

CORONAL TYPE AND COLLISIONAL-RADIATIVE MODELLING OF RARE GASES PLASMAS

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Abstract. Detailed modeling of rare gases plasmas, taking into consideration their atomic structure specificity is relevant to laboratory studies and industrial applications and also essential for astrophysical research. We are here describing the general trends of their Collisional-Radiative modeling, insisting also to well known limiting cases, especially of the Coronal type. The atomic data used in such modelings are shown to be indispensable for the evaluation of the global plasma properties and to play a determinant role for obtaining a satisfactory optical diagnostics.

1. Introduction

Collisional-Radiative (C-R) modeling of plasmas has been early used [1] for evaluating the population of excited states in Non-Local Thermodynamic Equilibrium (NLTE) plasmas. This type of modeling is indispensable for evaluating the optical measurements of spectroscopic diagnostics whenever the Local Thermodynamic Equilibrium (LTE) conditions are not fulfilled. The method has been extended to frequently used plasmas containing more complicated species, notably to rare gas ones [2]. Furthermore, effective levels and complicated atomic structure have been introduced in order to describe in detail the NLTE plasma characteristics [3]. The success of the method greatly depends on the quality of the atomic database used in the evaluation of the rate coefficients occurring in the set of statistical equations based in the Boltzmann kinetic equation, which constitutes the basis of the C-R model. Such a database is now under development in our laboratory, concerning Xe and its ions, an element which is widely used in Stationary Plasma Thrusters (SPT) for space applications.

Another, important generalization is obtained in examining a multitude of ionized species instead of dealing only with the neutral and the first ionized ones. Such models are extensively used for high-Z laser produced plasmas, theta-pinchs etc, where the sufficiently high temperature justifies inclusion of numerous ionization stages. In such systems, which could demonstrate the relative importance of some particular processes [4], it is often advantageous to resolve especially modulated systems instead of the fully detailed configuration models including a huge amount of statistical equations. The study of plasma reactors is another field where the successively ionized species have to be taken into account in order to obtain realistic models of the C-R type. Except of the presence of twice and three-times ionized species of one element, other elements and molecules have often to be introduced in those models. Finally, C-R type modeling become of interest to magnetically confined plasmas in order to take account of the mechanisms related to the presence of a Scrape-off Layer (SOL) in the outmost region of the confined plasma near the edge and of the functioning of the limiter.

Inversely, reducing the number of reactions taken into account in the formulation of the aforementioned statistical equations may occasionally lead to a better insight of the plasma equilibrium conditions, provided that these conditions comply with such a simplification. Whenever it is the case we may advantageously use models of the Coronal (CO) or Coronal-Radiative (CO-R) type, as it will be explained later on. This can happen when the equilibrium described by the C-R model, the so-called Collisional-Radiative Equilibrium (C-RE) with decreasing density goes to a Coronal Equilibrium (CE) limit. Then, the Electron collision Ionization (EI) and the Radiative Recombination (RR) play a determinant role, see e.g. [3]. Additional consideration of the autoionization – dielectronic recombination couple, of the three-body recombination and of some radiative processes as electron collision de-excitation and spontaneous decay could be more adapted in other cases (CO-R models).

We have used the CO model for studying the SPT and their hollow cathode plasmas, both in the plume and the spot mode. The necessary ionization data have been evaluated by semi-classical calculations based in the Classical Trajectory Monte Carlo (CTMC) method [5] and by standard empirical formulas [6]. Recombination data have been taken from a NIFS review [7], from original publications and calculated using quasi-classical approximations. The obtained results for a Xe propellant gas are in agreement with recently published [8,9] experimental ones, justifying especially the observed quasi-absence of Xe I lines

and the fast mode change from 1 to 3 eV. We have observed lately an analogous variation in a prototype SPT-50 made available to us at the LPTP Laboratory of the Ecole Polytechnique [10].

Although the modelings described here are useful in various applications, in this paper emphasis is given to SPT applications and examples. We first (§ 2) present some general considerations on the C-R, CO and CO-R modeling. In § 3 these are applied to the SPT modeling for Ar and Xe low temperature plasmas. Section 4 is dealing with Atomic and Molecular (A+M) data needed for the type of modeling we are addressing here. Our conclusions and perspectives for future work are given in § 5; they will be discussed further in the Meeting.

2. Zero-Dimension Modeling of Rare Gas Plasmas.

As it was mentioned previously, the ions circulating in the SPT are mainly produced by electron collisions with the propellant gas and its previously formed ions. The available electric energy is principally spent to their production and acceleration. We have then a typical case of ionizing plasma. It is therefore very important to correctly evaluate the rates of the ion production through the corresponding ionization cross sections, which are integrated over an electron distribution; in a first approximation, for the formulation of the latter a sole Maxwellian can be used. For higher electron temperatures, one is obliged to take into consideration an increasing number q of ionized species. We address subsequently the problem of determining an adequate value of q for each modeling case. In the case of SPT plasmas we have to take into consideration that the successive ionizations of the rare gases are concerning six equivalent valence electrons with relatively low ionization thresholds [11]. By simple inspection of the cross sections corresponding to each ionic species production [12] we can be sure that no satisfactory modeling can be obtained and, at the same time, no correct optical diagnostics, if we take only one sole ionization stage into consideration. It has to be stressed that the significant tail part of the Maxwellian distribution attributed to the electrons plays an important role in the plasma ionization mechanism. Furthermore, the excited states of these species have to be somehow taken into consideration, especially the long living metastable ones. Finally, sufficiently intense transitions, which are handy for the non-intrusive optical diagnostics must be incorporated into the model. The atomic structure of the rare gas and of their ions has also to be correctly taken into account, in order to be at least able to evaluate the contribution of the lines included inside their multiplets. The Boltzmann statistical equation can constitute a basis for writing down a system of differential equations, which in principle could be resolved and give the population of any species in any place. This is a too ambitious scheme, which can be simplified in various ways. Especially when the local characteristics of the plasma are of principal importance as is the case for local diagnostic purposes, we may neglect both the moments in the LHS of the Boltzmann equation and the time variation as well, and keep only its RHS part. By means of those simplifications we arrive to a Homogeneous and Stationary (HS) plasma hypothesis, which can be proved sufficient for local diagnostics of quiet plasmas.

Thusly, a set of statistical equations is written down for each species, which include the corresponding atomic parameters. The sets of all the considered species have to be put together and the obtained system including all the creation and destruction terms has to be resolved together with a quasi-neutrality closure condition for the plasma of the type

$$n_i^{(1)} + 2 n_i^{(2)} + 3n_i^{(3)} + 4n_i^{(4)} + 5n_i^{(5)} + \dots + qn_i^{(q)} = n_e \quad (1)$$

where the subscript i pertains to all the considered excited states of each species and the numerical superscript corresponds to the charge of each species. Eq. (1) shows that each multi-charged species, even when less abundant, plays a role weighted proportionally to its charge.

A very tedious work is necessary for writing down the statistical equations with the proper atomic coefficients even for a sole ionization stage. As this has been well documented elsewhere we do not report it here in detail (see e.g. in [13] for the construction of the energy levels and transition probabilities scheme). An example of C-R model written previously for the Ar atom in jK-coupling scheme that is mostly prevailing for the rare gases atoms and their ions is available in [3]. Subsequently, this work has been often followed and extended [14-17] for the needs of the argon modeling. Whenever more than one ionization stage is included, direct transitions from one species equation set to another not contiguous, are also to be included, as is the case e.g. for the double ionization which has a smaller but evidently not negligible cross section value [18]. Also, for higher electron densities, the two-electrons transitions of autoionization and its

inverse, resonant capture, play an increasingly important role in the plasma ionization and hence in the energy balance [4].

Because of the huge number of unknown atomic data for Ar, and much more for Xe and their ions which are needed for a satisfactory C-R modeling, a consistent effort has been devised and is now underway [12, 19] in order to produce consistent sets of A+M data for modeling of rare gas plasmas needed in a broad spectrum of applications.

Hopefully, prior to the completion of the aforementioned detailed and extensive work, a number of important conclusions can be reached by means of simplified models suitable for application in some cases. As is well known, whenever the plasma is in LTE with a Maxwellian distribution function pertaining to the common temperature, the Boltzmann distribution law for the excited states, the Saha equation relating the densities of the ionic species and the radiation law of Planck, are sufficient for its description. This is the complete LTE case. A partial LTE is established if the Saha equation is at least valid for the higher excited levels, although the lower part of them is somehow detached in a corona-like situation that is described subsequently. If the plasma is not in LTE, still non-local thermodynamic equilibrium conditions may prevail, attributing to each species a characteristic temperature (NLTE case). A C-R model becomes then necessary for its description.

In the SPT case, the strongly ionizing conditions are not allowing a simple evaluation according to the LTE laws, but we can take advantage of another well studied condition instead, which has been known from the study of astrophysical plasmas [20] namely the CE, because it prevails in the solar corona. Its significance is easier understood if we look to the bulk compartment of the plasma. More generally, the description of a plasma, i.e. the number densities n_i of each species as a function of its electronic density n_e for a defined electronic temperature T_e , is given by a curve which has two asymptotes. Suppose for simplifying that we are looking for singly ionized plasmas with Ground State (GS) density n_1 . In a region where the plasma has high n_e and also sufficiently high n_1 , and, consequently, the equilibrium criteria can be satisfied [21] provided T_e is also sufficiently high, the populations approximate those foreshown by LTE. An asymptotic variation law

$$n_1 = f(T_e)n_e^2 \quad (2)$$

is reached then, coming from the Saha law which foresees a variation $n_e/n_1^* \sim 1/n_e$; the star indicates as usually the LTE population. Due to the high temperature, a great number of excited states may then exist, but the GS population of the neutrals follows simply the Saha law. On the opposite side, in a lower n_e and n_1 region, another kind of equilibrium is reached, with asymptotic variation according to a

$$n_1 = \phi(T_e)n_e \quad (3)$$

law. It corresponds to a special case where the excited states, if any, play a non-important role. It means that the ionization of the GS neutrals by EI and the recombination by RR to the GS are the sole essential processes. Bates has given an approximate criterion for the region where CE is expected [8]. Note that the CE is favored whenever the plasmas are not reabsorbing the radiation provided by the atomic processes involving their own constituents, a case known as optically thin plasmas.

If we neglect the presence of multiply charged ions the system of statistical equations shrinks in the case of CE limit to one sole equation, which leads to Eq. (3) because for a given T_e the rates of the EI and RR processes are constant. We obtain directly Eq. (3) on the basis of the micro-reversibility principle, because here the significant processes are only ionization from GS to continuum and back, having densities n_1 and $n_+ = n_e$. The same reasoning that is giving the Boltzmann law relating the excited states with the GS populations $n_i = n_1 \exp(\chi T)$ leads also here to Eq. (3).

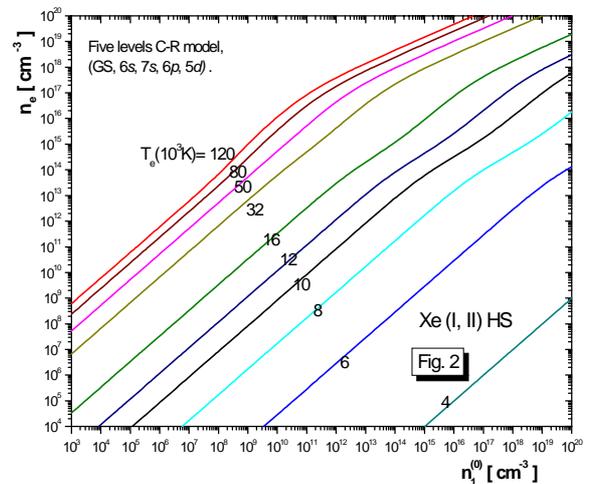
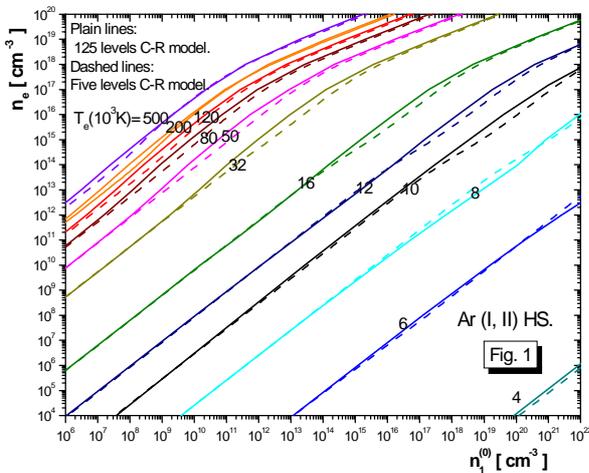
The two asymptotes are connected by a more or less smooth curve following a NLTE variation described by the C-R model. For the same T_e , as the n_e lowers we pass from ETL with $n_1 \sim n_e^2$ to a partial LTE curve according to the C-R model, followed by a CO model variation with $n_1 \sim n_e$. This character of the plasma in the corresponding regions was previously observed in the case of Ar plasma C-R calculations [3] as is discussed in § 3.

We remind here that the C-R type models have mainly “zero” dimensions. This means that the populations $f(T_e)$, expressed by the model can be only locally valid and for HS plasmas, if the LHS of statistical equations are set to zero. However, it is possible to adapt the C-R models to non-stationary and inhomogeneous plasmas, keeping in the LHS the moments concerning time and space variation.

3. Corona modeling at the limit of C-RE.

Given the conditions often encountered in SPT we can expect that a CO model could be sufficient for the local description of the plasma, at least in some regions. In order to illustrate the expected validity regions of various types of modeling, we show in Fig. 1 the results (plain lines) of a one ionization stage model for Ar (noted Ar I, II) from [3]. The applicability of this model is severely restricted to sufficiently low temperatures, where the presence of doubly and more ionized species can be neglected. Hence, the curves corresponding to higher temperatures are only given here in order to illustrate the general trends. Also, we remind that Fig. 1 concerns only HS plasmas because the LHS part of the Boltzmann equation was set to zero and the results shown can be applied only locally. The upper part of the figure shows straight lines of slope $\frac{1}{2}$, giving the LTE condition variation according to the Eq. (2). These lines subsequently bend due to the installation of a C-RE and their slopes increase generally smoothly. In the lower part of the figure the isotherm lines become again straight with a slope reaching 1, in accordance with Eq. (3). The corona limit is reached in this region. Let us compare now the results of a full 125 level C-R model as is this of [3] with a very simplified model containing only five levels, namely the GS and the excited states $4s$, $5s$, $4p$ and $3d$, without discrimination between the core j_c values $1/3$ and $2/3$, also shown in Fig. 1 (dashed lines). In view of the logarithmic scale used here, the differences illustrated in Fig. 1 are not overwhelming; nevertheless, we arrive up to half an order of magnitude differences, mostly in the regions where the C-RE prevails. Lack of precise C-R modeling in this region hinders any successful diagnostics. In regions where the LTE is prevailing, the discrepancy between the two models results tends to zero, because the presence of more excited levels play no role in the Saha equilibrium with the continuum. Moreover, we observe that for sufficiently low T_e the excitation of the GS is negligible, be it either fully or partially described in the model. The simplified model offers no interest at all, and a CO model is giving satisfactory results. Still, the corona model gives only the populations of the GS, in fact for the simplified case examined here only this of the neutral atom GS. Therefore, it cannot help for any diagnostics based in excited levels. A simplified model including selected diagnostic lines may therefore be useful, provided we are not going too far in the C-R region, where the approximation is too crude. Such models, the interest of which appears basically in diagnostics, are known as CO-R.

An analogous picture is shown in the Fig. 2 for the case of a Xe I, II model. In the sole model shown here, only four excited states $6s$, $7s$, $6p$ and $5d$, plus the GS of the neutral atom are taken into consideration. Because of the lower ionization potential (I_p) of the Xe atom, the whole system of curves is skipped to higher electronic densities, but the general trends remain obviously the same. This means that in a SPT the same T_e and GS densities correspond to higher n_e for a Xe propellant than for Ar. Equivalently, for the same n_e (and consequently the same ion density) the Xe plasma is significantly (about 25 %) cooler. The Xe – Ar analogy suggests that for local modeling of the SPT with Xe propellant, in regions where the T_e is low, the CO model can give important information. This complies with the experiment [8,9], where the authors claim that the Xe plasma reaches in fact the coronal limit. Moreover, where the CO modeling for Ar functioning is valid, it is expected to be also valid for the Xe gas.

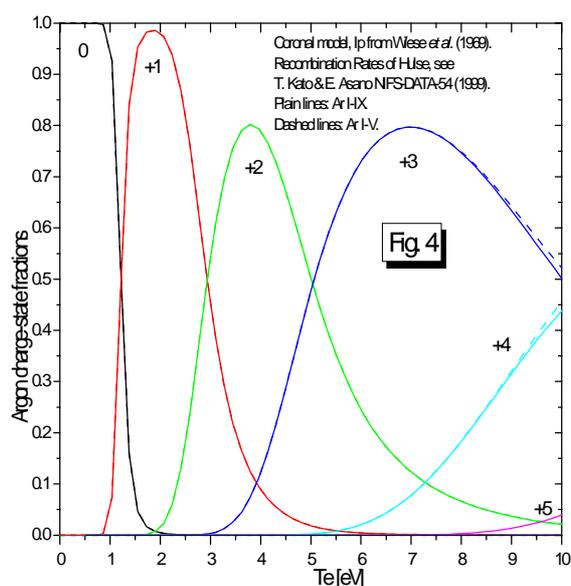
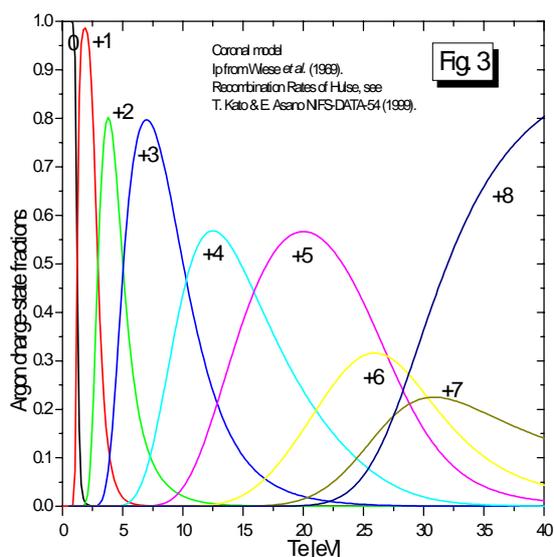


3.1. Multiply ionized species.

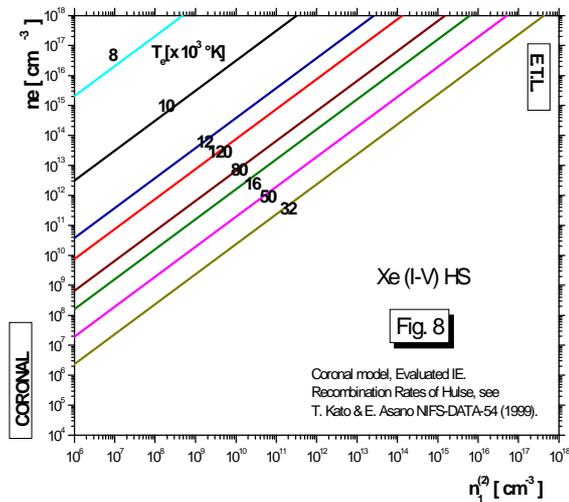
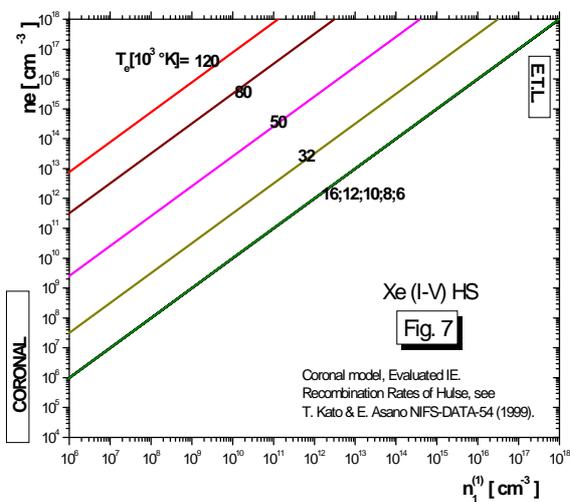
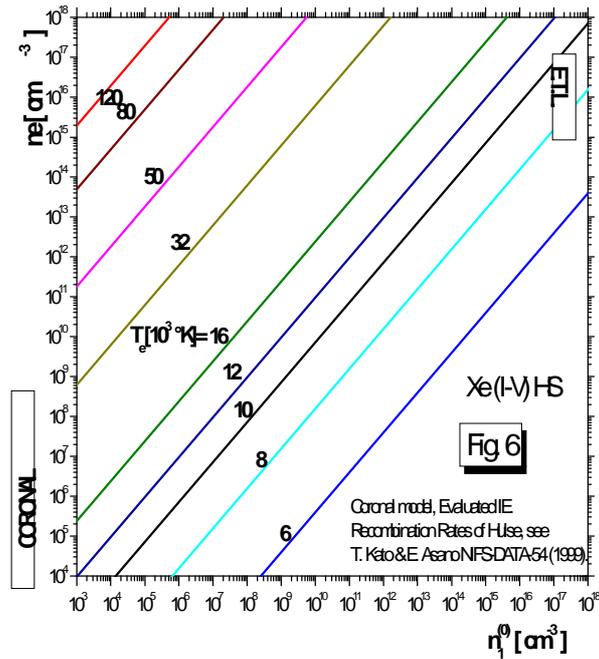
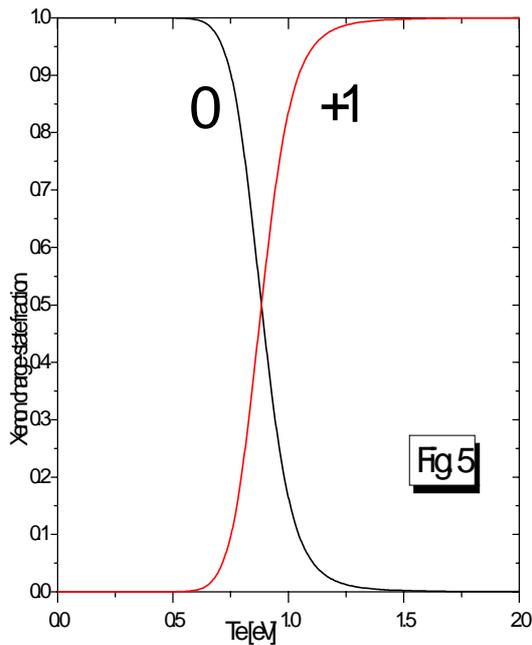
The observations of the previous paragraph made for only once ionized rare gases can be extended to plasmas containing multiply ionized atoms. In order to simplify the system we restrict again to the HS case. Then, for relatively low T_e , more important is to include many ionization stages, even in a coronal scheme neglecting the role of the excited states, than the full treatment of the neutral atom. In the former case we arrive to a system of N statistical equations, one for each ionization stage included, plus the quasi-neutrality condition (1), for $N+1$ unknowns including the $n_i^{(0)}$ population for the neutral atom, with $i = 1$ for the GS populations. Because in this system the off-diagonal elements are zero, it is possible to get the solution $n_i^{(q)} = f(T_e, n_e)$ with q varying from 0 to N , simply by successive divisions. Could we choose to include some excited states, say four as was the case with the Xe I, II simplified model, on the top of the GS and this for each ionization stage including the neutral, except for the last one, the corresponding model would call for a non-diagonal $5x(N+1)$ system solution. Still, with only four excited levels per stage, we have seen that the expected result is not satisfactory in the C-RE region. On the contrary, in the low T_e, n_e region where the coronal equilibrium prevails, the multi-species CO model is expected to correctly describe the global plasma properties and an insufficient excited level addition could only complicate the problem.

The question of the choice of the number q value in Eq. (1), i.e. of the maximum value of N taken into consideration in a multi-stage model, has now to be addressed. The reply depends of course on the temperature region of interest. Because of the atomic structure of the rare gas atom, one is tempted here to consider up to eight times ionized species which have their outer shell totally stripped, after losing the $6p$ electrons and two s ones. The results of the corresponding Ar I-IX CO model can be presented under the form of percentages of the $n_i^{(q)}$ densities for each T_e (see Fig. 3) that are not a function of the n_e as far as the CE is valid. Anticipating that the most important ions in the case of the SPT are the first four, we have also elaborated an Ar I-V CO model, the results of which are given in Fig. 4. It is easy to conclude that for a SPT modeling the latter is sufficient, as it gives practically the same results for temperatures up to 10 eV. The one sole ionization stage model Xe I, II of the Fig. 2 is giving the percentage curves of Fig. 5, which are manifestly of significantly different type and can only be used in very low temperature considerations, e.g. in the hollow cathode region as was the case in [8]. Again, CO models are typically giving the same population percentage for any n_e (the curves in Figs. 1, 2 in the lower part become straight lines for any n_e with n_e proportional to n_i) but in fact this is only valid for sufficiently low T_e in order to satisfy the Bates' criterion for CE.

The CO modeling is giving an approximate evaluation of the plasma composition if the temperature is known. E.g. for a SPT fed by Ar, a small variation of the temperature from 0.8 to 1.5 eV can lead to a variation of the $n^{(1)}, n^{(0)}$ percentages from 1 %, 99 % to 93 %, 7 % correspondingly, according to Fig. 4.



As another example we are giving in Fig. 6 the results of the Xe I-V CO model in the form of the n_e variation for each T_e . Using this figure as a template, we can evaluate the single ion density $n_1^{(0)}$ if the electronic density and temperature n_e, T_e are known. For a temperature of 0.5 eV and an electron density of 10^{12} cm^{-3} the corresponding $n_1^{(0)}$ is $3 \cdot 10^{16} \text{ cm}^{-3}$. Also, we can get the electronic temperature if the n_e and $n_1^{(0)}$ of any ion can be measured (see e.g. Fig. 7 for the case of the Xe^+ ionized species density $n_1^{(1)}$ and Fig. 8 for the case of the Xe^{++} doubly ionized species density $n_1^{(2)}$). Still, all these results are practically useful for diagnostics only if we can measure the continuum density(ies). For the common diagnostic using discrete lines of the neutral or other species, a CO-R model including the measured lines has to be used, and in case that C-RE prevails, a full C-R one.



4. The A+M database.

The A+M evaluation part of the C-R, CO and CO-R modelings is mandatory for obtaining confident results, useful for explaining the global comportment of the plasma and to elucidate its properties. They must also be sufficiently precise in order to allow a satisfactory plasma diagnostics. The quality of the input data is directly conditioning the validity of the obtained output [17]. Especially for A+M data requirements of SPT a detailed inventory has been given and the available evaluation methods to be used in each case succinctly described [19]. The first results obtained within our evaluated database project were reported in [12]. They include CTMC method evaluation of cross sections for electron – atom (ion) collision processes, mainly ionization and excitation [23]. The electron collision single ionization being an essential process in the SPT case, we addressed it with priority and in detail. In so doing, a convenient formula has been devised for the EI cross sections evaluation, which is directly derived to the often-used Drawin formula [6]. Besides describing correctly the ionization process both for low and high energies, as was already the case for the latter, the maximum of each ionization is now found in the collision energy given by $E_{n+2} = E_n + E_{n+1}$ with n , $n+1$, $n+2$ indicating the successive ionization stages; the maximum value include always the number of equivalent electrons ξ available for ionization in the external shell having the same quantum number (here l) but its absolute value is calculated within the CTMC method approximation. This method is giving the correct variation of the cross section near the threshold, in accordance with the Wannier law [24], where the low energy of the electrons in the SPT is mostly contributing to the ionization rate.

The cross section for the RR process was also evaluated, on the basis of semi-classical approximations [3, 25, 26] in comparison with previous evaluations [7]. More detailed evaluation will be needed for this process; it can be tackled on the basis of full quantum calculations giving the total photo-recombination spectra [27].

Work is in progress, both for refining the available atomic data for Xe and other rare gases and also for evaluating other data categories, as transition probabilities, desexcitation etc. The whole evaluation effort of our group is eased by the collaboration within the A+M Data Network managed by the IAEA. Many data centers participating in this network are keeping databases in the Web, containing useful material for evaluation.

We finally note that A+M cross sections are entering in the equations of various models under the form of rates, which we have obtained by integrating according to a Maxwellian distribution of the electrons as a function of temperature. Although this is a commonly followed procedure, cannot always be guaranteed, and we are looking forward to consider two Maxwellians or other distributions for the rate calculation.

5. Conclusions and perspectives of the C-R modeling.

The modeling of the types we are describing here is giving significant results reflecting the intrinsic characteristics of the plasma constituents. These results are necessary for a trustful optical diagnostics. They can also be introduced in detailed modeling codes in order to improve their capacity to describe the SPT functioning. Valuable modeling work has been done lately in this direction. Also, complex codes existing for the description of the plasma properties under confinement, as is e.g. the well-known EIRENE code, developed for the needs of the controlled fusion, could be adapted to the SPT conditions.

An extension the C-R type modeling to transient plasmas can be obtained if we keep the time variations of the distribution functions df_i/dt in the LHS part of the Boltzmann equation [28]. As the population of the GS is expected to be significantly higher than this of the excited states, the variations of the latter are often neglected before this of the former. Then, the equation corresponding to the GS becomes differential and has a nonzero LHS. Of course, this approximation has to be somehow modified in case we consider plasmas with more than one continuum, belonging to various ionized species. Depending on the temperature, the population of the GS of the neutral atom for such plasmas, may even become negligible before the population of the GS of various ionization species, as a simple CO model can show.

Finally, it seems that the improvement of the A+M database of Xe and of its ions and experimental validation of the elaborate rare gases C-R models are of paramount importance before the industrial application of the latter. We are actually working toward this aim. In so doing, the results obtained as a function of the evaluated input data are studied and will be compared between them and with the

experiment [10]. Using adequate searching engines for bibliographic and numerical databases available in the Web is greatly helping this task [29].

This work is effectuated in the frame of the Groupement de Recherche CNRS/CNES/SNECMA/ONERA 2232 “Propulsion à Plasma pour Systèmes Spatiaux”.

Abbreviations

A+M Atomic and Molecular
C-R Collisional-Radiative
C-RE Collisional-Radiative Equilibrium
CE Coronal Equilibrium
CO Coronal
CO-R Coronal-Radiative
CTMC Classical Trajectory Monte Carlo
EI Electron collision Ionization
GS Ground State
HS Homogeneous and Stationary
I_p Ionization Potential
IAEA/AMD International Atomic Energy Agency / A+M Data Unit, Vienna
LHS, RHS Left, Right Hand Side
LTE Local Thermodynamic Equilibrium
NIFS National Institute of Fusion Science, Japan
NLTE Non-Local Thermodynamic Equilibrium
RR Radiative Recombination (Photo-recombination)
SOL Scrape-off Layer
SPT Stationary Plasma Thruster
SPT-50 Stationary Plasma Thruster prototype of 50 mm diameter

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