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STUDY OF THE EVOLUTION OF THE DENSITY OF METASTABLE ATOMS FOLLOWING INTERRUPTION OF AN ARC

S. VACQUIE, J. P. DINGUIRARD, H. KAFROUNI and I. PAGES

Equipe de Recherche Associée au CNRS No. 217, Centre de Physique Atomique, 118, route de Narbonne 31077 Toulouse Cedex, France

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Abstract—We have measured the density of four metastable levels of argon during extinction of an arc discharge at atmospheric pressure. Complete LTE was not attained and overpopulation of metastable levels was observed. The metastable levels are depopulated at a lower rate than the radiating levels.

1. INTRODUCTION

THERE are only few methods available for measuring the characteristic plasma dimensions of an extinguishing arc at atmospheric pressure. Application of spectroscopic methods may necessitate the assumption of LTE between electrons, ions and neutral atoms. It is clear that if there are important LTE departures, the values obtained could be inaccurate and the notion of temperature would not apply. Complicated processes must be employed to describe the system.[1] For diffusion-controlled plasmas, an analogy has been established between the density decay at a given instant and the time decay at a given point; for plasmas in extinction, the existence of LTE must also be verified everywhere. For reasons of convenience, partial (or complete) LTE is often assumed to be present.[2]

LTE is easily verified for radiating levels but not for metastable levels. If we assume that LTE exists at all times, we must also accept the existence of the microreversibility of the collision processes. It is therefore not certain that a population of metastable atoms has a longer lifetime than that of radiating atoms, in spite of the very long lifetime of all isolated particles. It is, however, very important to have a precise idea of the behavior of the population of these atoms because they play a determining role in the transport of energy, as well as in the reignition of a discharge, since they represent a reservoir of potential energy. Hence this work has a double purpose. We intend to measure in the decaying plasma of an arc at atmospheric pressure the evolution of the density of atoms excited at the $3p^54s$ levels, and to see if they are in LTE with the electrons. We will also compare their rate of change with that of a radiating level.

2. METHOD FOR DETERMINING THE DENSITIES OF METASTABLE ATOMS

3.2 *Basis of the method*

Figure 1 shows the first energy levels of the argon atom. We have studied the transitions between the $3p^54p$ levels and the metastable and resonant $3p^54s$ levels. The corresponding lines are self-absorbed in the discharge. They have been used to measure the densities of the metastable atoms. The method involves measurements of the attenuation of radiation at the transition under investigation.[3] Attenuation will be proportional to the density of the $4s$ level.

The stationary state. The density n_m of the metastable atoms is related to the monochromatic absorption coefficient k_λ of the line corresponding to the $4p$-$4s$ transition by

$$\int_{-\infty}^{+\infty} k_\lambda \, d\lambda = \frac{8\pi c}{\lambda_0^4} \frac{g_n}{g_m} n_m A_{nm} \left(1 - \frac{g_m}{g_n} \frac{n_n}{n_m}\right), \tag{1}$$

where g_m and g_n are the statistical weights of the levels, λ_0 is the central wavelength of the radiation, n_n is the number density of excited atoms at level n, and A_{nm} is the Einstein transition probability from state n to m.

Figure 2 shows the method used to measure the transmission coefficient, $\tau(x, \lambda)$, of a

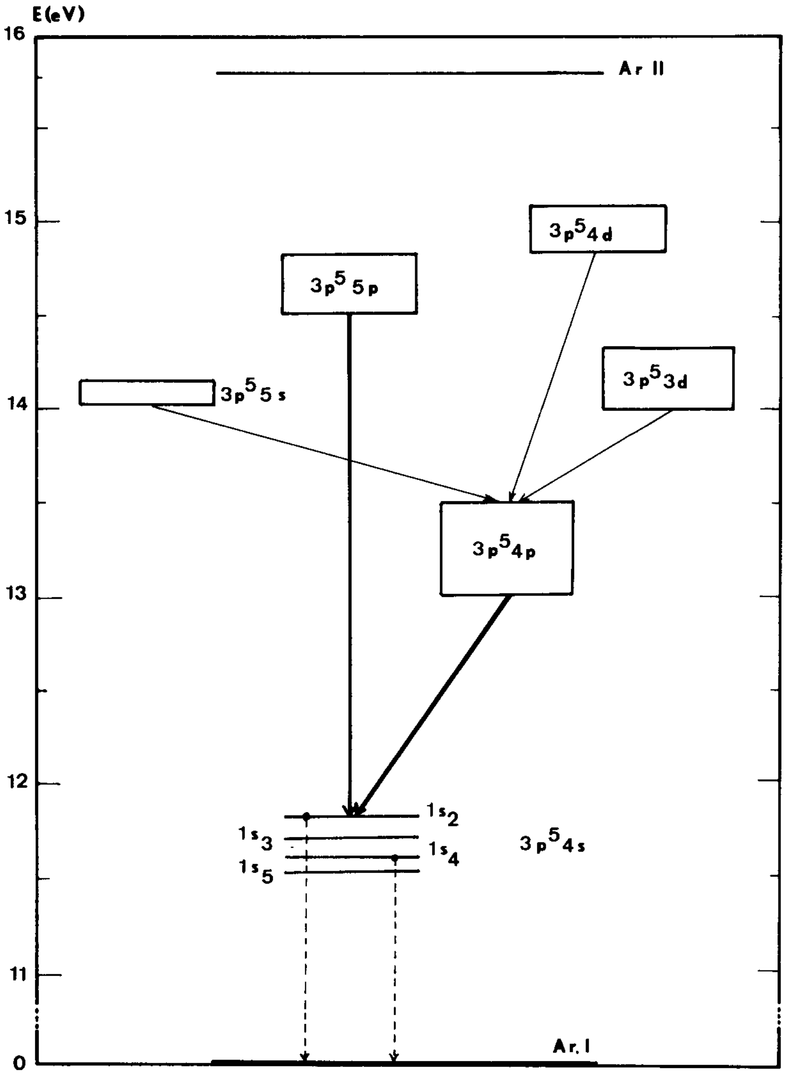

Fig. 1. Schematic representation of energy levels of the argon atom.

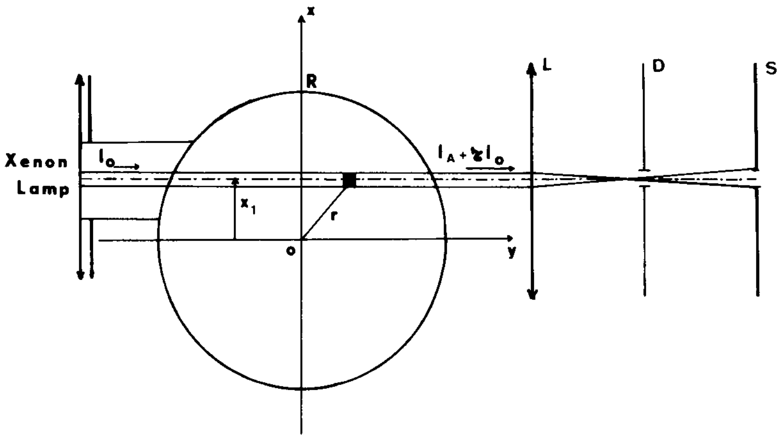

Fig. 2. Measurement of the transmission coefficient of the arc.

discharge with an axis of symmetry, thus allowing the determination of $k(r, \lambda)$. For the x_1-ordinate, the transmission coefficient is obtained by measuring the attenuation of I_0, viz.

$$\tau(x,{}_\lambda) = \frac{(I_0 + I_{\rm arc})_{x_1} - (I_{\rm arc})_{x_1}}{I_0}. \tag{2}$$

By definition, we have

$$\tau(x,\lambda)=\exp\left[-2\int_x^R k(r,\lambda)\frac{r\,\mathrm{d}r}{(r^2-x^2)^{1/2}}\right];\tag{3}$$

$k(r, \lambda)$ is given by the Abel inversion

$$k(r,\lambda)=-\frac{1}{\pi r}\frac{\mathrm{d}}{\mathrm{d}r}\int_r^R -\log\tau(x,\lambda)\frac{x}{(x^2-r^2)^{1/2}}\,\mathrm{d}x.\tag{4}$$

The transient state. The time must be considered as a third variable. When the induced emission is neglected, eqn (1) can be written as

$$n_m(t)=\frac{8\pi c}{\lambda_0^4}\frac{g_m}{g_n}\frac{1}{A_{nm}}\left[\int_{-\infty}^{+\infty} k(r,\lambda,t)\,\mathrm{d}\lambda\right].\tag{5}$$

One must then measure $k(r, \lambda, t)$ or, preferably, $\int_{\Delta\lambda} k(\lambda)\,\mathrm{d}\lambda$ directly.

On opening the exit slit of the spectroscope in such a way as to let the entire $4p$-$4s$ line pass, the photomultiplier will receive a flux equal to the total intensity of the line. This opening corresponds to a $\Delta\lambda$ wavelength interval. The transmission coefficient obtained is related to the monochromatic coefficient by the expression

$$\tau(x,\Delta\lambda,t)=\frac{1}{\Delta\lambda}\int_{\Delta\lambda}\tau(x,\lambda,t)\,\mathrm{d}\lambda.\tag{6}$$

The values so obtained are not monochromatic. It is therefore not possible to relate (by Abel's inversion) $\tau(x, \Delta\lambda, t)$ to $k(r, \lambda, t)$ and thus to n_m. Nevertheless, if the value of τ is close to unity (i.e. if the line is weakly absorbed), one can relate the integrated value $\tau(x, \Delta\lambda, t)$ to the local value $\int_{\Delta\lambda} k(r, \lambda, t)\,\mathrm{d}\lambda$.

By expanding the expression (3) into a series, we find in first order that

$$\tau(x,\lambda,t)=1-2\int_x^R k(r,\lambda,t)\frac{r}{(r^2-x^2)^{1/2}}\,\mathrm{d}r.\tag{7}$$

The Abel inversion is thus possible. If we write

$$h(x,\lambda,t)=2\int_x^R h(r,\lambda,t)\frac{r}{(r^2-x^2)^{1/2}}\,\mathrm{d}r,\tag{8}$$

we can integrate both sides of eqn (8) over the $\Delta\lambda$ interval, viz.

$$\int_{\Delta\lambda} h(x,\lambda,t)\,\mathrm{d}\lambda=2\int_x^R\left[\int_{\Delta\lambda} k(r,\lambda,t)\,\mathrm{d}\lambda\right]\frac{r\,\mathrm{d}r}{(r^2-x^2)^{1/2}}.\tag{9}$$

Equation (9) is an Abel inversion. The conjugate terms are

$$\int_{\Delta\lambda} h(x,\lambda,t)\,\mathrm{d}\lambda \quad\text{and}\quad \int_{\Delta\lambda} k(r,\lambda,t)\,\mathrm{d}\lambda.$$

It follows that

$$\int_{\Delta\lambda} k(r,\lambda,t)\,\mathrm{d}\lambda=-\frac{\Delta\lambda}{\pi}\int_r^R\frac{\mathrm{d}}{\mathrm{d}x}\left[1-\tau(x,\Delta\lambda,t)\frac{\mathrm{d}x}{(x^2-r^2)^{1/2}}\right].\tag{10}$$

From the value of $\tau(x, \Delta\lambda, t)$, we obtain $\int_{\Delta\lambda} k(r, \lambda, t)$ and thus n_m using the relation (1).

Two simplifying assumptions are necessary for this calculation.

(a) The first assumption involves disregarding, in eqn (1), the term $(g_m/g_n)\times(n_n/n_m)$ in the induced-emission term. In the worst possible case, this procedure leads to an error of slightly more than 10%. During the decay, or at a point out of LTE, this term is negligible because of overpopulation of the metastable level n_m with respect to the radiating level n_n.

(b) In order to carry out the Abel inversion for the $\tau(x, \Delta\lambda, t)$ term, we must be able to expand it in a series and retain the first-order term. This approximation is justified as long as the absorption is not greater than 20%.

2.2 *Correlations between the* 4s *states*

It may be shown that the 4s states are closely correlated and that it is not necessary to study variations in each of these. In fact, they are always in equilibrium and show the same departure from LTE. The variation of the number density of one then gives that of the others. This has been shown by calculating the excitation rate by electronic collisions between 4s states using the Gryzinski formulae.[4] We then compare these values with those corresponding to transitions between 4s levels and the most closely correlated (4p) levels. A difference of several orders of magnitude is found. What is more, the transition rates between the 4s states are very high. Some results are shown in Tables 1 and 2.

Table 1. Excitation rates by electronic collisions for some transitions between 4s levels

Transition / Temperature	$1s_5-1s_4$	$1s_4-1s_3$	$1s_3-1s_2$	$1s_5-1s_2$
12,000	1.45.E-4	8.01.E-5	9.31.E-5	1.07.E-5
10,000	1.33.E-4	7.24.E-5	8.30.E-5	8.98.E-6
8000	1.87.E-4	6.29.E-5	7.08.E-5	6.99.E-6
6000	9.87.E-5	5.09.E-5	5.37.E-5	4.81.E-6

Table 2. Excitation rates by electronic collisions between the 4s and 4p states

Transition / Temperature	$1s_2-2p_{10}$	$1s_2-2p_1$	$1s_3-2p_{10}$	$1s_3-2p_1$
12,000	1.58.E-10	1.21.E-9	1.43.E-9	2.97.E-8
10,000	1.04.E-10	7.19.E-10	9.26.E-10	1.94.E-8
8000	5.93.E-11	3.46.E-10	5.10.E-10	1.04.E-8
6000	2.54.E-11	1.12.E-10	2.08.E-10	3.79.E-9

Since the 4s levels are closely correlated by electronic collisions, a departure from LTE for one indicates the same departure for the other three. Collisions between metastable atoms and between neutral and metastable atoms strengthen these correlations.

In this study, we consider the $1s_2$ and $1s_4$ resonance levels in the same manner as the metastable levels. Resonance levels are optically bound to the ground state but, because of the strong dipole interaction between excited atoms in a resonant level and atoms in the ground state, the resonance radiation is immediately reabsorbed. We have calculated the excape factor, which is of the order of 0.1%.

2.3 *Choice of level*

We have studied the transmission coefficient $\tau(x, \Delta\lambda)$ of the discharge for eight lines in the steady state. We chose to use the 6965 and 7067 Å lines, corresponding to the $2p_2-1s_5$ and $2p_3-1s_5$ transitions, respectively. The measurements of their absorption coefficients $k(r, \Delta\lambda, t)$ allows us to calculate the number density of the $1s_5$ metastable level.

These lines were chosen for the following reasons. The reproducibility of the transmission-coefficient measurements is good. The value of $\tau(x_0, \Delta\lambda)$ on the arc axis were confirmed by another method. The profile $\tau(x_0, \lambda)$ of the *monochromatic transmission-coefficient* was plotted point by point for each line with a 0.04 Å wavelength step. The integration over the entire line profile gave (for the 6965 Å line) transmissions of 92.7, 92.5 and 92.8% in three successive measurements. The experimental value is between 92 and 93%. The total absorption on the axis is thus about 7.3% when $I = 54$ Å and $T = 11{,}500$°K. Under the same conditions, the total absorption on the axis for the 7067 Å line is about 6.7%. It is necessary to study a line with sufficient absorption to be measurable without too much error. But the weak-absorption approximation that is necessary for the calculation should still hold. The absorption coefficients for the 6965 and 7067 Å lines meet these criteria and have similar values.

We were able to use the 7514 Å ($2p_5 - 1s_4$) and 7273 Å ($2p_2 - 1s_4$) lines in the study of the $1s_4$ level. The absorption coefficient of the first has an acceptable value but that of the second is too weak for accurate measurements. The investigation of the $1s_4$ level was therefore abandoned.

The only usable line for measurement of the $1s_3$ level corresponds to the $2p_3 - 1s_3$ transition (7948 Å). Its absorption coefficient is too high. The $1s_2$ level can only be measured from the 7504 Å line ($2p_1 - 1s_2$). Consequently, the 4s levels were studied using the variations of the $1s_5$ level.

2.4 *Experimental method*

In order to explore completely a whole discharge radius, it is necessary to measure $\tau(x, \Delta\lambda, t)$ for $0 < x < R$ (Fig. 2). We must first record $\tau(x_1, \Delta\lambda, t)I_0 + I_{arc}(x_1, \Delta\lambda, t)$ during the decay. Then, keeping the experimental conditions constant, I_{arc} $(x_1, \Delta\lambda, t)$ is measured. This operation is repeated for several values of x between 0 and R. The number density of metastable atoms may then be calculated.

3. EXPERIMENTAL ARRANGEMENT

The study was carried out using a wall-stabilized arc[5] with graphite electrodes and burning in argon at atmospheric pressure. The arc current is about 55 amps. The beam I_0 is produced by a high-pressure xenon lamp (900 W). An optical set-up is adjusted to send a parallel beam through the arc. The light emitted by a chord is focused on the entrance slit of the monochromator. The spectroscope has a linear dispersion of 8.3 Å mm^{-1}. The arc interruption is obtained by a fast thyristor connected to the electrodes, which short-circuits the discharge. The turn-on time is close to 1 μs. The current is shunted in 17 μs (Fig. 3).

The signal from the photomultiplier is sent to a transient recorder. The signal is stored and subsequently displayed on an $X - Y$ recorder (or on an oscilloscope). The curves obtained are computer-analyzed.

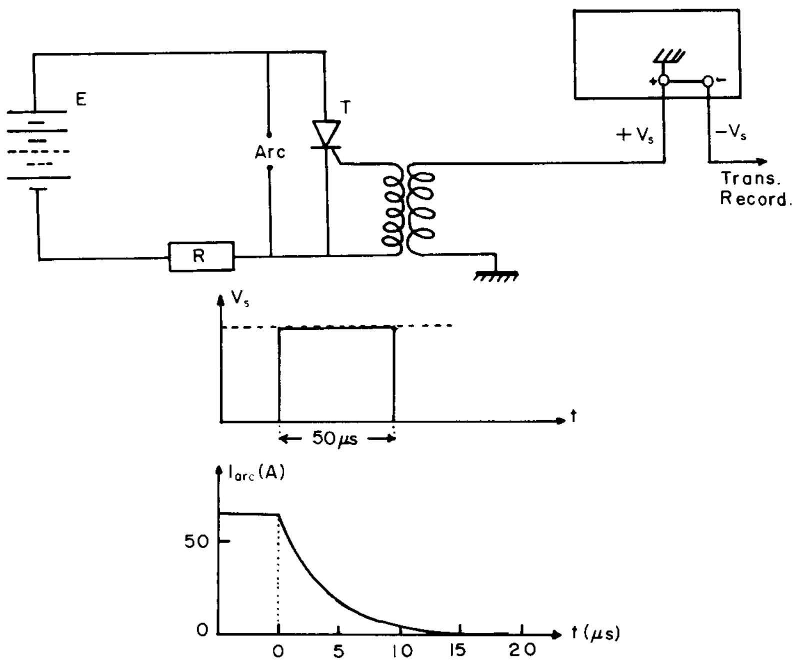

Fig. 3. Circuit breaker.

4. EXPERIMENTAL RESULTS

4.1 *Measurement of the electron density during decay*

In order to define a standard for the existence of LTE, it is necessary to measure a reference variable that is independent of the factors governing the existence of equilibrium. This statement obviously does not apply for temperature. We chose to measure the variations of electron density $n_e(t)$. Knowing the laws governing plasma composition in complete LTE, we relate one metastable atom number-density value with an n_e-value. This value n_m, corresponding to n_e in a complete LTE plasma, is called a theoretical value and may then be compared with the experimental value. When complete LTE is established, the values are the same. The $n_e(t)$ is determined from the absolute intensity of the free-bound continuum at $\lambda = 4230$ Å. Calibration is carried out by using a tungsten ribbon lamp. We have

$$\epsilon_\nu^{f.b.} = 5.44 \times 10^{-39}\, n_e \cdot n_i T^{1/2} \xi (\mathrm{erg} \cdot \mathrm{cm}^{-2} \cdot \mathrm{st}^{-1}),$$

with $\xi^{(6)} \simeq 1.6$, $n_e \simeq n_i$, n_i being the ionic density and T the temperature. This method is not very sensitive to the departure from LTE conditions; thus, n_e is proportional to $T^{1/4}$.

Figure 4 represents the variations of n_e with t for three values of r. Figure 5 represents the variations of n_e with r for three values of time t. It may be seen that the variation is quasi-exponential as a function of time and that the decrease in n_e is slow (which is characteristic of an argon plasma). The values obtained allow us to calculate $n'_m(t)$, which may be compared with experimentally determined values.

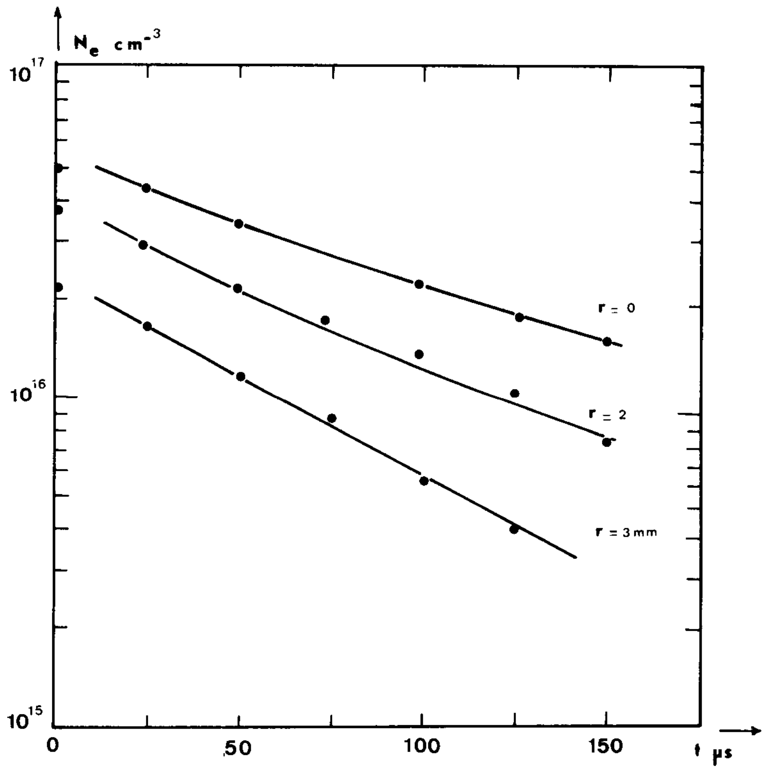

Fig. 4. Variations of n_e with time for 3 values of r.

4.2 *Metastable atom-density decay*

Figure 6 represents the comparison between experimental and calculated values, as functions of time, of the density of the $1s_5$ state for points at $r = 2$ and $r = 2$ mm. Figure 7 represents the comparison between experimental and calculated density values of the $1s_5$ level as functions of the radius for $t = 0$ and $t = 100\ \mu$s. It may be seen that, on the axis, the decay is exponential as a function of time and there is no difference between the experimental and calculated values on the discharge axis during the decay period. At the steady state ($t = 0$), the $n_m(r)$ and $n'_m(r)$ curves are identical. However, when $r > 2.5$ mm and $t > 100\ \mu$s, the curves differ. At 3.5 mm from the axis,

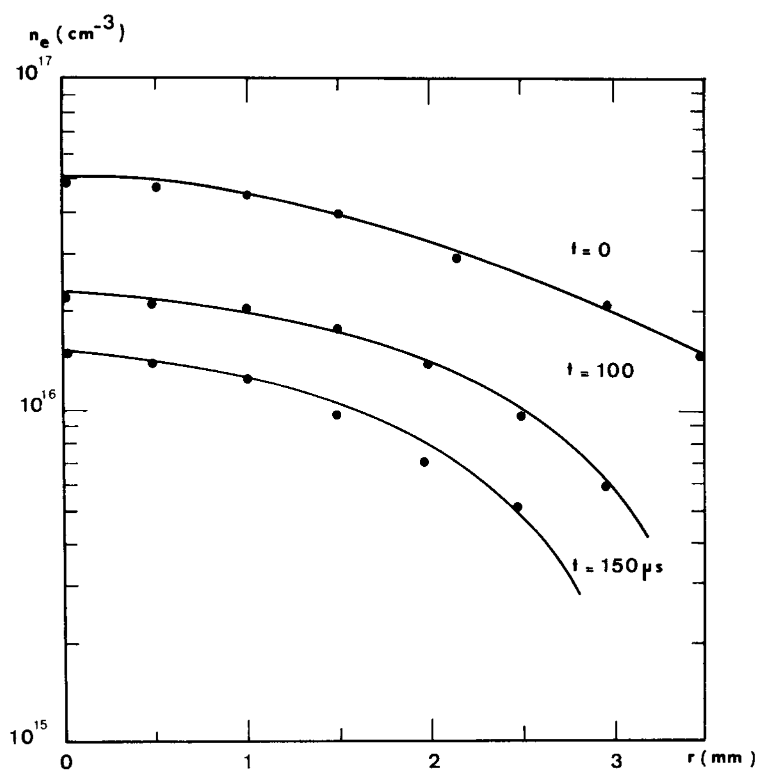

Fig. 5. Variations of n_e with r for $t = 0$, $t = 100$, and $t = 150\ \mu$s.

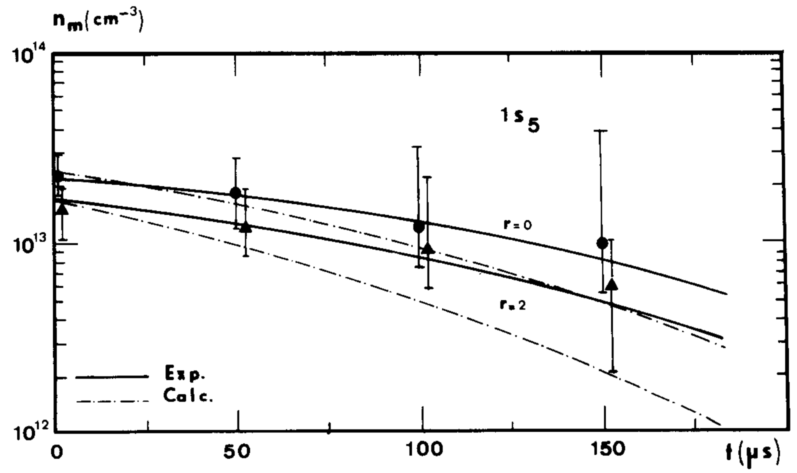

Fig. 6. Comparison between experimental and calculated values of the $1s_5$ level vs time for $r = 0$ and $r = 2$ mm.

the experimental value is greater by about an order of magnitude than the calculated value. This shows a departure from LTE and an overpopulation of the 4s level. Measurements carried out on the $1s_4$ (7514 Å line) and the $1s_2$ (7504 Å line) states confirm this observation.

4.3 *Validity of the data*

Each measurement was repeated six or seven times. This allowed us to find the scatter of the experimental results. The least scatter was found for the steady state: for example, on the axis ($r = 0$) and for $t = 0$, the $1s_5$ level number density was always found to be between 1.91×10^{13} cm^{-3} and 2.45×10^{13} cm^{-3}. On the other hand, for $t = 0$ and $r = 3$ mm, the number density of the same level was between 6×10^{12} cm^{-3} and 1.6×10^{13} cm^{-3}.

The measurements carried out during the decay lead to a much larger dispersion. On the axis, the absorption is high and the divergences between the values obtained are not very large. At $t = 100\ \mu$s, they are between 7.5×10^{12} and 2.1×10^{13} cm^{-3}. However, at $r = 3$ mm and $t = 100\ \mu$s,

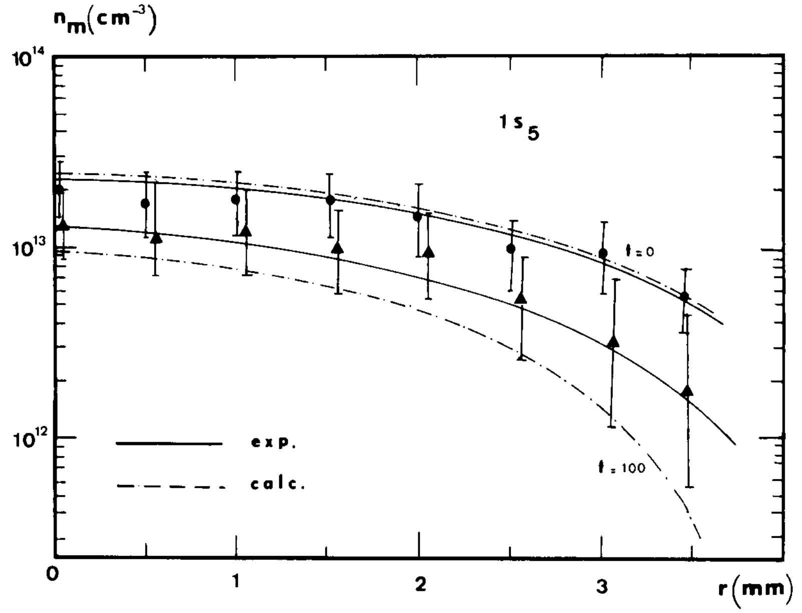

Fig. 7. Comparison between experimental and calculated values of the $1s_5$ level vs r for $t = 0$ and $t = 100\ \mu s$.

the values are between 1.3×10^{12} and $8 \times 10^{12}\ cm^{-3}$. This large divergence is due to the low absorption coefficient $k(r, \Delta\lambda, t)$ for $r \geq 3$ mm.

The error introduced by the calculation is much less important. The absorption coefficient of the 6965 and 7067 Å lines being low, expansion into a series in $\tau(x, \lambda)$ limited to the first order is justified. The Abel inversion is carried out by computer on a conveniently "smoothed" profile. A parabolic interpolation method is used. The maximum error due to the calculations is estimated to be about 25%, i.e. negligible when compared with the experimental errors. The error of the calculated curves is not greater than 15%. Under these conditions, the departure from LTE observed when $r > 3$ mm and $t > 100\ \mu s$ cannot be due to an uncertainty in the measurements.

Measurements actually carried out on the excited atom densities, as well as on electron densities, confirm these results.[7]

5. COMPARISON WITH A RADIATING LEVEL

The deexcitation of metastable atoms can only come about on collision with other particles. It is interesting to measure their rates of decay compared with those of a radiating level. We have compared the density variations of the $1s_5(3p^54s)$ level with that of the radiating $3p_8(3p^55p)$ level, the density of which is measured by using the absolute intensity of the 4300 Å line.

Figure 8 shows the density of the $1s_5$ metastable level along the abscissa and the density of the

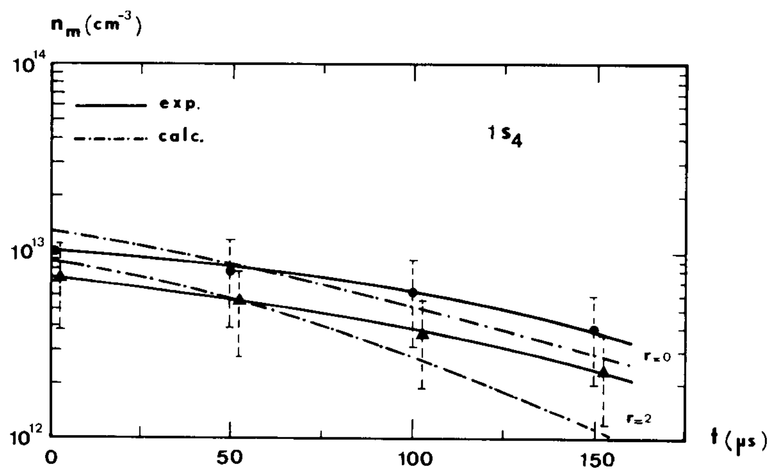

Fig. 8. Relationship between the number density of the $3p_8$ radiating level and of the $1s_5$ metastable level for the decaying plasma.

$3p_8$ radiating level along the ordinate. The solid line represents the theoretical relationship between the densities of these two levels at complete LTE. The experimental points represent the decay of density values obtained at $r = 0$ and $r = 2$ mm. Complete LTE is achieved when the points are on the curve. The other points indicate that the metastable level is relatively more populated than the radiating level.

6. DISCUSSION

In the steady state, LTE depends on the presence of an electron density greater than (or equal to) a critical density n_e^c. When $n_e < n_e^c$, the collision frequency between electrons and atoms is not sufficient to give $T_a = T_e$, T_a and T_e being the atom and electron temperatures, respectively. This critical density was measured for hydrogen and helium by Griem[8] and Drawin.[9] Its extension to argon is, however, not easy.

Figure 9 shows the results of recent studies on the departure from LTE in a steady-state argon arc at atmospheric pressure.[10] The metastable atom number density is plotted along the abscissa

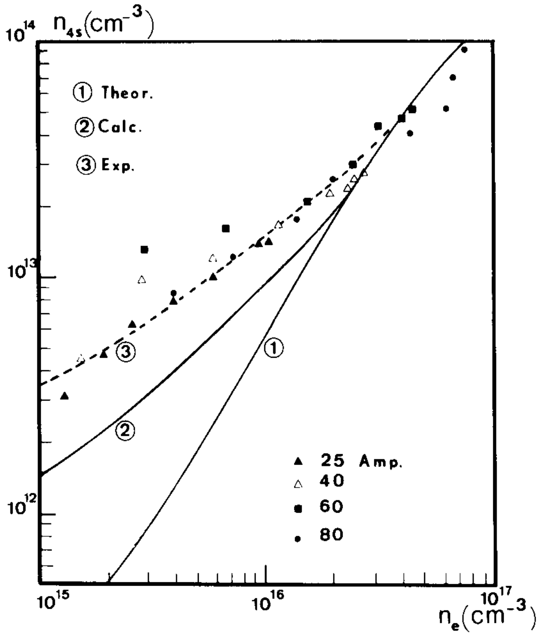

Fig. 9. Theoretical (1), calculated (2) and experimental (3) relations between electron number densities and metastable level number densities in a steady argon discharge.

and the electron number density along the ordinate. Three curves are plotted. Curve 1 gives the theoretical relationship at complete LTE between the number density of a metastable level and the electron number density. Curve 2 gives the values of the metastable number densities calculated from a B.K.M. type model[11] taking into account all possible interactions between different groups of levels. Curve 3 is an experimental curve. For each electron number density (measured by the recombination continuum), we have plotted the corresponding value of the metastable number density (measured by absorption). The agreement between calculated and experimental data is not very good because of uncertainties of some of the reaction rates. However, for $n_e > 2 \times 10^{16}$ cm^{-3}, the curves are the same and complete LTE is established. This result confirms the existence of a critical density above which, in a transient plasma, the discharge is in equilibrium. Our results show that an identical analysis can be carried out for a transient state. The arc discharge at atmospheric pressure does not decay in accord with the laws of complete LTE, which is contrary to a hypothesis that is often made.[12] As for the steady state, a critical number density exists. For $n_e < n_e^c$, collision processes are not sufficiently fast compared with radiative processes to maintain LTE during extinction. We also note that the radiating levels are more rapidly depopulated than the metastable level.

The accuracy of the data obtained is, in any case, insufficient to give the n_e^c value under transient conditions. In order to determine n_e^c, we measured the electron number density by two methods. The first, using the recombination continuum (free–bound), is practically independent of LTE. The second involves the absolute intensity of the 4300 Å line. From Boltzmann's and Saha's formulae, one can calculate n_e. This method is only valid when LTE is established. The curves thus obtained diverge from the point at which $n_e \simeq 2\times10^{16}\,\mathrm{cm}^{-3}$. Below this value, complete LTE is not established. These results confirm those obtained at the steady state. They are important in studies on ion-electron recombination. We can define an apparent collisional-radiative coefficient γ by

$$dn_e/dt = -\gamma n_e n_i.$$

But γ is a composite coefficient because both recombination and ionization occur in a decaying plasma. We have

$$dn_e/dt = -\alpha n_e n_i + \beta n_0 n_e;$$

α is the true recombination coefficient, β is the ionization coefficient and n_0 is the atom number density in the ground state. The magnitude of γ depends on β, which is closely related to the thermal conductivity coefficient. In fact, the ionization term depends on the rate of energy diffusion in the discharge, which is strongly dependent on the presence of overpopulation of metastable atoms.

The preceding results show that the term denoting the loss of metastable atoms was smaller than it should have been in LTE. This result also shows that a source term has greater importance during decay than if the discharge were in complete LTE at all times.

Processes leading to the creation or destruction of metastable atoms.[13] Destruction by collisions either leads to excitation of a higher level or to deexcitation during a superelastic collision. The latter process, which is particularly important with slow electrons, leads to very large energy transfers in the discharge.

Only the electrons in the tail-end of the distribution function have a high enough energy to excite neutral atoms. The energy of the electrons decreases so fast during the extinction period that this process rapidly becomes negligible. The metastable atoms are also created by deexcitation of higher levels (for example, $3p^54p-3p^54s$). The relative importance of this process increases as the energy of the electrons decreases. A third reaction mechanism involves the recombination either by direct creation or by "cascade" after the formation of an excited neutral atom. This process is the same as the preceding one.

The departure from LTE leading to an overpopulation of metastable atoms compared to the electron number density shows that the formation processes are more important than expected. The calculation carried out using the B.K.M. model showed that the process of population by cascade was more important than the others and gave rise to an overpopulation of the $4s$ level.[10] This result confirms the importance of the radiating processes during the post-discharge period for an n_e value lower than the threshold value.

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