Experimental evidence for the complete saturation phase in the argon neutral system

EXPERIMENTAL EVIDENCE FOR THE COMPLETE SATURATION PHASE IN THE ARGON NEUTRAL SYSTEM

J.J.A.M. van der MULLEN, B. van der SIJDE and D.C. SCHRAM
Physics Department, Eindhoven University of Technology, Eindhoven, The Netherlands

Received 3 January 1980
Revised manuscript received 18 June 1980

An experimental study of the population densities of excited states confirms the existence of the complete excitation saturation phase in the argon neutral system. Collisional radiative coefficients $r_m^{(1)}$ are independent of n_e and decrease with decreasing ionization energy. At higher n_e-values the levels are observed to come consecutively into Saha equilibrium (PLTE).

In the literature, the problem of the population of excited states in radiative plasmas has been treated within the framework of Collisional Radiative (CR) models. In particular, these studies have been focussed on hydrogen [1–3], or hydrogenic ions [4,5], as here the atomic system is relatively simple and expressions are available for electron excitation and ionization rates. For diagnostic applications also CR-models for more complex systems are of importance, which, in view of the complexity of the systems, should be supported by reliable measured data.

We have performed absolute measurements of the densities of excited argon atoms in the electron density range from $n_e = 8 \times 10^{18}\ \mathrm{m}^{-3}$ to $4 \times 10^{20}\ \mathrm{m}^{-3}$. This n_e-range is intermediate between the Corona and PLTE-regimes. Fujimoto et al. [2,3] showed from CR-model calculations for hydrogen, that in this intermediate density regime also a relatively simple systematic trend exists, which was called the complete saturation phase. We will show from measurements that this systematic behaviour exists also for Ar (neutral) I, in spite of the larger complexity of the atomic system, and in this regard this work forms an extension of the work of Tachibana and Fukuda [6].

In CR-models the density of the mth excited state is described by an equation of the form:

$$n_m = r_m^{(0)} n_m^{\mathrm{S}} + r_m^{(1)} n_m^{\mathrm{B}} , \qquad (1)$$

in which n_m^{S} is the level density according to the Saha equation and n_m^{B} the density according to the Boltzmann equation. The coefficients $r_m^{(1)}$ and $r_m^{(0)}$ are the collisional radiative (CR)-coefficients, characteristic for the atomic system. The first term of the r.h.s. of eq. (1) relates n_m to the density of the continuum of the considered system (n_{Ar^+}), the second term to the density of the ground state atoms, n_{Ar}. For some atomic systems (H[1], He^+ [5], He^0 [5,7], Ar [8]) model calculations have been performed and CR-coefficients have been tabulated.

Following previous work by Drawin [1], Webb [9], Romanov [7] and in particular Fujimoto [2,3], the following regimes or phases can be distinguished with increasing n_e:

(1) *The Corona phase*, in which electron excitation is balanced by radiation; only the second term of the r.h.s. of eq. (1) is important and $r_m^{(1)}$ is proportional to n_e.

(2) *The complete (excitation) saturation phase*, in which electron excitation is balanced by electron deexcitation; in this phase $r_m^{(1)}$ "saturates", i.e. is independent of n_e and only weakly dependent on T_e.

The transition between (1) and (2) is relatively slow and is called the quasi-saturation phase [3]; here, full CR-models are required.

(3) *Partial LTE*; here recombination is significant and the level m is in equilibrium with the continuum. Now the first term of the r.h.s. of eq. (1) is dominant and $r_m^{(0)}$ approaches 1. If $n_{\mathrm{Ar}^+} = n_e$, it follows that $n_m \propto n_e^2$.

The complete excitation saturation phase is due to the dominance of collisional (de)excitation over radiative processes, far before the considered levels come into Saha–Boltzmann equilibrium. For ionizing plasmas with overpopulated ground state densities, this results in an upward flow of excitation, the so called ladder-climbing process. For higher lying levels (lower ionization energy, E_m^{ion}) the energy differences between neighbouring levels decrease, and consequently, electron excitation cross sections will increase, as will the statistical weights of neighbouring levels.

It has been shown for hydrogen [3] that these systematics lead to a dependence of $n_m \propto (E_m^{\mathrm{ion}})^3$. These systematics suggest the possibility of significant simplifications with respect to the use of full CR-models. Experimentally, similar trends are also observed for more complex systems: He^+ [4], Na [10], Ar [6,11–13, this paper], Ar^+ [13].

For neutral argon, only one extensive model calculation exists [8], but it has been concluded from comparison with measurements [12,13], that in this model the electron (de)excitation is largely underestimated. Consequently, the CR-coefficients $r_m^{(1)}$ are too small and are not constant in the model. Absolute measurements as function of n_e do not exist; even relative measurements are scarce [6] and n_e and T_e are not accurately known. The transition between saturation and PLTE-phases has not been observed experimentally. The emphasis of our work is on absolute measurements of excited level densities, accompanied with accurate measurements of the plasma parameters.

We have measured the absolute intensities of a number of Ar I lines with upper levels in the 6s–10s, 4p–7p, 4d–8d groups. The system has been calibrated with a tungsten ribbon lamp; the transition probabilities were taken from Wiese [14]. The measurements were performed on a highly ionized (20–95%) magnetically confined plasma column of a hollow cathode arc. The plasma parameters n_e and T_e were in the range $8 \times 10^{18} < n_e < 4 \times 10^{20}$ m^{-3} and $3 < T_e < 5$ eV and were measured with Thomson scattering [15]. It has been verified that the electron energy distribution is maxwellian [13]. From intensity measurements of the Ar III, 328.6 nm line it can be concluded that the density of doubly ionized ions can be neglected with respect to the Ar^+-ion density, so that $n_e = n_{Ar^+}$ [13]. The ground state neutral density ranges from 2 to 0.5×10^{19} m^{-3} and was determined with a method

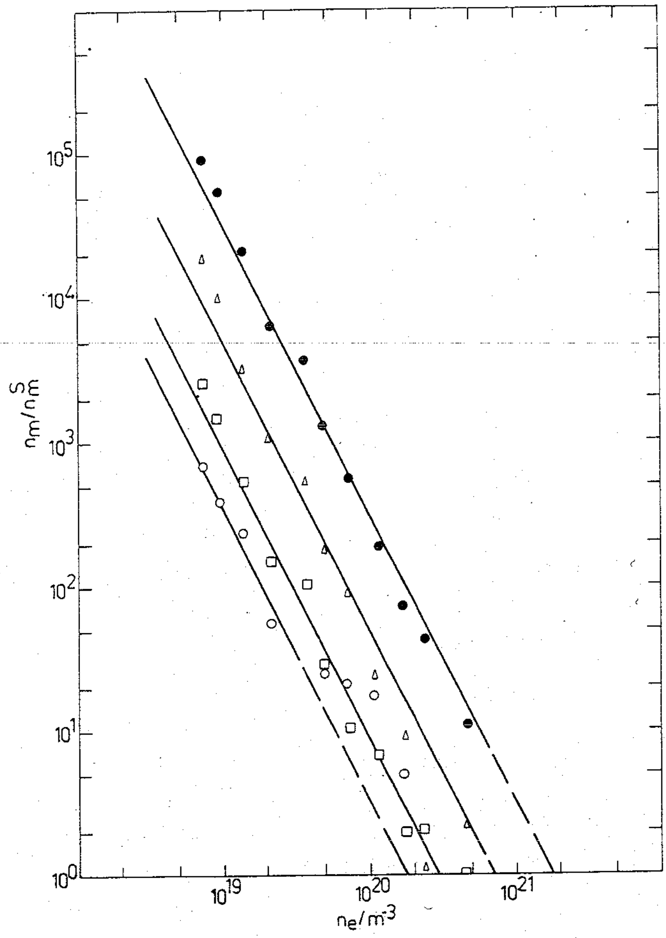

Fig. 1. The ratios n_m/n_m^S, excited group density normalized to its Saha-density, as functions of the electron density n_e for the 4p (•), 4d (△), 5d (□) and 6d (○) group. The solid lines are according to an n_e^{-2} slope. T_e = 3.5 eV and $n_a = 10^{19}$ m^{-3}.

described by Pots [13,16] which is based on the use of the gas law and of the ion energy balance.

In fig. 1 we present the measured ratios n_m/n_m^S for the 4p, 4d, 5d and 6d groups as functions of n_e for plasmas with a constant T_e of 3.5 eV and a roughly constant neutral density ranging from $n_a \approx 2 \times 10^{19}$ m^{-3} at low n_e to 0.5×10^{19} m^{-3} at high n_e-values. The ratios n_m/n_m^S decrease uniformly with n_e^{-2} until the value $n_m/n_m^S = 1$ is reached; this occurred at the values indicated in table 1. Since the ground state density n_{Ar} is approximately constant, this dependence is equivalent to constant level densities and constant CR-coefficients, $r_m^{(1)}$. The values of the $r_m^{(1)}$-coefficients are also tabulated in table 1, together with the n_e-ranges for the complete excitation saturation phase. The accuracy of the $r_m^{(1)}$-coefficients is mainly determined by the n_{Ar} and T_e-determinations and is estimated to be within a factor 2. Note that the levels are observed to

Table 1
The $r_m^{(1)}$ coefficients and n_e-ranges for the complete saturation phase for some groups.

Level	$r_m^{(1)}$	n_e-range for complete saturation phase	
		lower bound (m^{-3})	upper bound (m^{-3})
4p	2×10^{-4}	1×10^{19}	$\approx 2 \times 10^{21}$
4d	3×10^{-5}	1×10^{19}	7×10^{20}
5d	5×10^{-6}	1×10^{19}	3×10^{20}
6d	2×10^{-6}	1×10^{19}	2×10^{20}

come consecutively into Saha equilibrium with increasing n_e-values.

The measurements clearly indicate the existence of the complete excitation saturation phase for Ar I. From the observed transition to the saturation phase we can estimate the required electronic deexcitation rates. For the 4p-group (e.g. $\Sigma A = 4 \times 10^7$ s^{-1}) collisional deexcitation is larger than radiative decay for $n_e > 1 \times 10^{19}$ m^{-3} (cf. table 1), if the electronic deexcitation rate is $\langle \sigma v_e \rangle_{4p} \approx 4 \times 10^{-12}$ m^3 s^{-1}. The downward deexcitation rate 4p–4s is 10^{12} m^3 s^{-1} according to hydrogenic formulas [17]. Therefore, a total deexcitation rate of 4×10^{-12} m^3 s^{-1} due to 4p → 5p, 3d, 5s, 4d transitions seems reasonable in view of the small ΔE_{exc} and the large number of levels. The measurements indicate the need of these large deexcitation rates. A first implementation of these can be found in ref. [13].

The ratios n_m/n_m^S for several levels m are presented in fig. 2 as functions of the ionization energies of the levels, E_m^{ion}, for three values of n_e. The general trend is, that the level densities decrease with decreasing E_m^{ion}, even such that the slopes become more negative with decreasing E_m^{ion}, with distribution "temperatures" T' as low as 0.4–0.2 eV. Note, that T', defined as

$$kT' \equiv -\left(\frac{1}{n_m}\frac{dn_m}{dE_m^{ion}}\right)^{-1} = kT'(E_m^{ion}) , \qquad (2)$$

is not equal to the electron temperature T_e (like it is in PLTE), but is merely a measure of the slope of the Boltzmann plot. It should be stressed again [7] that in the saturation regime, excited density measurements are not a good way to determine T_e [18].

For $n_e = 1.5 \times 10^{19}$ m^{-3} the excited level densities scale roughly with $(E_m^{ion})^3$, equivalent to the anticipated scaling for hydrogen [3]. At higher n_e-values

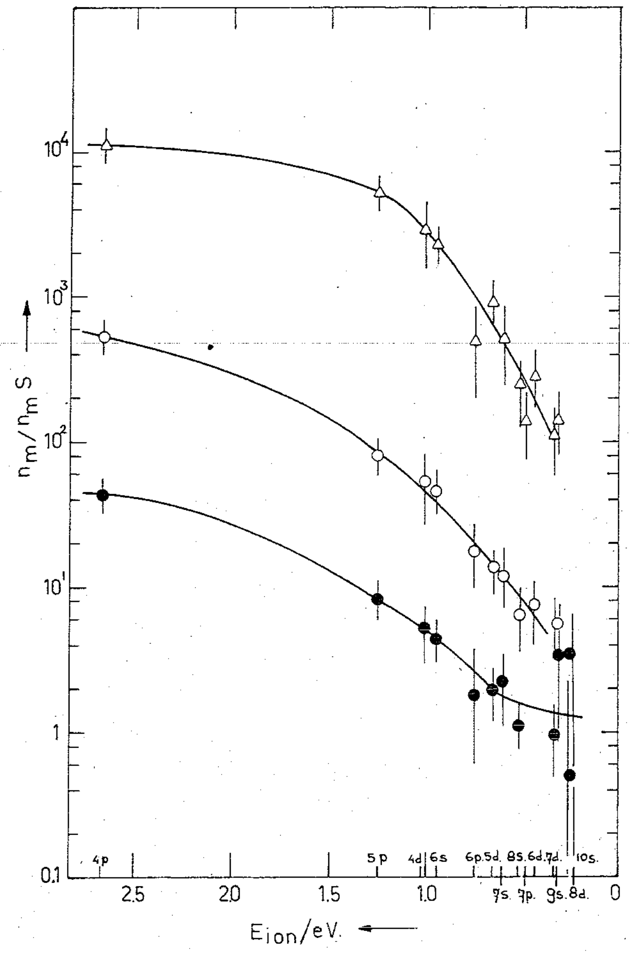

Fig. 2. The ratios n_m/n_m^S as functions of the ionization energy E_{im}^{ion} of the groups concerned; △: $n_e = 1.5 \times 10^{19}$ m^{-3}; ○: $n_e = 6.7 \times 10^{19}$ m^{-2}; ●: $n_e = 1.5 \times 10^{20}$ m^{-3}; T_e = 3 to 5 eV.

the slopes become smaller, especially for higher levels, which reflects the transitions to the PLTE regime. For the highest n_e-value the 7s, 6p, 5d and higher levels are in Saha equilibrium with the continuum (PLTE), as indicated by the values $n_m/n_m^S = 1$. In this regime, PLTE, of course $T' = T_e$. The observed form of the population distribution in the saturation phase [$n_m \propto (E_m^{ion})^3$] has been observed for more atomic systems and appears to be a general characteristic. The slope of the excitation distribution is such, that there is a roughly constant relation between the decreasing distribution "temperature", T', and the average energy difference between neighbouring levels, ΔE_m^{exc}. For argon, one can deduce from fig. 2 that kT' is an order of magnitude larger than ΔE_m^{exc} for subsequent levels, which again would indicate a ladder-climbing excitation flow from lower to higher excited states, up to the levels which are in PLTE. It should be worthwile to reconsider existing collisional radiative models for argon in view of the presented observations.

Furthermore, the expression for the density of the excited level m:

$$n_m \approx r_m^{(1)} n_m^{\mathrm{B}} = r_m^{(1)} n_{\mathrm{a}} \exp(-E_m^{\mathrm{exc}}/kT_{\mathrm{e}}) \,, \qquad (3)$$

suggests a method for the determination of T_e, if the level m is in the complete saturation phase. With known values for $r_m^{(1)}$, the excitation energy of the mth level, E_m^{exc} and n_a (from the gas law), eq. (3) yields a logarithmic expression for T_e in terms of the absolute density n_m.

We thank Mrs. M. v.d. Sande and A.A. Lubrecht for their contributions to the experiments.

References

[1] H.W. Drawin, Z. Phys. 186 (1965) 99; report Eur-CEA-FC-383 (1966).
[2] T. Fujimoto, Y. Ogata, I. Sugiyama, K. Tachibana and K. Fukuda, Japan J. Appl. Phys. 11 (1972) 718.
[3] T. Fujimoto, J. Phys. Soc. Japan 47 (1979) 265, 273.
[4] W. Kohsiek, J. Quant. Spectrosc. Radiat. Transfer 16 (1976) 1079.
[5] H.W. Drawin and F. Emard, report Eur-CEA-FC-534 (1970); Physica 85C (1977) 333.
[6] K. Tachibana and K. Fukuda, Japan J. Appl. Phys. 12 (1972) 895.
[7] N.P. Romanov, Opt. Spectr. 24 (1968) 468.
[8] K. Katsonis, thesis Univ. Paris Sud (1976).
[9] C.E. Webb, J. Appl. Phys. 39 (1968) 5441.
[10] L. Vriens, J. Appl. Phys. 49 (1978) 3814.
[11] J. Jolly, J. Quant. Spectrosc. Radiat. Transfer 20 (1978) 503.
[12] J.J.A.M. van der Mullen, B. van der Sijde, B.F.M. Pots and D.C. Schram, Proc. 4th Escampig (Essen, FRG, 1978) p. 101.
[13] B.F.M. Pots, thesis Eindhoven Univ. of Technology (1979).
[14] W.L. Wiese, M.W. Smith and B.M. Miles, Atomic transition probabilities, part II (Nat. Bureau of Standards, 1969).
[15] B. van der Sijde et al., Proc. XIVth ICPIG (Grenoble, 1979), J. de Phys. C7 (1979) 851.
[16] B.F.M. Pots, P. van Hooff, D.C. Schram and B. van der Sijde, to be published.
[17] C.M. Ferreira and J.L. Delcroix, J. de Phys. 36 (1975) 1241.
[18] M. Ribau Teixeira and F. Carvalho Rodrigues, J. Phys. D12 (1979) 2173.