COLLISIONAL-RADIATIVE TYPE MODELLING AND APPLICATION IN PLASMA DIAGNOSTICS

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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 type modeling including some examples. It is shown how the atomic data to be used in such modeling are determining the global plasma properties description and therefore data evaluation is indispensable for obtaining a satisfactory optical diagnostics.

Keywords: Collisional-Radiative, Coronal, Coronal-Radiative, Modeling, Diagnostics.

1. Introduction

Modeling of the Collisional-Radiative (C-R) type has been extensively used in the study of Non-Local Thermodynamic Equilibrium (NLTE) plasmas encountered in the laboratory, of natural plasmas (terrestrial and stellar atmospheres) and plasmas obtained in various industrial applications (e.g. controlled fusion, plasma reactors, ionic propulsion). In an effort to cover a wide field of applications, C-R modeling was extended from the initial H plasmas study [1] to plasmas containing less simple species, beginning with the often used in the laboratory rare gas plasmas [2]. Furthermore, effective levels and complicated atomic and molecular structure have been introduced in order to describe in detail the NLTE plasma characteristics [3] and also numerous successive ionization stages whenever the temperature of the studied plasmas becomes higher [4]. The success of the method greatly depends on the quality of the atomic database used in the evaluation of the rate coefficients, which are occurring in the set of statistical equations formulated using the Boltzmann kinetic equation and constituting the basis of the C-R model. Such a database is now under development in the LPGP laboratory, concerning Ar and Xe and their ions. The first of them is very commonly used in laboratory and industrial applications; the second is widely used in Stationary Plasma Thrusters (SPT) for space applications.

C-R models are so popular because they become indispensable whenever the universal laws of Boltzmann and Saha, valid only in plasmas with at least Local Thermodynamic Equilibrium (LTE), are failing and the velocity distribution of the particles (and photons gas...
distribution) is not anymore following the Maxwell velocity distribution function (and the frequency distribution radiation law of Plank). Then, detailed evaluation of the expected populations of both levels generating the measured lines becomes only possible by using C-R modeling. For a given electron density \( n_e \) and distribution function (defining an electron temperature \( T_e \) of Maxwellian) C-R models are used in evaluating the local populations of the atomic (and molecular, if any) levels of the species encountered in the plasmas (atoms, molecules and ions), for the calculation of the abundance of the ionized species and also to appreciate the contribution of each atomic or molecular process in the overall properties of homogeneous and stationary (HS) plasmas; in this simplified case the RHS of the Boltzmann kinetic equation is taken equal to zero and the system of equations becomes a linear and non-differential set of the populations. Moreover, C-R type models may evaluate the transport contribution and the time variation in non-homogeneous transient plasmas if the LHS terms of the Boltzmann equation are taken into consideration. This equation containing too much information, its successive moments are taken for the formulation of a system of (in general differential) equations describing the plasma. Furthermore, the so obtained contributions of each species and level are used in the elaboration of specific types of plasma modeling. Here we are oriented towards diagnostic applications of the C-R type modeling, related to spectral line intensity measurements, allowing e.g. plasma temperature evaluation. Notably, simplified Coronal (CO) and Coronal-Radiative (CO-R) models are to consider, because they can already give selected plasma characteristics, through the measurement of the continuum radiation and also of the intensities of selected emission lines for which atomic data are known. Common C-R type modelings are briefly described, together with some examples of application in the diagnostics of rare gas plasmas.

An important generalization of the C-R type modeling consists 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-pinches etc, where inclusion of numerous ionization stages becomes compulsory due to the sufficiently high temperature. In such systems, in which the relative importance of some particular processes has been demonstrated [5], it is often advantageous to resolve especially modulated systems instead of fully detailed configuration models which are including a huge amount of statistical equations. The study of plasma reactors is another field where successively ionized species have to be taken into account in order to obtain realistic models of the C-R type. Besides twice and three-times ionized atomic species, various molecules and their ions 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. We also mention plasma-surface interaction processes, which, although may become very important depending on the plasma density, are not addressed here.

Inversely, reduction of the number of reactions taken into account in formulating 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 or Kinetic Equilibrium (C-RE, KE) 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 considered in some cases.

We have used the CO model for studying the SPT and their hollow cathode plasmas, both
in the plume and the spot mode. Especially for the hollow cathode plasmas which are
expected to be nearer to the CO equilibrium, our calculations for Xe propellant gases are in
agreement with previously available experimental results for Xe [6,7], justifying the quasi-
absence of Xe I lines and the fast mode changing for variation of the electron temperature
from 1 to 3 eV reported for these experiments. We have been able to observe lately a similar
variation for both Ar and Xe propellants feeding of the cathode in the absence of the main
stream, in a prototype SPT-50 propulsion device, made available to us at the LPTP
Laboratory of the “Ecole Polytechnique” [8]. Furthermore, when the main discharge is on, an
important variation of the ionized versus neutral species percentage has also been seen as a
function of the plasma region and of its temperature. The experimental details and obtained
spectra will be given separately [9]. For the CO model formulation, the necessary ionization
data have been evaluated by semi-classical calculations based in the Classical Trajectory
data have been taken from original publications calculated using quasi-classical
approximation and compared with a NIFS review [12]. In order to perform a detailed optical
diagnostics, a CO-R modeling including at least part of the observed lines of the neutral and
ionized species has to be developed. Towards this aim, the necessary transition probabilities
are being calculated in the Coulomb approximation and the excitation-de-excitation cross
sections are calculated with the empirical Drawin formulas [11] compared with CTMC
calculations [13].

Although the types of modeling reported 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. Section 3 is dealing with Atomic
and Molecular (A+M) data needed for the C-R type of modeling, with SPT modeling for Ar
and Xe low temperature plasmas in mind. Our conclusions together with perspectives for
future work are given in § 4; they will be discussed further in the Meeting.

2. Modeling of Plasmas in Kinetic Equilibrium

Electron collisions with the propellant gas and its previously formed ions are mainly
producing the ions circulating in the SPT. We have then a typical case of ionizing plasma in
which the available electric energy is principally spent to the production and acceleration of
the ions. It is therefore very important to correctly evaluate the rates of the ion production
through the corresponding ionization cross sections, which are subsequently integrated over
the 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. The situation is similar in various
applications, where the rare gas plasmas are very common; note that the successive
ionizations of the rare gases are concerning six equivalent valence electrons with relatively
low ionization thresholds [14]. By simple inspection of the cross sections corresponding to
each ionic species production [15] 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, except in very low temperatures. 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, in presence of abundant
electrons 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 coming from lines with $j_c = 1/2, 3/2$ core values for Ar, Kr and Xe atoms, included inside each multiplet.

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 minimum Homogeneous and Stationary (HS) plasma hypothesis, which may be proved often 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 constituents’ densities, of the type

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

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. With increase of the temperature, the abundances of the neutral and even of the first ionized species may become negligible and the equilibrium is based on the presence of the more ionized species.

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 [16] for the construction of the energy levels and transition probabilities scheme). An example of C-R model written previously for the Ar atom in $j/K$-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 [17-20] 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 [21]. 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 [5].

In most application cases a simple evaluation according to the LTE laws is not possible [22]. Yet, in the SPT case in presence of strongly ionizing conditions we can take advantage of another well studied equilibrium type instead, which has been known from the study of astrophysical plasmas [23] namely the CE, because it prevails in the solar corona. It corresponds to a special case where the excited states, if any, play a non-important role and applies when for a given temperature, $n_e$ is varying proportionally to $n_i^{(0)}$ (here only the first ionization state is considered for simplicity). In this case the ionization of the GS neutrals by EI and the recombination by RR to the GS are the sole essential processes [24]. Bates has given an approximate criterion for the region where CE is expected [6]. 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. Neglecting again the presence of multiply charged ions the system of statistical equations shrinks in the case of CE limit to one sole equation.

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 only for stationary 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. But in case that the plasma is definitely far from HS conditions, detailed codes have to be developed taking into account the magnetic fields, the instabilities, the containment vessel surface etc. We mention here the code EIRENE (see: www.eirene.de) developed in the Institute für Plasmaphysik, Jülich for the cold Tokamak plasmas modeling near the wall. This type of integrated numerical experiment includes the plasma physics, the A+M processes and the plasma-material interaction. Therefore, this code can also be directly adapted to a wide class of applications, as the simulation of high intensity discharge for lighting, of plasmas appearing in space applications as reentrance and propulsion etc., provided:

1) The code has been adapted to the geometry of the application we are interested in and to the form of any present E/M field
2) The A+M data concerning the encountered gases are evaluated and incorporated.

In view of the success of the exhauster’s implementation in Tokamaks, a study for the optimization of the magnetic field in the particular case of SPT is also foreshown, in order to obtain an exhausting of the charged particles under ergodic condition, meant to minimize the erosion damages. In such a configuration, instabilities could play a lesser role. Also, the molecular effects, which are very important in Tokamak edge plasmas, have in general to be investigated and probably included in the C-R modeling meant for various rare gas plasmas encountered in experiment and industrial devices and also in the nature.


As mentioned in the introduction, C-R type modeling can give the percentage of ionization stages corresponding to the electron density and temperature and the populations of the excited levels for each species and also allows estimation of the importance of the various processes. We have seen that for low temperature Ar plasmas, presumably containing only $\text{Ar}^0$ and $\text{Ar}^+$ species, various models have been developed which are giving essentially the same results. This is clearly insufficient for the presently studied experimental and industrial plasmas, where also Kr and Xe C-R modeling is needed. Therefore, we are running an extended project of C-R type modeling from early 2000 (see e.g. [25]). Seeking a validation of our models, we perform in parallel optical diagnostics in the aforementioned SPT-50 prototype [8]. Measurements include optical and UV lines in the following four plasma regions:

a) hollow cathode, axial direction, without the main discharge,
b) exit of the channel,
c) region of merging of the cathode electrons with the ion jet,
d) advanced jet region (typically 50 cm from the channel exit).

All these measurements are done separately with Argon and Xenon feeding in various optical angles. We have now advanced the position of the prototype inside the vessel, in order to be able to repeat the measurements with the available ports, in a direction perpendicular to the jet. Thus, a complete optical study of the plasma will be obtained for the Ar/Xe cases.
Figure 1: Recorded spectrum in the cathodic zone

Figure 2: Recorded spectrum in the anodic zone

Figure 3: Recorded spectrum in the plasma jet
The continua radiations of the plasmas will be also diagnosed whenever the necessary technical facilities could be implemented. Also, the hollow cathodes will be changed and the SPT functioning conditions varied, in order to reach an optimization for the case of powerful plasma thrusters. As an example, in Figs. 1, 2, 3 are shown typical unresolved spectra of the cases a), c) and d) correspondingly, with pure Argon feeding. These figures are illustrating the aforementioned (§1) important variation of the spectrum according to the plasma regions. Note that the wavelengths scale given in Fig. 3 is also valid for Figs. 1,2. Except the noted impurities, the shown lines are mostly identified as coming from Ar and its ions. As this is not the subject of the present review, the corresponding detailed spectra and the optical characteristics of the spectrograph will be given in a poster [9] of the Meeting. The OH bands appearing in Fig. 1 allowed an evaluation of the corresponding rotational temperature for the hollow cathode alone to more than 1.5 keV, provided the distribution follow the Boltzmann law [26]. Similar measurements with Xe gas feeding, also in presence of the main discharge, are underway, which will allow for a better comparison with analogous data obtained in the PIVOINE test facility [27].

C-R type modeling in which a huge amount of evaluated atomic data is included is needed for the evaluation of the experimental results. Precision of the A+M data evaluation is mandatory for obtaining confident results by C-R, CO and CO-R modeling; particularly they must be sufficiently precise in order to allow also a satisfactory plasma diagnostics. The quality of the input data is directly conditioning the validity of the obtained output [20]. 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 [25]. The first results obtained within our evaluated database project were reported in [15]. They include CTMC method evaluation of cross sections for electron – atom (ion) collision processes, mainly ionization [28] and excitation [13]. 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 [11]. 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 $E_{n+1} = E_n + E_{n+1}$ with $n, n+1$, $n+2$ indicating the successive ionization stages; the maximum value include always the number of equivalent electrons available for ionization in the external shell having the same quantum number (here 1) 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 [29], 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, 30, 31] in comparison with available evaluations [12]. 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 [32].

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, de-excitation 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 in our case have been obtained by integration according to a
Maxwellian distribution of the particles as a function of their temperature(s). Although this is a commonly followed procedure, cannot always be guaranteed as sufficient, and we are looking forward to consider two Maxwellians or other distributions for the rate calculation.


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 aforementioned EIRENE code, developed for the needs of the controlled fusion, could be adapted to the SPT conditions.

It is possible to obtain an extension the C-R type modeling to the transient plasmas case by keeping the time variations of the distribution functions df/dt in the LHS part of the Boltzmann equation [3]. 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 [4]. 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 for the industrial application of the latter. We are actually working toward this aim, also using adequate searching engines for bibliographic and numerical databases available in the Web, which are greatly helping this task [33]. The results obtained are compared as a function of the evaluated input data and also with the experiment [8, 9].

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Abbreviations and symbols

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<tr>
<th>Abbreviation</th>
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<tr>
<td>A+M</td>
<td>Atomic and Molecular</td>
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<tr>
<td>C-R</td>
<td>Collisional-Radiative</td>
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<td>C-RE</td>
<td>Collisional-Radiative Equilibrium</td>
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<td>CE</td>
<td>Coronal Equilibrium</td>
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<td>CO</td>
<td>Coronal</td>
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<td>CO-R</td>
<td>Coronal-Radiative</td>
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<td>CTMC</td>
<td>Classical Trajectory Monte Carlo</td>
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<td>EI</td>
<td>Electron collision Ionization</td>
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<td>E/M</td>
<td>Electric/Magnetic</td>
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<td>GS</td>
<td>Ground State</td>
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<tr>
<td>HS</td>
<td>Homogeneous and Stationary</td>
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<tr>
<td>Ip</td>
<td>Ionization Potential</td>
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<td>IAEA/AMD</td>
<td>International Atomic Energy Agency / A+M Data Unit, Vienna</td>
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<tr>
<td>KE</td>
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