Diagnostics of Argon Plasma Using Reliable Electron-Impact Excitation Cross Sections of Ar and Ar⁺

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Abstract: Comprehensive collisional radiative (CR) models have been developed for the diagnostic of argon plasma using Ar and Ar⁺ emission lines. The present CR models consist of 42 and 114 fine-structure levels of Ar and Ar⁺, respectively. Various populating and depopulating mechanisms are incorporated in the model. A complete set of electron-impact fine-structure resolved excitation cross-sections for different excited levels in Ar and Ar⁺ are used, which are obtained by employing relativistic distorted wave theory. Along with this, the electron-impact ionization, radiation trapping, diffusion, and three-body recombination are also considered. Further, to demonstrate the applicability of the present CR model, we applied it to characterize the Helicon-plasma utilizing the optical emission spectroscopy measurements. The key plasma parameters, such as electron density and electron temperature, are obtained using their measured Ar and Ar⁺ emission line intensities. Our results are in reasonable agreement with their anticipated estimates. The matching of our calculated intensities of the different Ar and Ar⁺ lines shows excellent agreement with the measured intensities at various powers.

Keywords: Ar and Ar⁺ collisional radiative model; relativistic distorted wave theory; electron-impact excitation cross-sections; argon plasma

1. Introduction

Non-invasive spectroscopic plasma diagnostic utilizing the optical emission spectroscopy (OES) measurements offers a powerful tool to study the deriving physics of plasma kinetics and gain knowledge of the production rate of different species present in the plasma. Since most of the laboratory plasmas significantly deviate from equilibrium conditions, giving rise to the need for the development of collisional radiative (CR) models to extract the information of key plasma parameters such as electron temperature ($T_e$) and electron density ($n_e$). We observe from the literature [1–3] that either the plasma or inert gases are often considered or added in small amounts in any plasma to characterize it. We find primarily that neutral argon gas is commonly used, and the intensities of its emission lines are measured and utilized to characterize various plasmas. In many high-density argon plasmas, along with the neutral emission lines, significant emissions from higher ionic states of argon, particularly from its first ionic state (i.e., Ar⁺), are also observed [4–7]. However, most of the CR models have used for the characterization of the plasma emission lines of the neutral argon, and not the lines of Ar⁺. Further, in such types of plasma, the electron-impact processes are the dominant processes [4]. The most important are the electron-impact excitations/de-excitations from the ground and the first excited manifold levels. It is due to the fact that the population densities of the ground and the first excited manifold are much higher as compared to other higher levels. Therefore, it would be interesting and important to carry out plasma diagnostics by incorporating reliable electron-impact excitation cross sections of the various fine-structure levels of the Ar and Ar⁺, along with other contributing processes in the CR models. To the best of our
knowledge, it would be the first study where we presented the calculation of the plasma parameter results obtained from CR models using both the atomic and ionic emission lines.

Further, plasma with low discharge pressure, high electron density, and low ion and electron temperatures are very useful and important for practical plasma sources [8]. From this point of view, Helicon Plasma (HP) sources, which use Helicon waves (HW) [8,9], are quite promising as these sources can produce plasmas with high \( n_e \approx 10^{13} \text{ cm}^{-3} \) and ionization efficiency [10,11] for input power smaller than a kW at radio frequency (rf) with some magneto-static field. Thus, HP sources have various applications, such as in fusion research [12,13], material processing [14–16], plasma propulsions and thrusters [17–19], plasma Wakefield accelerators [20], etc. In the world’s largest TOKAMAK fusion device, which is the International Thermonuclear Experimental Reactor (ITER), the neutral beam injection (NBI) ion sources are all inductively coupled plasma (ICP) type [21]. In ITER, ICP is generated inside the driver at rf 1 MHz and high powers up to 90 kW. However, it is beneficial to lower the value of the required rf power while maintaining the same plasma densities to obtain a more operational margin with accuracy and cost diminution. In this respect, various experimental studies [4,5,22] have shown that the Helicon heating is a very effective rf coupling technique to achieve plasma densities in Helicon discharges up to one order higher than the ICP discharges at the same rf power. With these developments, researchers worldwide have started investigating the HP source for the ITER nuclear beam system [11,21–26]. In most HP source [4] experiments, the working noble gas is argon; thus, it makes the diagnostics of argon plasma in these sources important and worth exploring.

In this regard, Soltani and Habibi [4] recently designed and developed an HP source for the NBI system. In their work, they identified and measured the intensities of the emission lines from both the Ar and Ar\(^+\). Thus, it would be beneficial from the application point of view to take up the diagnostic study of the HP of argon gas in light of their OES measurements [4]. Therefore, in this study, we aim to develop the detailed CR models of Ar and Ar\(^+\) for HP in light of the experimental study of Soltani and Habibi [4]. The present CR models of Ar and Ar\(^+\) have been developed by incorporating important atomic processes, such as electron-impact excitation/de-excitation, electron-impact ionization, radiation trapping, diffusion, and three-body recombination. It is worth mentioning that the electron-impact processes play a dominant role in plasma kinetics. Therefore, the cross-sections of these processes should be incorporated in a consistent manner [27]. However, the measurements and benchmark non-perturbative calculations are preferred, but these are available for selected transitions and also at a very limited energy range [28–33]. Mostly these considered transitions are unresolved fine-structured and thus are not suitable for developing a plasma model. However, in the plasma model, cross-sections for a large number of fine-structured transitions are required in the wide range of impact energy. Therefore, calculations from a reliable variant of the perturbative approach are often useful. In this regard, our group has reported electron-impact cross-sections for various atoms and ions using the relativistic distorted wave approach (RDW). In this RDW theory, the bound state atomic wave functions of Ar and Ar\(^+\) are at the Dirac–Fock level obtained from GRASP2K code [34]. Further, the projectile electron wave functions are computed numerically by solving the Dirac equations, which naturally incorporates relativistic effects such as spin-orbit interaction and jj coupling. Our calculated RDW cross-sections [35–38] are in reasonable agreement with the available respective measurements [29,30,39] and non-perturbative \( R \)-matrix calculations [31,32], though later better B-spline \( R \)-matrix calculations [33] for excitation from the ground state to some upper excited states were also reported, which reasonably compare with our RDW cross-sections. Additionally, our reported fine-structure resolved cross-sections have been successfully used in various plasma diagnostic studies [40–42]. All the incorporated electron-impact excitation cross-sections [35,37,38,43] of Ar and Ar\(^+\) in the present CR model are calculated from the RDW theory.

Further, we use a spectroscopic diagnostic approach which is based on comparing the peak intensity of emission lines obtained experimentally with the intensity obtained using our CR model. It would be worth mentioning here that in the present work, we
have not included the line-broadening in the CR model. We have obtained the plasma
patterns by employing the OES measurements of Soltani and Habibi [4] for Ar and Ar
emission lines. Moreover, we would like to add that Evdokimov et al. [7] have reported
their OES measurements of the magnetron discharge plasma considering Ar and Ar
lines and presented their modeling results. They primarily developed a CR model to characterize
their magnetron plasma, following our earlier reported CR model of Gangwar et al. [36]
for neutral Ar plasma, and also took only a single emission line of Ar (i.e., 488 nm). In
their approach, they have considered a single electron-impact transition in Ar+, which can
only be suitable for magnetron plasma. However, in the case of different plasmas, where
all other possible electron excitation processes that can occur from the ground and different
excited states of Ar+ and various other population transfer mechanisms are essential, this
approach might not be appropriate. All these processes (mentioned above) are considered
in the present CR models, and details are given in the next section.

2. Collisional Radiative Model

The particle balance equation of the present CR model, which is used to obtain
the population density \( n_j \) as a function of electron density and electron temperature
-corresponding to an excited level \( j \) of any considered system, can be written as:

\[
\sum_{i=1}^{x} k_{ij}(T_e)n_i n_e + \sum_{i>j} A_{ij}^{\text{eff}} n_i n_e n_j + n_e n_{j+1}(T_e) - \sum_{i=1}^{x} k_{ij}(T_e)n_j n_e - \\
\sum_{i<j} A_{ji}^{\text{eff}} n_j n_i n_{j+1}(T_e) - n_{j+1}(T_e) - k_{j+1}^{\text{diff}} = 0.
\]  

(1)

All the positive and negative terms, shown in Equation (1), represent the population
and depopulation channels for the excited level \( j \), respectively. The first and fourth terms
are the population transfer by electron collisional excitation and de-excitation, respectively,
wheras the second and fifth terms denote the radiative decays, respectively, from the
upper levels and to the lower levels. The third and sixth terms represent the three-body
recombination and ionization processes, respectively. The last term of the equation shows
the depopulation by diffusion of excited states through the chamber walls. In Equation (1),
\( x \) refers to the number of fine-structure (FS) levels taken in the CR model for the Ar/Ar+
. The selection of these levels is made such that, in each iteration, we added most of the
fine structure levels of an excited atomic state configuration until the populations of the
radiating excited levels no longer change significantly by further addition of the levels. In
Equation (1), \( n_i \) refers to the population density of fine-structure levels and \( n_{-1} \) is the ion
density of the first ionic state of the concerned atomic system, i.e., neutral Ar or Ar+.
Further, as mentioned above, the most important processes are the electron-impact excitations/de-
excitations from the ground and the first excited manifold levels in the considered plasma
conditions [4]. On the other hand, out of the two-body and three-body recombination
processes, we have included only the latter, as it is crucial for low-temperature plasma [44].
The charge exchange process is also not included in the present model, though this can
affect the charge-state balance of the low-temperature and low-density plasma. However,
we omit these processes for the sake of simplicity, noting that the purpose of this study is
to demonstrate the utility and accuracy of our recently calculated cross-sections. In fact,
the laser-produced plasma in [45–47] is at a lower temperature (\( T_e \approx 1 \text{ eV} \)) than the present
Ar plasma studies and significantly higher electron density (\( n_e \approx 1\times10^{16}–1\times10^{17} \text{ cm}^{-3} \), therefore
enhancing three-body recombination), yet they still found the significance of radiative
recombination. Further, the population transfer due to electron-impact excitation and
ionization in Equation (1), are included through rate-coefficients \( k_{ij} \) and \( k_{j+1}^{\text{diff}} \), respectively.
The rate-coefficients for electron-impact excitation can be evaluated using the following expression:

\[ k_{ij} = \sqrt{\frac{2}{m_e}} \int_{E_{ij}}^{\infty} \sigma_{ij}(E) \sqrt{E} f(E) dE. \]  

(2)

Here, \( m_e \) is the mass of electrons, \( E_{ij} \) denotes the excitation threshold energy of \( i \to j \) transition. \( \sigma_{ij}(E) \) is the electron-impact excitation cross-section for the \( i \to j \) transition. \( f(E) \) represents the Maxwellian electron energy distribution function (EEDF), which is related to the electron energy probability function through \( f(E) = E^{1/2} F(E) \). This choice of EEDF is made for the sake of simplicity and to be in consistency with the experimental plasma of Soltani and Habibi [4], where they have also taken the Maxwellian EEDF. Similarly, the rate-coefficient for the electron-impact ionization can be obtained by replacing the upper-state transition \((\sigma_{ij}^+\) \) in place of excitation cross-section. The rate-coefficients for reverse processes, e.g., electron-impact de-excitation and three-body recombination, are incorporated using the detailed balance principle [41,44,48]. Thus, the electron-impact de-excitation rate-coefficient \((k_{ji})\) can be written in terms of excitation cross-section as:

\[ k_{ji} = \frac{g_i}{g_j} \sqrt{\frac{2}{m_e}} \int_{E_{ij}}^{\infty} \sigma_{ij}(E) EF(E - E_{ij}) dE. \]  

(3)

Here, \( g_i \) and \( g_j \) are the statistical weights of the initial and final states, respectively. Further, the rate-coefficient for the three-body recombination \((k_{+i})\) can be obtained by using the Saha relation [44] as follows:

\[ k_{+i} = \frac{g_i}{2g_+} \left( \frac{\hbar^2}{2\pi m_e T_e} \right)^{3/2} \sqrt{\frac{2}{m_e}} \int_{E_{+i}}^{\infty} \sigma_{+i}(E) EF(E - E_{+i}) dE. \]  

(4)

Here, \( g_+ \) refers to the statistical weight of the singly ionized state of \( \text{Ar}/\text{Ar}^+ \) ground state and \( E_{+i} \) is the ionization threshold energy of \( \text{Ar}/\text{Ar}^+ \). In order to account for the radiation trapping effect, the transition probability \((A_{ij})\) in Equation (1) is replaced by effective transition probability \((A_{ij}^{eff})\), having relation \( A_{ij}^{eff} = A_{ij} \times A_{ji} \), for \( i \to j \) transition. Here, the escape factor \((\Lambda_{ij})\) is a function of gas temperature, dimensions of the plasma chamber [4], and population density of the lower level. We have used Mewe’s approximation [49] to calculate the escape factor in the present work as given below,

\[ \Lambda(K_{ij}\rho) = \frac{2 - e^{-K_{ij}\rho/1000}}{1 + K_{ij}\rho}. \]  

(5)

The required values of transition probabilities are taken from the NIST database [50]. The radius of the plasma chamber \((\rho)\) is taken as 2.4 cm, and the gas temperature is equal to room temperature. We solve first the particle balance equation (Equation (1)) to obtain the population densities of different excited fine-structure levels of \( \text{Ar} \). We have taken 42 fine-structure levels \((i.e., x = 42)\). These levels are the ground state \((3p^6)\) and the excited configurations \(3p^54s\) (with 4 FS), \(3p^54p\) (with 10 FS), \(3p^55d\) (with 12 FS), \(3p^55s\) (with 4 FS), \(3p^55p\) (with 10 FS) states, as well as the first ionization state of neutral argon, i.e., \(3p^5\). All of these states are given in Table 1 in the more familiar LS coupling notation, which we specifically derived for the purpose of the present work. Additionally, these were reported elsewhere in Paschen notation [36]. Table 1 also shows the electronic configuration of the different states and their energies [50]. All these considered levels are connected through different radiative and collisional transitions such as electron-impact excitation/de-excitation, ionization, diffusion, and three-body recombination, as shown in Figure 1. In the case of neutral argon, \( n_+ \) is the population density of \( \text{Ar}^+ \) in Equation (1). The electron-impact excitation rate-coefficients are obtained using Equation (2) by employing RDW electron-impact excitation cross-sections calculated by our group [36]. In order to evaluate
the ionization rate-coefficient, the required electron-impact ionization cross-sections are
taken from the experiments [51,52]. The rate-coefficients for the diffusion of metastable
states of Ar are taken from Kolts et al. [53]. It is worth mentioning that the escape factors
required in Equation (1) are computed for the transitions which decay to the ground state
of Ar, as only these have a significant contribution compared to other transitions.

Table 1. The energy levels of Ar, considered in the present CR model.

| Level No. | Level       | LS Coupling | Excitation Energy (eV) |
|-----------|-------------|-------------|------------------------|
| 1         | 3p6         | 1S0         | 0                      |
| 2         | 3p5(2P3/2)4s| 3P2         | 11.548                 |
| 3         | 3p5(2P1/2)4s| 3P1         | 11.623                 |
| 4         | 3p5(2P3/2)4s| 3P0         | 11.723                 |
| 5         | 3p5(2P1/2)4s| 1P1         | 11.828                 |
| 6         | 3p5(2P3/2)4p| 3D1         | 12.907                 |
| 7         | 3p5(2P1/2)4p| 3D2         | 13.076                 |
| 8         | 3p5(2P3/2)4p| 3P1         | 13.153                 |
| 9         | 3p5(2P1/2)4p| 3P0         | 13.172                 |
| 10        | 3p5(2P3/2)4p| 3S1         | 13.273                 |
| 11        | 3p5(2P3/2)4p| 1D2         | 13.302                 |
| 12        | 3p5(2P1/2)4p| 1P1         | 13.328                 |
| 13        | 3p5(2P3/2)3d| 1S0         | 13.480                 |
| 14        | 3p5(2P1/2)3d| 3P2         | 13.845                 |
| 15        | 3p5(2P3/2)3d| 3P1         | 13.863                 |
| 16        | 3p5(2P1/2)3d| 3P0         | 13.903                 |
| 17        | 3p5(2P3/2)3d| 3P4         | 13.979                 |
| 18        | 3p5(2P1/2)3d| 3P3         | 14.012                 |
| 19        | 3p5(2P3/2)3d| 3P2         | 14.063                 |
| 20        | 3p5(2P1/2)3d| 3P1         | 14.100                 |
| 21        | 3p5(2P3/2)3d| 3D1         | 14.153                 |
| 22        | 3p5(2P1/2)3d| 3D2         | 14.214                 |
| 23        | 3p5(2P3/2)3d| 1D2         | 14.234                 |
| 24        | 3p5(2P1/2)3d| 1P1         | 14.236                 |
| 25        | 3p5(2P3/2)5s| 1P1         | 14.303                 |
| 26        | 3p5(2P1/2)5s| 3P2         | 14.068                 |
| 27        | 3p5(2P3/2)5s| 3P1         | 14.090                 |
| 28        | 3p5(2P1/2)5s| 3P0         | 14.241                 |
| 29        | 3p5(2P3/2)5s| 1P1         | 14.255                 |
| 30        | 3p5(2P1/2)5s| 3D1         | 14.464                 |
| 31        | 3p5(2P3/2)5s| 3D2         | 14.500                 |
| 32        | 3p5(2P1/2)5s| 3P1         | 14.506                 |
| 33        | 3p5(2P3/2)5p| 3P2         | 14.525                 |
| 34        | 3p5(2P1/2)5p| 3P0         | 14.529                 |
| 35        | 3p5(2P3/2)5p| 3S1         | 14.575                 |
| 36        | 3p5(2P1/2)5p| 1P1         | 14.680                 |
| 37        | 3p5(2P3/2)5p| 1D2         | 14.687                 |
| 38        | 3p5(2P1/2)5p| 1S0         | 14.738                 |
| 39        | 3p5(2P3/2)5p| 2P3/2       | 15.760                 |
Figure 1. Energy level diagram of Ar along with the various collisional and radiative processes considered in the present CR model. The solid lines show excitations and de-excitation from the ground state of Ar. Wavy lines represent the radiative transitions. Here, FS stands for fine-structure.

In the next step, we solve the particle balance equation (Equation (1)) to obtain the population densities of different excited fine-structure levels of Ar\(^+\). We considered 114 fine-structure levels (i.e., \(x = 114\)). These are 3\(p^5\) (ground state) and excited 3\(s^2 3p^6\), 3\(p^4 3d\) (with 28 FS), 3\(p^4 4s\) (with 8 FS), 3\(p^4 4p\) (with 21 FS), 3\(p^4 5s\) (with 8 FS), 3\(p^4 4d\) (with 26 FS), 3\(p^4 5p\) (with 19 FS) states as well as first ionization level of Ar\(^+\) 3\(p^4\). These considered levels are listed in Table 2, along with their electronic configurations and associated energies [50]. The CR model framework outlining the interconnection between FS levels via different radiative and collisional transitions is shown in Figure 2, such as electron-impact excitation/de-excitation, electron-impact ionization, and three-body recombination. Here, \(n_+\) is the population density of Ar\(^{++}\) in Equation (1). The required RDW cross-sections of 114 fine-structure levels to evaluate electron-impact excitation rate-coefficient (Equation (2)) are taken from our previous work [38]. In addition, we have also calculated the additional RDW electron-impact excitation cross-sections of 3\(p^4 4s \rightarrow 3\(p^4 4p\) transitions because we did not report these cross-sections earlier [38]. These transitions play a vital role in the population and depopulation of the considered emission lines in the present work. The electron-impact ionization cross-sections of Ar\(^+\) needed to calculate the ionization rate-coefficients are taken from the reported measurements [54]. The required radiation trappings in Equation (1) are calculated for the transitions which decay to the ground state of Ar\(^+\), as we have found that only for these transitions, giving significant contributions.

Table 2. The energy levels of Ar\(^+\), considered in present CR model.

| Level No. | Level | LS Coupling | Excitation Energy (eV) | Level No. | Level | LS Coupling | Excitation Energy (eV) |
|-----------|-------|-------------|------------------------|-----------|-------|-------------|------------------------|
| 1         | 3\(p^5\) | \(^2p_{3/2}\) | 0                      | 59        | 3\(p^4(1S) 4p\) | \(^2p_{3/2}\) | 23.802                 |
| 2         | 3\(p^5\) | \(^2p_{1/2}\) | 0.177                  | 60        | 3\(p^4(1S) 4p\) | \(^2p_{1/2}\) | 23.846                 |
Table 2. Cont.

| Level No. | Level   | LS Coupling | Excitation Energy (eV) | Level No. | Level   | LS Coupling | Excitation Energy (eV) |
|-----------|---------|-------------|------------------------|-----------|---------|-------------|------------------------|
| 3         | 3s3p⁶   | 2S¹/²       | 13.480                 | 61        | 4⁵P₅/₂ | 22.515      |
| 4         | 4⁴D₇/₂  | 16.406      | 62                     | 5         | 3p⁴(3P)5s | 3P₁/₂      | 22.683 |
| 5         | 4⁴D₅/₂  | 16.425      | 63                     | 6         | 2⁵P₃/₂ | 22.700      |
| 6         | 4³D₃/₂  | 16.444      | 64                     | 7         | 2⁵P₁/₂ | 22.802      |
| 7         | 4⁴D₁/₂  | 16.457      | 65                     | 8         | 3p⁴(1D)5s | 2D₅/₂      | 24.284 |
| 8         | 4⁴F₉/₂  | 17.629      | 66                     | 9         | 3p⁴(1S)5s | 2D₃/₂      | 24.284 |
| 9         | 4⁴F₇/₂  | 17.695      | 67                     | 10        | 3p⁴(1S)5s | 5S¹/₂      | 26.665 |
| 11        | 3p⁴(3P)3d | 2P₁/₂       | 17.942                 | 12        | 4⁴F₃/₂ | 22.773      |
| 12        | 2P₁/₂   | 18.061      | 71                     | 13        | 2⁴P₃/₂ | 22.788      |
| 14        | 4⁴P₁/₂  | 18.254      | 72                     | 15        | 4⁴P₉/₂ | 22.949      |
| 16        | 4⁴P₅/₂  | 18.334      | 74                     | 17        | 4⁴F₇/₂ | 22.014      |
| 18        | 2⁴F₇/₂  | 18.496      | 75                     | 19        | 3p⁴(1P)4d | 2F₅/₂      | 23.070 |
| 20        | 2⁴F₅/₂  | 18.616      | 76                     | 21        | 2⁴F₃/₂ | 23.082      |
| 22        | 2⁴F₇/₂  | 18.776      | 77                     | 23        | 2⁴Gₙ/₂ | 23.070      |
| 24        | 2⁴F₃/₂  | 18.776      | 78                     | 25        | 2⁴F₅/₂ | 23.119      |
| 26        | 2⁴Gₙ/₂  | 19.116      | 79                     | 27        | 2⁴F₉/₂ | 23.162      |
| 28        | 2⁴F₇/₂  | 19.119      | 80                     | 29        | 2⁴F₇/₂ | 23.171      |
| 30        | 2⁴F₅/₂  | 20.246      | 81                     | 31        | 2⁴F₉/₂ | 23.258      |
| 32        | 2⁴F₇/₂  | 20.272      | 82                     | 33        | 3p⁴(1D)3d | 2F₅/₂      | 23.549 |
| 34        | 2⁴F₅/₂  | 20.367      | 83                     | 35        | 3p⁴(1D)3d | 2F₅/₂      | 23.549 |
| 36        | 2⁴F₇/₂  | 21.248      | 84                     | 37        | 3p⁴(1D)4d | 2F₅/₂      | 23.404 |
| 38        | 2⁴F₅/₂  | 21.262      | 85                     | 39        | 3p⁴(1S)3d | 2G₉/₂      | 23.438 |
| 40        | 2⁴F₇/₂  | 21.675      | 86                     | 41        | 3p⁴(1D)4d | 2G₉/₂      | 23.438 |
| 42        | 2⁴F₅/₂  | 22.266      | 87                     | 43        | 3p⁴(1S)3d | 2G₉/₂      | 23.438 |
| 44        | 2⁴F₇/₂  | 22.309      | 88                     | 45        | 3p⁴(1D)4d | 2G₉/₂      | 23.438 |
| 46        | 2⁴F₅/₂  | 22.825      | 89                     | 47        | 3p⁴(1S)3d | 2G₉/₂      | 23.438 |
| 48        | 2⁴F₇/₂  | 22.933      | 90                     | 49        | 3p⁴(1D)4d | 2G₉/₂      | 23.438 |
| 50        | 2⁴F₅/₂  | 23.487      | 91                     | 51        | 3p⁴(1S)3d | 2G₉/₂      | 23.438 |
| 52        | 2⁴F₅/₂  | 24.082      | 92                     | 53        | 3p⁴(1S)3d | 2G₉/₂      | 23.438 |
| 54        | 2⁴F₅/₂  | 24.096      | 93                     | 55        | 3p⁴(1S)3d | 2G₉/₂      | 23.438 |
| 56        | 3p⁴(3P)4p | 2D₅/₂       | 19.642                 | 58        | 3p⁴(1D)4p | 2D₅/₂      | 21.948 |
| 57        | 2D₅/₂   | 21.426      | 114                    | 59        | 3p⁴(1D)4p | 2D₅/₂      | 21.948 |

 Energy (eV)
Figure 2. Energy level diagram of Ar\(^+\) along with the various collisional and radiative processes considered in the present CR model. The solid lines show excitations and de-excitation from the ground state of Ar\(^+\) (\(^2\)P\(_{3/2}\)). Wavy lines represent the radiative transitions. Here FS stands for fine-structure.

We have solved the coupled particle balance equations (Equation (1)) numerically by utilizing the standard matrix inversion approach, similar to our previous CR models [36,42]. The evaluation of the rate-coefficients of the different processes has been performed as an intermediate step by providing the necessary cross-sections and the required transition probabilities. The needed transition probabilities are taken from the NIST database [50]. Simpson’s integration method is used to calculate the integrals corresponding to rate-coefficients in Equations (2)–(4). The required ground state population of the Ar atom in the CR model is evaluated from the standard gas law at the experimental conditions mentioned in Ref. [4], whereas in the case of the Ar\(^+\), following the charge neutrality condition, we take the ground state population density as equal to the electron density that we obtained from our CR model for argon. Since Equation (1) depends on the values of \(T_e\) and \(n_e\) for solving it, we assume a tentative wide grid of the different values of \(T_e\) and \(n_e\) separately. Then, by taking each value of \(T_e\) and different values of \(n_e\), we solve Equation (1) for each set of \((T_e, n_e)\). After solving the coupled particle balance equations, we obtain the population densities \(n_j\) of each considered fine-structure level as a function of different sets of \(T_e\) and \(n_e\) in the wide range of their values. Further, using these obtained population densities, we have calculated the intensities \((I)\) of the considered emission lines of Ar and Ar\(^+\) using the following equation,

\[
I_{ji} \propto n_j \frac{hc}{\lambda_{ji}} A_{ji}^{\text{eff}}.
\]  

Here, \(h\) and \(c\) represent Planck’s constant and the speed of light, respectively. \(\lambda_{ji}\) \((j \rightarrow i)\) stands for the emitted wavelength. Now, to obtain the plasma parameters, i.e., values of \(T_e\) and \(n_e\), which exactly correspond to the experimental plasma, the calculated CR model intensities need to be optimized or matched with their corresponding OES measured intensities [4]. A conventional way to achieve this optimization or matching is to calculate standard deviation parameters between the CR model simulated intensities and respective OES measured intensities.
To match our calculated model intensities for the considered emission lines of Ar and Ar\(^+\) with the experimentally measured intensities [4], we have first normalized these intensities individually by using the following relation,

\[
I_{\text{normalized}}^{j,\text{OES(\text{Model})}} = \frac{I_{j,\text{OES(\text{Model})}}}{\sum_{j=1}^{n'} I_{j,\text{OES(\text{Model})}}} \times 100. \tag{7}
\]

Here, \(I_{\text{normalized}}^{j,\text{OES}}\) and \(I_{\text{normalized}}^{j,\text{Model}}\) are the normalized intensities estimated from the OES measurements [4] and CR model, respectively. The \(n'\) represents the number of considered emission lines of Ar and Ar\(^+\), which is four (i.e., \(n' = 4\)), and the choice of these lines is justified in the next section. Further, we have employed the minimum scatter or least square approximation approach to check the best match of the calculated and experimental intensities [4] by obtaining the following deviation parameter to be minimum,

\[
\Delta = \sum_{j=1}^{n'} (I_{j,\text{OES}} - I_{j,\text{Model}})^2 T_{e n_e}. \tag{8}
\]

As mentioned above, the deviation parameter is a way to measure the agreement of the model intensities with the corresponding measurements [4]. Thus, its magnitude represents the least square difference between the normalized intensities obtained from the OES measurements [4] and the CR model. Therefore, the intensities obtained from the CR model for different sets of \(T_e\) and \(n_e\) are used in the above relation in order to find when the deviation parameter has the minimum value. Once this minimum condition with a particular combination of \((n_e\ and \ T_e)\) is achieved, the corresponding value of \(T_e\) and \(n_e\) are considered to represent the actual plasma parameters. This we will refer to as the obtained/extracted parameters \((T_e\ and \ n_e)\) for the specific case. This method has also been used successfully by us in our earlier work [36,41,42] to find the plasma parameters by matching the CR model and experimentally measured intensities.

3. Results and Discussion

Before discussing the present diagnostic results obtained from our CR models, we wish to briefly highlight the significance of using a CR Model in extracting the plasma parameters from OES measurements. The literature reveals that to extract \(n_e\) and \(T_e\) from OES measurements, the intensity of Ar-750.2 nm (for relative electron density estimation) and the ratio of Ar-811.5 and Ar-750.2 nm is often used to study the E-H-W mode transitions [55,56]. In the determination of line-ratio as a function of electron temperature, the upper radiating levels are assumed to be populated from the ground and metastable states through the electron-impact excitation process. This approach is very relevant; however, it should be noted that the line-ratio showed significant dependence only up to 3 eV [56]. Beyond that, the ratio is almost insensitive to the change in electron temperature [56]. Therefore, the scheme may not be suitable for helicon mode, where the presence of significant emission from Ar\(^+\) suggests a relatively higher electron temperature. A recent study reported a considerable discrepancy between the electron temperature values obtained from the Langmuir probe and the line-ratio [55] in Ar Helicon plasma. Therefore, applying CR models to extract plasma parameters from OES measurements is relevant.

As mentioned above, we employed the present CR models to study the experimental argon gas plasma reported by Soltani and Habibi [4]. They designed and developed a Helicon plasma source and showed the mode change from ICP to Helicon using OES measurements. In ICP mode, the magneto-static field \((B)\) is zero, whereas, in the case of Helicon mode, it varies from 350 to 750 G. Simultaneously, in both the modes, they recorded the spectra with changing rf power from 300–1000 W keeping the neutral background pressure at 0.7 mTorr. In their spectra, they have identified the specific emission lines of Ar in the range of 650–850 nm in the case of ICP mode, whereas in Helicon mode, they
observed emission lines representing Ar and Ar⁺ in the spectral range of 400–850 nm. In the present work, we have analyzed the data of OES measurements at \( B_r = 0 \) G (ICP mode) and \( B_r = 750 \) G (Helicon mode) of argon gas plasma provided by Soltani and Habibi [4]. It is worth mentioning that the 3p⁵4p FS levels of Ar and 3p⁴4p FS levels of Ar⁺ are quite crucial for the present plasma diagnostics. The reason is that Soltani and Habibi [4] identified emission lines in their spectra of Ar and Ar⁺, which corresponds to the decay from these levels. Here, we also selected the emission lines of Ar: 738.4, 751.4, 772.4, and 811.5 nm, and for Ar⁺: 434.8, 458.9, 480.6, and 487.9 nm that are originating from the 3p⁵4p and 3p⁴4p FS levels of Ar and Ar⁺, respectively. In addition, only these lines have substantial intensities, which are necessary for the calculation at all powers. Further, only these combinations of lines were sensitive with respect to the plasma parameters, i.e., electron temperature and electron density. Consequently, the evaluation of the population density distributions of such FS levels is the main goal of our CR models. We have used the Ar lines in the ICP mode and Ar and Ar⁺ lines in Helicon mode for plasma diagnostics. The Ar emission line at 772.4 nm is unresolved consisting of two lines at 772.38 nm and 772.42 nm. Therefore, while performing comparison, the theoretical model intensity is the summed value of these two lines.

Using our CR models of Ar and Ar⁺, we have calculated intensities of their emission lines as mentioned above through Equation (7) at several combinations of \( T_e \) and \( n_e \) in the wide range. These calculations are performed separately at various plasma operating powers of 300, 600, 800, 900, and 1000 W. Thereafter, the deviation parameters are calculated using the intensities of Ar lines in ICP mode and Ar as well as Ar⁺ lines in Helicon mode through Equation (8). Further, these deviation parameters are plotted as a function of \( T_e \) and \( n_e \) in each mode at different powers. Then, we analyzed them to ascertain the minima to obtain the electron temperature and electron density values, which eventually characterize the plasma. As an illustration, the variation of the deviation parameter with respect to the electron temperature (at fixed \( n_e \), which we found corresponds to the minimum deviation parameter) at 1000 W for both the modes are shown in Figure 3. From this figure, we can see that for \( B_r = 0 \) (ICP mode), the minimum value of deviation parameter is found at 4.0 eV with \( n_e = 1 \times 10^{17} \) m\(^{-3}\) and for \( B_r = 750 \) G (Helicon mode) at 8.13 eV with \( n_e = 1 \times 10^{19} \) m\(^{-3}\). Thus, these values of the \( T_e \) and \( n_e \), we can take as the extracted plasma parameters at 1000 W in different modes. It should be noted that in both cases, the minimum values of deviation parameters are less than 10 units. As explained earlier, the deviation parameter measures the agreement of the model intensities with the corresponding measurements [4]. Thus, 10 units can be said to as a good match of the intensities obtained from the CR model and experiment [4]. Similarly, we obtained the \( T_e \) and \( n_e \) values at all other powers for ICP and Helicon modes.

Figure 3. Deviation parameters as a function of \( T_e \) at the extracted value of \( n_e \) for argon plasma for (a) ICP mode (using Ar lines) and (b) Helicon mode (using Ar⁺ lines) at 1000 W.
We have shown all the obtained plasma parameters (i.e., $T_e$ and $n_e$) in both the modes in Table 3. From this table, one can observe that for the case of ICP mode, the electron temperature slightly increases from 3.39 to 4.00 eV with an increase of the power from 300 to 1000 W. Our values are quite close to the experimental value of electron temperature 4.0 eV for the ICP mode as reported by Soltani and Habibi [4]. We also found from our results that in the ICP mode, the electron density did not change much with respect to the variation in power and remained close to the value of $1 \times 10^{17}$ m$^{-3}$. Further, our model predicts a similar feature of the variation of electron temperature with respect to the power obtained using Ar lines in Helicon mode as was in ICP. We observe that the $T_e$ varied from 5.65 to 7.95 eV in the Helicon mode as the power increased from 300 to 1000 W. However, in contrast to ICP mode, the electron density is not constant but varies from the value of $3 \times 10^{18}$ to $1 \times 10^{19}$ m$^{-3}$ with the increase of power. We also observe from the present values that when the power is increased beyond 600 W in Helicon mode (with Ar lines), the electron temperature is abruptly increased. This suggests that up to the 600 W power, the argon plasma is in ICP mode, and thereafter, the plasma gets ionized, which is in confirmation with the observation of Soltani and Habibi [4]. Because of this reason, in Helicon mode, at lower powers, i.e., 300–600 W, Ar$^+$ lines are not observed experimentally, whereas at higher powers, i.e., 800–1000 W range, both Ar and Ar$^+$ spectral lines have been observed. Since both the spectral lines from Ar and Ar$^+$ represent the same Helicon plasma, the plasma parameters obtained from the CR models should ideally be the same for a particular power. We also find from Table 3 that our CR models using Ar and Ar$^+$ lines give quite close values of $T_e$ and $n_e$ in the range of 800–1000 W.

**Table 3.** Electron temperature ($T_e$) and electron density ($n_e$) for argon plasma at different modes and powers obtained using the present CR models.

| Power (W) | $T_e$ (eV) ICP Mode (Using Ar Lines) | $T_e$ (eV) Helicon Mode (Using Ar Lines) | $T_e$ (eV) Helicon Mode (Using Ar$^+$ lines) | $n_e$ (m$^{-3}$) Helicon Mode (Using Ar Lines) | $n_e$ (m$^{-3}$) Helicon Mode (Using Ar$^+$ Lines) |
|-----------|-------------------------------------|----------------------------------------|------------------------------------------|------------------------------------------|------------------------------------------|
| 300       | 3.39                                | 5.56                                   | -                                        | $3.00 \times 10^{18}$                   | -                                        |
| 600       | 3.59                                | 5.95                                   | -                                        | $4.48 \times 10^{18}$                   | -                                        |
| 800       | 3.61                                | 7.39                                   | 7.96                                     | $9.79 \times 10^{18}$                   | $9.70 \times 10^{18}$                   |
| 1000      | 4.00 (Exp. [4])                     | 7.95                                   | 8.13                                     | $1.00 \times 10^{19}$                   | $1.0 \times 10^{19}$                    |

Since Soltani and Habibi [4] have recorded the spectra with rf power variation in two different modes, it would be worth seeing how our plasma parameters behave with these variations. In Figure 4, we have shown the plot between the modes and obtained electron temperatures at 1000 W. There is a huge difference in the value of $T_e$ obtained using Ar lines in the ICP and Helicon modes. This may be due to the fact that in Helicon mode, the plasma gets more ionized as compared to the ICP mode. The appearance of Ar$^+$ emission lines in Helicon mode suggests that a higher fraction of electron-neutral collisions are resulting in excited argon ions rather than excited neutrals. It is only possible if the average electron energy is higher, as the threshold excitation energy is much higher for Ar ion levels than Ar-neutral excited states. The higher average energy also supports a higher ionization degree in Helicon mode. In this mode, the electron temperatures obtained using Ar and Ar$^+$ lines are nearly equal as expected because the plasma is the same. Further, for the Helicon mode, the variation of electron temperatures obtained using Ar lines with respect to powers is given in Figure 5. As shown in this figure, we observe a sudden jump in the temperature, which represents the transition from ICP to Helicon mode, as pointed out earlier.
Figure 4. Variation of obtained electron temperature ($T_e$) using the present CR model in different plasma operating modes at fixed power of 1000 W.

Figure 5. Variation of obtained electron temperatures ($T_e$) using present CR model at various powers for Helicon mode using Ar lines. The jump from point A to B reflects the transition of plasma operating mode.

Furthermore, in Figures 6–8, we have compared the normalized intensities obtained from the CR models (at the plasma parameters as given in Table 3) with the OES measurements [4]. In Figures 6 and 7, we find that the intensities of the considered transitions of Ar show good agreement with the experimental results [4] within the 4%. The largest differences are observed for the spectral lines 738.4 and 772.4 nm. Further, in Figure 8, we observe good agreement for all considered emission lines of Ar$^+$ with the experimental intensities [4]. The maximum difference was observed for 487.9 nm, which is 2%. Since the largest discrepancies are small in all three cases (Figures 6–8), we can say that predictions from our CR model are quite reasonable. Consequently, the obtained plasma parameters ($T_e$ and $n_e$) given in Table 3 at various powers are reliable.
As population densities are the main output of the CR models through which we have calculated the intensities in different modes at various powers, it would be thus worth seeing how the population densities vary with respect to the experimental values, which we have obtained through the measured intensities [4] using Equation (6). For this purpose,
in Figure 9, we plotted the ratio of upper-level population densities of the considered four emission lines in the present work, with respect to the wavelengths for the ICP and Helicon modes at 1000 W. To calculate this ratio, we divided all the upper-level population densities of the considered lines of Ar (738.4, 751.4, 772.4, and 811.5 nm) and Ar$^+$ (434.8, 458.9, 480.6, and 487.9 nm) with the density corresponding to the largest wavelength among these lines in each case, i.e., 811.5 and 487.9 nm. The population densities with which the ratios are obtained are $3.24 \times 10^{17}$ m$^{-3}$ in ICP mode and $1.97 \times 10^{17}$ m$^{-3}$ and $1.17 \times 10^{14}$ m$^{-3}$ in Helicon mode. These are shown in Figure 9a–c, respectively. On comparing our results with the values obtained from measurements [4], we find a reasonable agreement between both sets of results for the ICP and Helicon modes at 1000 W.

**Figure 9.** Comparison of upper-excited level population ratio of the considered emission lines with measurements [4] for argon plasma in (a) ICP mode (using Ar lines), (b) Helicon mode (using Ar lines), and (c) Helicon mode (using Ar$^+$ lines), at 1000 W.

### 4. Conclusions

In the present work, we have developed a fine-structure resolved CR model using Ar and Ar$^+$ emission lines for the diagnostics of the argon gas plasma. The required OES measurements are taken from the work of Soltani and Habibi [4]. Our models contain many possible populations transfer mechanisms among the fine-structure levels of Ar and Ar$^+$. The intensities of the different lines observed in the measurements for Ar and Ar$^+$ are calculated using the present CR models. These values are compared with the experimental intensities to extract the plasma parameters ($T_e$ and $n_e$) in ICP and Helicon modes at various powers. The extracted plasma parameters for both these modes at different powers seem to characterize the argon gas plasma of Soltani and Habibi [4] appropriately. We have also presented and compared the upper-level population density ratios with the measured values [4] and found good agreement. We believe our present extensive CR models consider
most population transfer mechanisms and can be easily extended to characterize any other Ar and Ar+ plasma.

**Author Contributions:** N.S. carried out the calculations, analysed the results, and took the lead in writing the manuscript. R.K.G. was involved in the discussions. R.S. supervised the project and arranged the essential funds to perform the research. All authors contributed to finalising the manuscript. All authors have read and agreed to the published version of the manuscript.

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