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A collisional–radiative model applicable to argon discharges over a wide range of conditions. II: Application to low-pressure, hollow-cathode arc and low-pressure glow discharges

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Abstract. The extensive collisional–radiative model for an argon atom plasma is applied to a low-pressure, hollow-cathode arc discharge and to the positive column of a low-pressure glow discharge in order to clarify the mechanisms by which the excited levels in these discharges are populated, the results being compared with experimental investigations in the literature. Computations are carried out for various sets of input parameters, such as the electron kinetic temperature T_e, the atom temperature T_a, the ion temperature T_i, the electron number density n_e, the ground state atom population n_1, the plasma column radius R and the escape factors Λ_{mn} and Λ_m, characterising the non-equilibrium plasmas under consideration. The predictions of our model, i.e. the populations in the excited levels as a function of the electron number density, the effective principal quantum number and the discharge current, are compared with the experimental results and in two cases also with the theoretical results of other authors. It is shown that all calculated dependences are fairly close to the corresponding experimental curves referring to both discharges. The results presented confirm the applicability of the so-called 'analytical top model' of van der Mullen *et al* and Walsh's formula for Λ_{1n} interpreted according to Mills and Hieftje.

1. Introduction

In the preceding paper (Vlček 1989), a collisional–radiative (CR) model applicable over a wider range of conditions than those described in the literature (Giannaris and Incropera 1973, Katsonis 1976, Gomés 1983, van der Sijde *et al* 1984a, Hasegawa and Haraguchi 1985) was established for an argon atom plasma.

Atom–atom inelastic collisions and diffusion losses of the metastable states, together with the electron–atom inelastic collisions and radiative processes usually included, are considered in this model, taking into account 65 effective levels.

Analytical expressions used for the corresponding cross sections are in good agreement with currently available experimental and theoretical data for argon. In particular, the measurements of Chutjian and Cartwright (1981) and the computations of Kimura *et al* (1985) have been used extensively.

With the help of the previously established numerical method (Vlček 1989), we can calculate the population coefficients determining the populations in all excited effective levels. This enables us to study the mechanisms by which these levels are populated under various conditions in a non-equilibrium argon plasma characterised (even in the case of a non-Maxwellian electron energy distribution function) (EEDF) by the set of parameters T_e, T_a, T_i, n_e, n_1, R, Λ_{mn} and Λ_m.

The main aim of the present paper is to check the reliability of our extensive CR model under the conditions in the low-pressure hollow cathode arc, studied experimentally by van der Mullen *et al* (1978, 1980) and by van der Sijde *et al* (1984a, b), and in the positive column of the low-pressure glow discharge,

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investigated experimentally by Kagan *et al* (1963b). A further motivation for this study is our interest in understanding the mechanisms by which the excited levels are populated in these two argon discharges of practical interest.

For experimental verification of our CR model, a combination of these discharges seems to be suitable because the population mechanisms differ appreciably in them. Moreover, because the electron energy distribution function in the hollow-cathode arc investigated is Maxwellian (Pots 1979) and the radiation trapping is negligibly small (van der Sijde *et al* 1984a, b), the calculated excited level populations obtained under these conditions cannot be affected by possible inaccuracies arising from the solution of the Boltzmann equation and from determination of the optical escape factors.

2. Results and discussion

Assuming that the quasi-stationary state model can be applied (see for example Cacciatore *et al* 1976, Biberman *et al* 1982) and the fundamental mode of diffusion of the metastables to the wall is dominant in the discharge tube (see for example Delcroix *et al* 1976, Ferreira *et al* 1985), we obtain a set of coupled linear equations

$$\sum_{n=2}^{65} a_{mn} n_n = -\delta_m - a_{m1} n_1 \qquad (1)$$

where $m = 2, \ldots, 65$, from which the unknown excited level populations n_n may be calculated, provided that the coefficients a_{mn} and δ_m are known (Vlček 1989) and the ground state atom population n_1 has been determined experimentally.

Owing to the possibility of investigating the effect of the upward ionisation flow of electrons from the ground state atom and their downward recombination flow from a continuum on the populating of the excited levels, the system (1) is solved, in spite of the fact that n_1 is not an independent parameter in our case, in the standard form

$$n_n = n_n^{(0)} + G_n^{(1)} n_1 \qquad \text{for } n = 2, \ldots, 65 \qquad (2)$$

where the population coefficients $n_n^{(0)}$ and $G_n^{(1)}$ represent the solutions of (1) with $n_1 = 0$ or $n_1 = 1$ and $\delta_m = 0$, respectively inserted into their right hand sides.

The expression (2) can be rewritten as

$$n_n = r_n^{(0)} n_n^{S} + r_n^{(1)} n_n^{B} \qquad (3)$$

where n_n^S and n_n^B are the corresponding Saha population and Boltzmann population, respectively, $r_n^{(0)}$ and $r_n^{(1)}$ are the so-called CR coefficients relating the actual populations n_n to n_n^S and n_n^B, respectively.

In a general case, the numerical method developed allows us to calculate the population coefficients $n_n^{(0)}$ and $G_n^{(1)}$ as functions of the following input parameters: T_e, T_a, T_i, n_e, n_1, R, Λ_{mn} and Λ_m with the possibility of neglecting the atom–atom inelastic collisions when their influence on the population mechanism is studied.

Under a reasonable assumption that only the reabsorption of the resonance radiation may be important in the discharges investigated, we have used the analytical formulae for the escape factors (Mills and Hieftje 1984) in which Λ_{1n} are dependent only on T_a, n_1 and R, where $n = 3, 5, 12, 15, 16, 17, 20, 21, 26, 27$ and 33 (see table 1 of Vlček 1989).

In the special case, when the Boltzmann equation need not be solved due to the Maxwellian form of the EEDF in a plasma, computations become straightforward and relatively rapid. Furthermore, the ion temperature T_i does not then appear among the input parameters (Vlček and Pelikán 1985, Vlček 1989).

The basic data characterising the excited levels considered in our CR model, including all individual 4s and 4p states, are given in table 1 to help in comparing the calculated and experimental results.

Table 1. Data characterising the excited effective levels including all actual 4s and 4p states.

Level number n	Designation $n_{pq}nl[K]_J$	Excitation energy ε_{1n} (eV)	Statistical weight g_n
2	$4s[3/2]_2$	11.548	5
3	$4s[3/2]_1$	11.624	3
4	$4s'[1/2]_0$	11.723	1
5	$4s'[1/2]_1$	11.828	3
6	$4p[1/2]_1$	12.907	3
7	$4p[3/2]_{1,2}$, $[5/2]_{2,3}$	13.116	20
8	$4p'[3/2]_{1,2}$	13.295	8
9	$4p'[1/2]_1$	13.328	3
10	$4p[1/2]_0$	13.273	1
11	$4p'[1/2]_0$	13.480	1

2.1. Hollow-cathode arc discharge

Hollow-cathode arc discharges have been widely used, for example in the investigation of argon ion laser plasmas (Pots *et al* 1978), in fusion-oriented and welding technology (Chall and Uhlenbusch 1982), in plasma centrifuges (Wijnakker *et al* 1979), or generally in plasmas in magnetic fields (Boeschoten *et al* 1979).

Valuable results contributing to the elucidation of the mechanisms by which the excited levels in a low-pressure, hollow-cathode arc are populated have been obtained in recent years by van der Mullen *et al* (1978, 1980, 1983) and van der Sijde *et al* (1984a, b).

In our case, the numerical results for the excited level populations n_n and for a CR coefficient $r_n^{(1)}$ are respectively compared with the corresponding values determined experimentally by van der Mullen *et al* (1978, 1980) and van der Sijde *et al* (1984a, b) who measured the absolute intensities of numerous Ar I lines as a function of the electron number density n_e for known values of T_e, T_a, n_1 and R in the highly ionised, magnetically confined plasma of a low-pressure, hollow-cathode arc.

It has previously been verified that the EEDF is

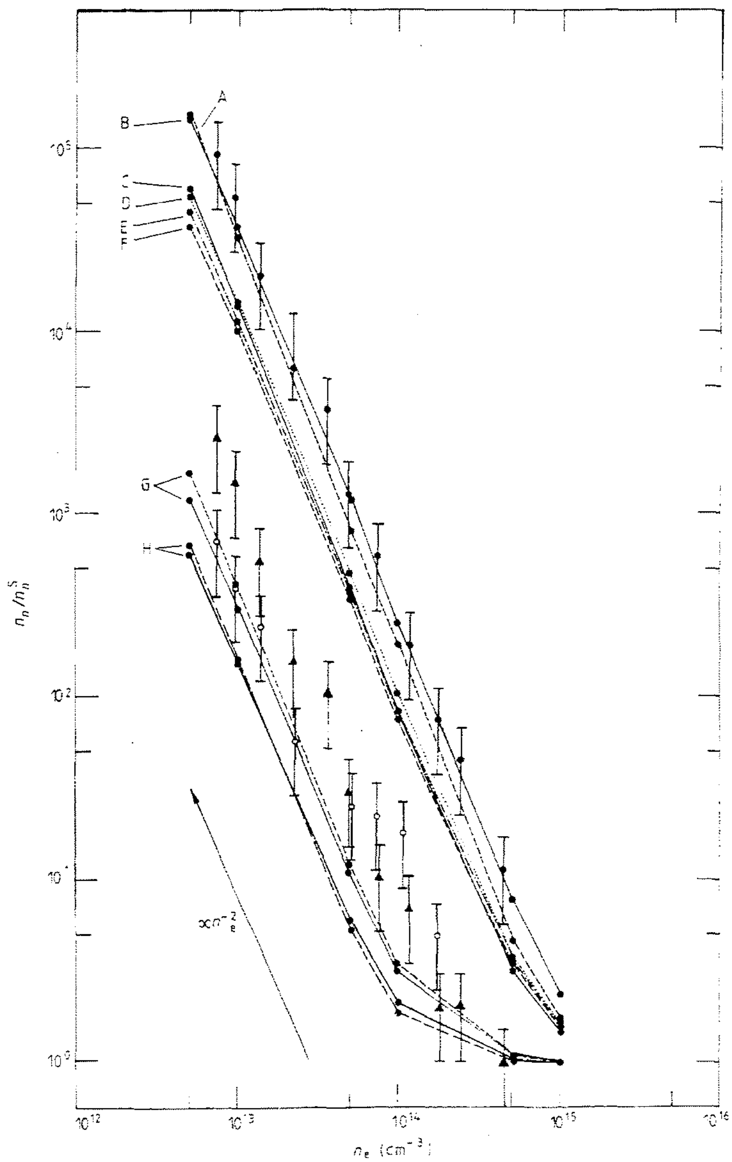

Figure 1. The excited level populations n_n related to the corresponding Saha values n_n^S for various electron number densities n_e at $T_e = 40600$ K, $T_a = 11600$ K, $R = 1$ cm and n_1 ranging from 2.35×10^{13} cm^{-3} for $n_e = 5 \times 10^{12}$ cm^{-3} to 3.62×10^{12} cm^{-3} for $n_e = 10^{15}$ cm^{-3}. Present work: curve A, $n = 6$; curve B, $n = 11$; curve C, $n = 7$; curve D, $n = 10$; curve E, $n = 8$; curve F, $n = 9$ (see table 1); curves G and H, the 5d and 6d groups, respectively, with $j_c = \frac{1}{2}$ (full curves) and $j_c = \frac{3}{2}$ (broken curves). Symbols ●, ▲, ○ show the experimental data of van der Mullen *et al* (1980) for the whole 4p, 5d and 6d groups, respectively.

Maxwellian (Pots 1979) and that doubly ionised ions can be neglected (van der Mullen *et al* 1980, Pots 1979) under the conditions considered.

In figure 1 we compare the ratios n_n/n_n^S measured by van der Mullen *et al* (1980) for the 4p, 5d and 6d groups with the corresponding values calculated by us at the plasma parameters T_e, n_e and n_1 presented in the above-mentioned paper and for $T_a = 11600$ K and $R = 1$ cm taken from similar experiments (van der Sijde *et al* 1984a) carried out with the help of the same set-up. According to van der Mullen *et al* (1978), the accuracy in the measurement of n_n is estimated to be 50%.

As can be seen in figure 1, the uniform decrease of the calculated values of n_n/n_n^S as n_e^{-2} in the so-called 'excitation saturation phase' agrees well with the measured dependences, but the theoretical values,

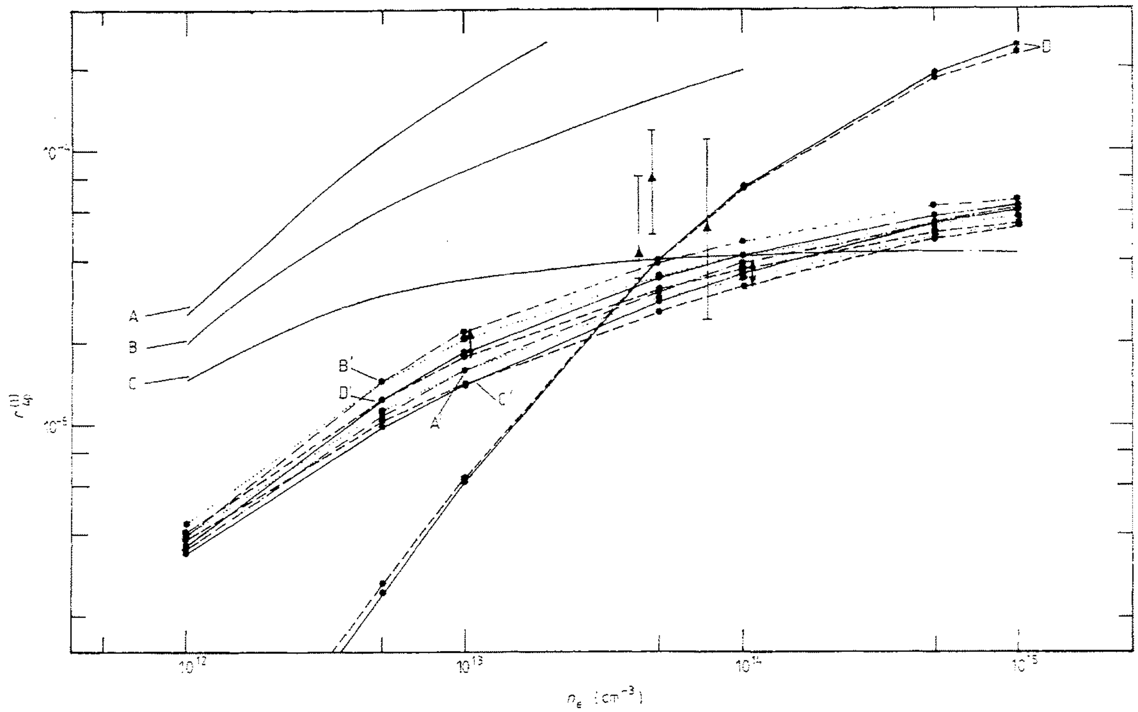

Figure 2. The $r_{4p}^{(1)}$ coefficient as a function of the electron number density n_e. Theoretical results: curve A, Pots (1979); curve B, van der Mullen *et al* (1977); curve C, van der Sijde *et al* (1984a) (all with $T_e = 34800$ K); curves D, determined by us using the model of Katsonis (1976) at $T_e = 35000$ K (full curve) and $T_e = 40000$ K (broken curve); present work: curve A′, $n_1 = 3 \times 10^{12}$ cm^{-3}, $T_a = 3480$ K, $R = 1$ cm, $T_e = 34800$ K (chain curve) and $T_e = 46400$ K (dotted curve); B′, as for A′ but at $n_1 = 9 \times 10^{12}$ cm^{-3}; C′, $n_1 = 3 \times 10^{12}$ cm^{-3}, $T_a = 6960$ K, $R = 1$ cm, $T_e = 34800$ K (full curve) and $T_e = 46400$ K (broken curve); D′, as for C′ but at $n_1 = 9 \times 10^{12}$ cm^{-3}. ▲, experimental results (van der Sijde *et al* 1984b).

except those obtained for the individual 4p states denoted by $n = 6$ and 11, are somewhat underestimated compared with the measurements. The values of n_e relating to the transition of the excited levels considered to the regime of the partial local thermodynamic equilibrium are in satisfactory agreement with those obtained by extrapolation of the measured data in the work of van der Mullen *et al* (1980).

It is known (see for example van der Sijde *et al* 1984b) that the values of the CR coefficients $r_n^{(1)}$, occurring in (3), may be important for spectroscopic diagnostics of a plasma, e.g. for the determination of the electron temperature T_e and the ground state atom population n_1, if the nth level is in the complete saturation phase.

In figure 2 we give our results for the coefficient $r_n^{(1)}$, where $n = 9$, as a function of n_e for comparison with the corresponding experimental values obtained by van der Sijde *et al* (1984b) under the following conditions in the discharge: $34800\ \mathrm{K} \leqslant T_e \leqslant 46400$ K, $3480\ \mathrm{K} \leqslant T_a \leqslant 6960$ K, $10^{13}\ \mathrm{cm}^{-3} \leqslant n_e \leqslant 10^{14}\ \mathrm{cm}^{-3}$, $3 \times 10^{12}\ \mathrm{cm}^{-3} \leqslant n_1 \leqslant 9 \times 10^{12}\ \mathrm{cm}^{-3}$. As van der Sijde *et al* (1984b) did not give the value for the plasma column radius in their work, we have used $R = 1$ cm again (van der Sijde *et al* 1984a) in our computations illustrating the dependence of the coefficient $r_n^{(1)}$ on the electron temperature T_e and also on the atom temperature T_a and the ground state atom population n_1. Assuming that the value of n_1 changes with n_e in a manner similar to the measurements of van der Mullen *et al* (1980), we have determined the most probable values of $r_n^{(1)}$ at $n_e = 10^{13}$ cm^{-3} and 10^{14} cm^{-3}. They are denoted by vertical arrows in figure 2. Our calculations have proved that the plasma investigated is completely optically thin also for all radiative transitions excepting the resonance transition from the $4s'[1/2]_1$ state at $n_1 = 9 \times 10^{12}$ cm^{-3} where we have obtained the value of $\Lambda_{15} = 0.46$ and 0.83 for $T_a = 3480$ K and 6960 K, respectively.

The numerical results obtained from several CR models under the assumption that the plasma is completely optically thin are also shown in figure 2.

When we used the extensive 65-level model of Katsonis (1976), in which all important excitations from the ground state of an atom to the levels lying above the 4s′ states are omitted, except the excitations to the effective levels with $n = 15$, 16 and 17 (Vlček 1989), the model curves for $r_n^{(1)}$ obtained at $T_e = 35000$ K and 40000 K are given. When the simplified models (van der Mullen *et al* 1977, Pots 1979) and the extensive 49-level model of van der Sijde *et al* (1984a) have been applied at $T_e = 34800$ K, the results for the 4p group as a whole are shown. Note that in the model of van der

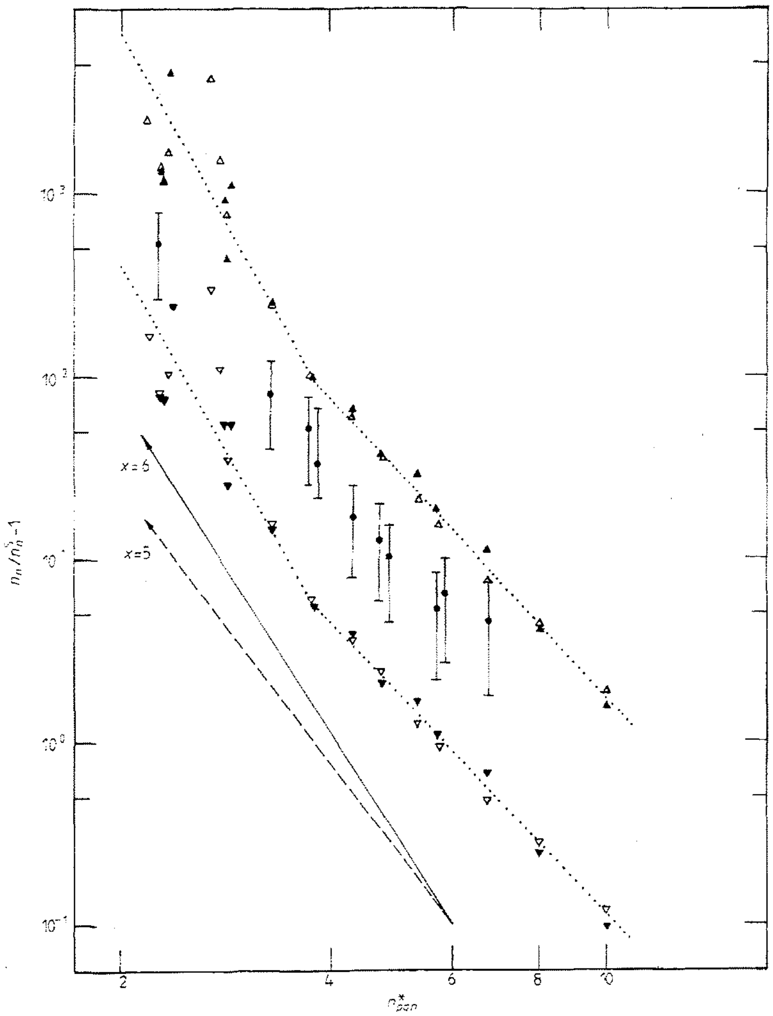

Figure 3. The population factor $n_n/n_n^S - 1$ as a function of the effective principal quantum number n^*_{pqn} obtained with the following input parameters: ▲, $j_c = \frac{1}{2}$ and △, $j_c = \frac{3}{2}$ for $T_e = 58\,000$ K, $T_a = 11\,600$ K, $n_e = 6.7 \times 10^{13}$ cm^{-3}, $n_1 = 10^{13}$ cm^{-3} and $R = 1$ cm; ▼, $j_c = \frac{1}{2}$ and ▽, $j_c = \frac{3}{2}$ for $T_e = 34\,800$ K, $T_a = 11\,600$ K, $n_e = 6.7 \times 10^{13}$ cm^{-3}, $n_1 = 10^{13}$ cm^{-3} and $R = 1$ cm. ●, experimental results (van der Mullen *et al* 1980). The full line $x = 6.0$ and the broken line $x = 5.0$ represent the slopes predicted on the basis of the analytical top model (van der Mullen *et al* 1983).

Sijde *et al* (1984a) the semiempirical formulae of Vriens and Smeets (1980) proposed for neutral hydrogen and alkali excited states are employed, all 4s states are separated and the statistical weights of the f groups are increased artificially.

Figure 3 shows our numerical results together with the corresponding experimental results (van der Mullen *et al* 1980) for the value of $n_n/n_n^S - 1$ as a function of the effective principal quantum number $n^*_{pqn} = (\varepsilon_1^H/\varepsilon_n)^{1/2}$, where ε_1^H and ε_n are the ionisation energies for atomic hydrogen in the ground state and for the nth level of argon respectively.

As can be seen, the measured values obtained at actual electron temperatures in the range from 34800–58000 K and at $n_e = 6.7 \times 10^{13}$ cm^{-3} are found to lie between the model curves determined by us at these two extreme values of T_e. Moreover, the slopes of the straight parts of our dependences agree well not only with the experimental result but also with that obtained on the basis of the so-called analytical top model (van der Mullen *et al* 1983). Using this model to describe the population mechanisms of high-lying excited levels in a real, collisionally dominated, ionising plasma with the Maxwellian EEDF, one can write the following simple power law for non-hydrogenic systems:

$$n_n/n_n^S - 1 = b_0(n^*_{pqn})^{-x}$$

where b_0 is a constant and the value of x is in the range

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