $N_1 = 10 \times N_1^{\text{HS}}$ due to diffusion processes, the first points in the diagram seem to be in a straight line and give a temperature value of 16700 K, that is 2.8 $\times T_{\text{e}}$. (N_1^{HS} is the ground level density in the homogeneous stationary

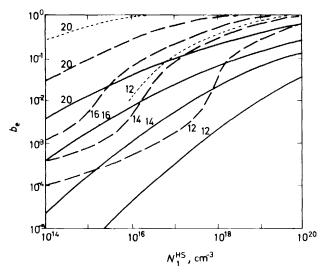


Figure 2. Homogeneous stationary helium plasma: ratio of electron density in homogeneous stationary state and in LTE. After H. W. Drawin¹⁴. Temperature is indicated in 10³K on each curve: ---- optically thin plasma, --- slightly optically thick plasma, strongly absorbing plasma.

plasma.) In the second case LTE would be reached at h = 3; when $N_1 = 10 \times N_1^{\text{HS}}$, the deviation from LTE will be clear if the Boltzmann diagram is drawn; but if the intensity ratio of H_{α} and H_{β} only is measured a temperature of 12600 K (1.4 $\times T_{\alpha}$) is obtained.

Thus the use of Boltzmann plots can be difficult when diffusion processes occur. Such diffusion often interferes in flames and plasmas and may explain an abnormally high rotational temperature observed in OH bands, and a Boltzmann diagram with two distinguishable regions of uniform slope.

METHODS USING N⁺₂ AND N₂ ELECTRONIC BANDS

When the temperature of a plasma does not exceed 15000 K its spectrum can include molecular bands, the examination of which may allow us to obtain the temperatures of rotation, vibration, or electronic excitation of molecules.

Several methods use bands as well as lines: absolute intensity of a rotational line or of a vibrational band, intensity ratio of two lines or two bands, and Boltzmann plots. The methods used and the results obtained depend not only on molecular species and observed bands, but also on instruments and mainly on the spectrometer resolution.

When there are nitrogen and carbon in the plasma, its spectrum shows high intensity CN bands, chiefly the violet electronic system $B^2\Sigma \rightarrow X^2\Sigma$. Since the work of Smit and Spier in 1942, this system can be used in several ways: profile of 0–0 band, intensity ratio of 0–1 and 1–2 bands, intensity ratio of heads of 0–0 and 1–1 bands or 0–1 and 1–2 bands, in addition to absolute and relative intensities of rotational lines¹.

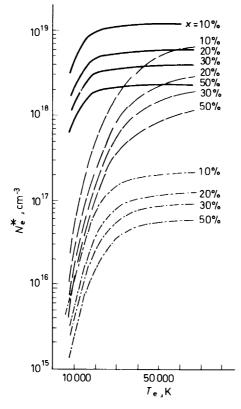
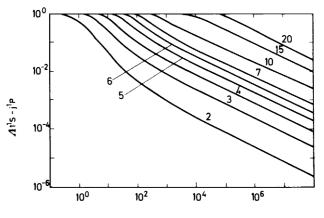


Figure 3. Helium plasma. Critical values Ne* for LTE. —— optically thin plasma, —— slightly optically thick plasma, —— strongly absorbing plasma. x means the maximum deviation of $N_1^{\rm HS}$ and $N_{\rm ILTE}$ values.



7 1'S - 2'P

Figure 4. Optical escape factors in helium plasma, after H. W. Drawin.¹⁶ $N_e = 10^{15}$ cm⁻³, $T_n = 2 \times 10^4$ K. Only Λ_{1j} (for resonance lines) are shown.

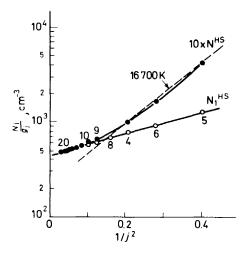


Figure 5. Boltzmann diagram showing deviation from LTE in diffusion dominated hydrogen plasma. After H. W. Drawin¹⁷. $N_e = 10^{12} \text{ cm}^{-3}$, $T_e = 6 \times 10^3 \text{ K}$, all $\Lambda_{1j} = 0$, $N_1^{\text{HS}} = 7.18 \times 10^{17} \text{ cm}^{-3}$, $b_1^{\text{HS}} = 2.95 \times 10^3$.

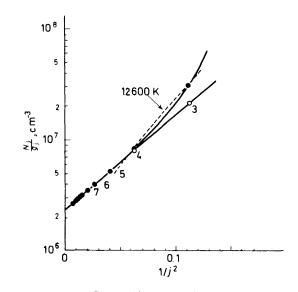


Figure 6. As Figure 5, but: $N_e = 10^{14} \text{ cm}^{-3}, T_e = 9 \times 10^3 \text{ K}, N_1^{\text{HS}} = 3.05 \times 10^{15} \text{ cm}^{-3}, b_1^{\text{HS}} = 14.94.$

The work of Fauchais on the first negative system $B^2 \Sigma_u^+ \rightarrow X^2 \Sigma_g^+$ of N_2^+ will be discussed; this system is as intense as the CN one and it is nearly always seen in plasmas containing nitrogen. P. Fauchais and J. M. Baronnet used a d.c. arc nitrogen plasma jet. The plasma is in LTE¹⁸, and the temperature of the region observed lies in the range 4000–12 500 K.

The methods which were used are as follows: absolute intensity of rotational lines and of heads of 0-0 and 0-1 bands, intensity ratio of two lines in the 0-0 band, intensity ratio of two groups of lines in the same band.

Absolute intensities of heads[†] of 0–0, 0–1 and 0–2 bands are given versus T in *Figure* 7; we see that the method is accurate below 7000 K, and cannot be used above 8000 K.

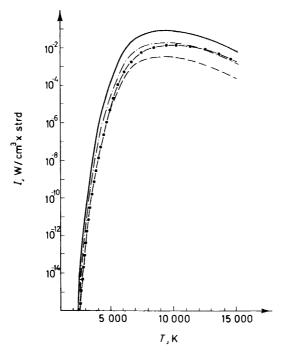


Figure 7. Absolute intensities of band heads, after J. M. Baronnet and P. Fauchais^{19, 20}. First negative N₂⁺ system: ---- 0-0 band, ---- 0-1 band, ---- 0-2 band. Second positive N₂ system: ---- 0-0 band

If band heads are overlapped by lines or bands of other molecular species (mainly overlapping of N_2^+ 0–0 band and CN 0–0 band) rotational lines can be resolved with a spectrometer of rather low resolution (20000) and the absolute intensity of a line, such as line 72 P at 3835.1 Å, can be measured (*Figure 12*).

In order to avoid absolute intensity measurements, needing an energy gauged spectrometer, the intensity ratio of two lines may be used, with the same precautions as before. For good accuracy, the quantum numbers K of lines must be quite different; due to the perturbation of the 0–0 band at K = 39-40 and 65–66, the lines will be those for $30 < K_1 < 38$ and $70 < K_2 < 78$. As shown in *Figure 8*, in the case of the lines 34 and 76, the intensity

[†] Here the band head is the spectral range of the 0-v' band which is not overlapped by the 1-v' band.

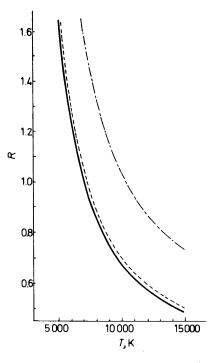


Figure 8. Intensity ratio of the two lines K = 34 and K = 76 of the N_2^+ 0-0 band. After Baronnet and Fauchais¹⁹. — calculated taking account of 1-1, 2-2 and 3-3 band overlapping, ---calculated taking account of only 1-1 band overlapping, ---- calculated without band overlapping.

ratio must be calculated taking account of 0–0, 1–1 and 2–2 bands overlapping. R values of other pairs of lines are given in Figure 9^{19} . An error of ten per cent in R gives an error of about ten per cent in the temperature up to 8000 K and thirteen per cent at 11 500 K.

The main error in measurement of R comes from the difficulty in determining the line wings.

This error is reduced if the intensity ratio of two groups of lines can be measured. For the highest sensitivity, the two groups of lines must be in the same band. Figure 10 gives R values for the two groups $K_1 = 32$ to 38 and $K_2 = 70$ to 76 in the 0–0 band. Here again, calculated R values must take account of band overlapping. An error of ten per cent in R gives an error of twelve per cent in T at 8000 K and fifteen per cent at 11 500 K.

The results obtained by all these methods are compared in Figure 11¹⁹.

Fauchais has also studied the use of the second positive system of N_2^{20} . His results show that absolute intensities of the head[†] of the 0–0 band and of several lines can be used.

[†] The R branch of the 0–0 N₂ band is not easily seen, so that here the band head is defined by all lines between 3371.3 and 3361.4 Å.

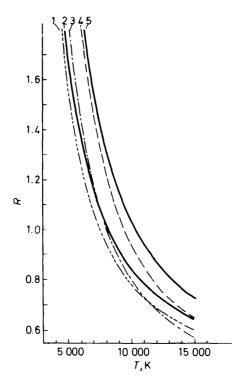


Figure 9. Intensity ratio of two lines of N_2^+ 0–0 band. After Baronnet and Fauchais¹⁹. 1, lines 38 and 72; 2, lines 34 and 70; 3, lines 30 and 74; 4, lines 32 and 78; 5, lines 36 and 78.

The intensity ratio of lines gives a 'rotational' temperature $T_{\rm rot}$ if LTE is established for rotational levels. Since molecular rotations are characteristically strongly coupled to translations by molecular collisions, $T_{\rm rot}$ often gives the gas temperature whatever the nature of band excitation. So, N_2 bands are excited by Ar metastable atoms outside an Ar plasma jet which flows out in the air, and they give a rotational temperature $T_{\rm rot}$ near the neutral one T_n^{20} . Also, N_2^+ bands excited by an electron beam allow us to measure the gas temperature in a low pressure wind tunnel²¹.

In the case of cylindrical plasmas the Boltzmann plot and intensity ratio methods become too complicated, and Abel transformations are needed. Chevaleyre in Lyon calculated how the Boltzmann diagram is modified in the case of a cylindrical flame with three kinds of temperature distributions: constant to a distance r from the axis and decreasing in a layer of thickness R - r with a linear, parabolic, or gaussian law. The Boltzmann plot remains straight, and a correction appears on $T_{\rm B}$ which is less than three per cent if (R - r)/r < 1, and which reaches between ten and fourteen per cent if $r = 0^{22}$.

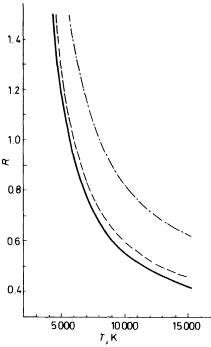


Figure 10. Intensity ratio of two groups of lines: lines 32 to 38 and lines 70 to 76 of N_2^+ 0–0 band. After Baronnet and Fauchais¹⁹ (the three curves calculated as in Figure 8).

USING THE KIRCHHOFF LAW

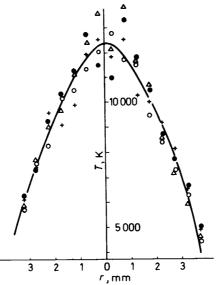
Finally, mention is made of a spectrometric method quite common in flames which has been employed for a long time. This involves application of Kirchhoff's law to measurements of the luminance I_v and the absorption coefficient α_v .

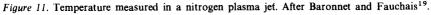
Most spectrometric methods require avoidance of self-absorption of radiation, which is perhaps why methods using Kirchhoff's law are so rarely used in plasmas. In many plasmas measurable absorptions are observed either at the centre of some lines, or in bands, or even in infra-red continuum spectra. In order to obtain an accurate measurement, the absorption coefficient must be between 0.05 and 0.95 (optical thickness between 0.05 and 3).

From Kirchhoff's law, $I_{\nu}(T) = \alpha_{\nu} \times I_{\nu}^{0}(T)$, the temperature T is deduced (I_{ν}°) is the black body luminance).

This method is convenient for plasmas with a dispersed solid phase in thermal equilibrium with the plasma, like luminous flames with soot particles. Above all, it is quite suitable for a medium temperature range: 10^3 to 5×10^3 K, although it has been applied up to 10^4 K¹.

SPECTROMETRIC PLASMA DIAGNOSTICS





- \bigcirc from intensity ratio of two groups of lines of N₂⁺ 0-0 band
- + from intensity ratio of two groups of hier of N_2^+ 0-0 band \triangle from absolute intensity of the N_2^+ 0-0 band head from absolute intensity of the N_2^+ 0-1 band head

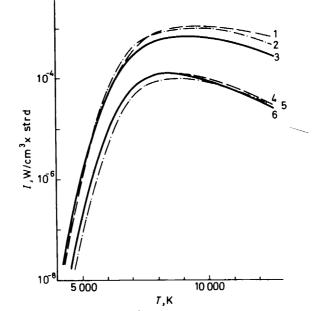


Figure 12. Absolute intensities of lines of N_2^+ and N_2^- 0–0 bands. After J. M. Baronnet et al.²⁰. N_2^+ 0-0 band: 1, line K = 26, 2, line K = 46, 3, line K = 72; N_2 0-0 band: 4, line $R_2 = 20$, 5, line $R_2 = 37$, 6, line $R_2 = 60$.

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