Diagnostics of plasmas with substantial concentrations of atomic oxygen

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An efficient method predicting the electron density N_e and temperature T_e in partially ionized plasmas with a fraction of atomic oxygen greater than about 10% of all heavy particles of the plasma is presented. The method is valid in plasmas where 10^{10} cm$^{-3} \lesssim N_t \lesssim 10^{16}$ cm^{-3} and 8000 K $\lesssim T_e \lesssim 15\,000$ K [$N_t = 2(N_a + N_e)$, and N_a is the density of the oxygen atoms].

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I. INTRODUCTION

Temperature and density of electrons in a partially ionized plasma which contains several neutral components (each consisting of one kind of atoms or molecules) can be determined from distribution of the internal states of the particles of one (any) of the components as long as the distribution is weakly affected by collisions of the particles of this component (called, hereafter, the "basic" component of the plasma) with the other heavy particles. Such a situation usually takes place when (1) the fraction of the particles of the basic component is substantial (that is, no less than about one-tenth of all heavy particles of the plasma), and (2) the ionization degree of the plasma is greater than about $10^{-3}-10^{-2}$. In most cases, these two requirements guarantee that the collisions of the plasma electrons with particles of the basic component will dominate the collisional processes shaping the distribution of the internal states of the particles. This includes partially ionized gases where atomic oxygen can be considered the basic component [1].

Knowledge of the electron density N_e and temperature T_e in plasmas where the fraction of the atomic oxygen is greater than 10% of all heavy particles and where the ionization degree is greater than 10^{-3} is an important issue because such plasmas are common in a wide range of applications. If the total density of the heavy particles in such plasmas is less than 10^{17} cm^{-3}, then the impact of inelastic collisions of the oxygen atoms with the other heavy particles on the distribution of the electronic energies of the atoms can be neglected when compared to the impact of the inelastic collisions of the electrons and photons with oxygen atoms. Then, the electron properties in the parent plasmas (where the atomic oxygen is the basic component) can be studied using a collisional-radiative model of a mixture of electrons, oxygen atoms, and their ions under the same physical conditions as the parent plasma under consideration.

General collisional-radiative models of various monatomic gases at $8000 \lesssim T_e \lesssim 15\,000$ K and $10^{10} \lesssim N_e \lesssim 10^{15}$ cm^{-3} were discussed in Refs. [1–3]. The present work is based on the results of Ref. [1] (called hereafter paper I) in which a nonlinear collisional-radiative model was formulated and solved for steady-state atomic oxygen plasma. (We refer the reader to paper I for a detailed discussion on the development, assumptions, and method of the solution of the model.) The model predicted the following properties of the plasma: (a) population of electrons, ions (both negative and positive), and excited atoms as a function of electron temperature and density, (b) intensities of spectral, continuum, and dielectronic recombination lines, and (c) the relaxation times for the electronic energy of the excited atoms.

A summary of important conclusions of paper I is given in Sec. II. These conclusions are the basis for formulation of the present kinetic model for steady-state, partially ionized atomic oxygen plasmas at $8000 \lesssim T_e \lesssim 15\,000$ K and $10^{10} \lesssim N_e \lesssim 10^{15}$ cm^{-3}. Such range of conditions exists in many plasmas with substantial concentration of atomic oxygen, including hypersonic flows (e.g., reentry problem), plasma processing (e.g., the oxygen discharge etching in plasma reactors), and environmental studies (e.g., dissociation of pollutants, such as the NO and CO molecules, in plasma phase).

The conclusions summarized in Sec. II are used in this work to simplify the complex, nonlinear collisional-radiative model of paper I, and to adopt the model to diagnostics of plasmas in situations where measurements of some spectral line intensities (or their ratios) in the plasmas are available. The simplification leads to a mathematically convenient and easy to interpret kinetic model of steady-state partially ionized oxygen plasmas at $8000 \lesssim T_e \lesssim 15\,000$ K and $10^{10} \lesssim N_e \lesssim 10^{15}$ cm^{-3}. The final formulation of the model consists of a set of two algebraic equations, the solution of which allows one to determine the electron density and temperature. [The accurate values of the coefficients occurring in the equations (the coefficients represent the rates of the collisional and radiative processes dominating the plasma) can easily be calculated]. In short, we present in this work an accurate and effective procedure on how to determine the electron density and temperature in plasmas under consideration if measurements of the plasma intensities (or intensity ratios) of a few spectral lines are available.

II. SUMMARY OF BASIC CONCLUSIONS OF PAPER I

(1) The populations of atoms excited to the electronic levels with $i=1$, 2, and 3 (i is the number of the level; $i=1$ denotes the ground level) almost always have Boltzmann distribution (see Fig. 6 of paper I).

(2) The populations of atoms excited to levels with $i \geq 8$ are always close to Saha equilibrium, and the presence of the strong $j \rightarrow i$ spectral lines (such as the lines produced in the $9 \rightarrow 1$, $9 \rightarrow 7$, and $8 \rightarrow 6$ spontaneous transitions) does not change this fact (see Fig. 10 of paper I).

(3) The bound-bound radiative transitions dominate the radiation produced by the plasma (see Figs. 21–23 of paper I).

(4) The radiation produced by the $5 \rightarrow 1$, $6 \rightarrow 4$, $7 \rightarrow 5$, $8 \rightarrow 6$, and $9 \rightarrow 7$ spontaneous transitions is weakly reabsorbed in the plasma (see Figs. 13–16 of paper I); in other words, the radiation escape factors κ_{ji} for these transitions are close to 1.

(5) The intensity of radiation produced by the $8 \rightarrow 6$ spontaneous transition is much weaker than the intensity of radiation produced by the $6 \rightarrow 4$ transition. Similarly, the intensity of radiation produced by the $9 \rightarrow 7$ transition is much weaker than the intensity produced by the $7 \rightarrow 5$ transition (see Figs. 18–20 of paper I). Therefore, the contribution of the $8 \rightarrow 6$ spontaneous transition to the population of the level $i=6$ can be neglected when compared to the contribution of the $6 \rightarrow 4$ transition. Similarly, the contribution of the $9 \rightarrow 7$ transition to the population of the level $i=7$ can be neglected when compared to the contribution of the $7 \rightarrow 5$ transition.

(6) The degree of ionization of the plasma is always greater than 10^{-2} (see Fig. 12 of paper I), that is, it is high enough to keep the energy distribution of free electrons close to the Maxwellian distribution [4,5].

(7) Since the radiation produced by the $5 \rightarrow 1$, $6 \rightarrow 4$, and $7 \rightarrow 5$ transitions is weakly reabsorbed in the plasma, the intensities of the radiation (or the intensities ratios) can be measured by standard spectroscopic techniques (see Figs. 13–16 of paper I). Subsequently, measured values of the intensities of these spectral lines can be used, in conjunction with the collisional-radiative model discussed below, to diagnose the density and temperature of the plasma electrons.

III. ATOMIC MODEL

The electronic energy structure assumed for atomic oxygen is shown in Fig. 1. The conclusions discussed in the preceding section suggest that the electronic levels of the atoms in the oxygen plasmas under consideration can be divided into three zones (see Fig. 1): (1) the "Boltzmann zone" that includes the $i=1$, 2, 3 levels which are always in Boltzmann equilibrium, (2) the "Saha zone" that includes the levels with $i \geq 8$ which are always in Saha equilibrium, and (3) the "active zone" that includes the

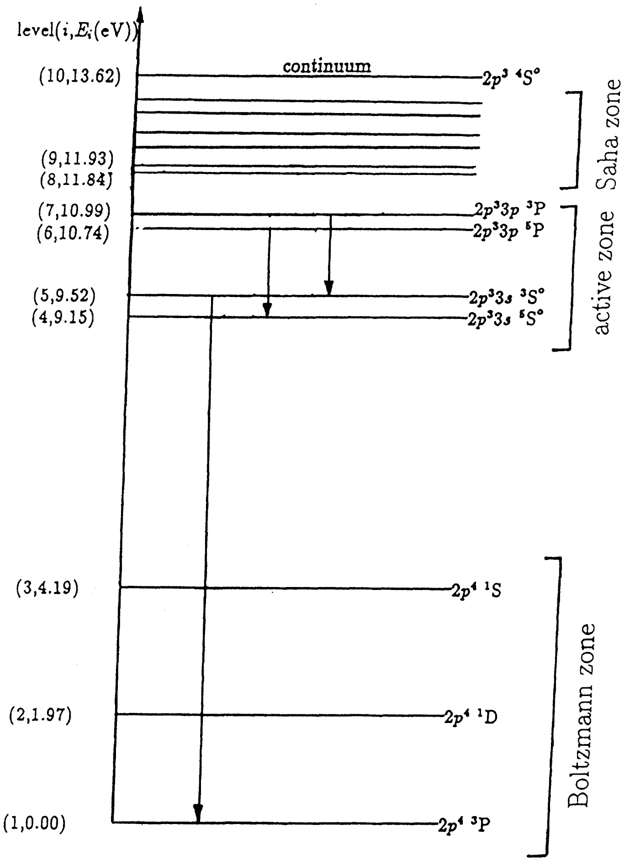

FIG. 1. Energy levels of the oxygen atom assumed in the present work. Energies E_i of the levels are measured with respect to the ground ($i=1$) state. The vertical arrows indicate the electric-dipole-allowed spontaneous transitions from the levels of the "active zone."

$i=4$, 5, 6, and 7 levels which can be in nonequilibrium (that is, they can be in neither Boltzmann nor Saha equilibrium). Therefore, it is sufficient to consider, in studies of thermal nonequilibrium in the oxygen plasmas of interest here, only the kinetics of the $i=4$, 5, 6, and 7 atomic levels.

IV. EXCITATION CROSS SECTIONS AND RATE COEFFICIENTS

A number of reliable measurements of cross sections for several electron-impact excitation transitions in oxygen atoms have been reported recently. The cross sections for the transitions for which experimental data are not available were calculated using theoretical approaches. The cross sections for the individual transitions were obtained as follows.

Measured cross sections [6,7] for the $1 \rightarrow 5$ and $1 \rightarrow 7$ electron-impact transitions were used in the present model. Comparison of the measured $1 \rightarrow 5$ cross section with other measurements [8] and with calculations of Julienne and Davis [9], Smith [10], and Rountree [11,12] gives an

TABLE I. Parameters in the rate coefficients (1) for electron-impact excitation of atomic oxygen. The designation of the inner shells $1s^22s^2$ is omitted in all electronic configurations. The numbers in brackets denote multiplicative powers of ten.

$i\to j$	Transition	B_{ij}	m_{ij}	α_{ij}
$1\to2$	$2p^4\,{}^3P\to2p^4\,{}^1D$	1.5840[−7]	−3.0304[−1]	1.688 70
$1\to3$	$2p^4\,{}^3P\to2p^4\,{}^1S$	2.5892[−9]	−1.2378[−1]	1.187 60
$1\to4$	$2p^4\,{}^3P\to2p^33s\,{}^5S^o$	5.8914[−13]	7.1849[−1]	1.028 30
$1\to5$	$2p^4\,{}^3P\to2p^33s\,{}^3S^o$	3.8914[−9]	4.4479[−2]	1.106 70
$1\to6$	$2p^4\,{}^3P\to2p^33p\,{}^5P$	2.5219[−10]	2.2426[−1]	1.037 30
$1\to7$	$2p^4\,{}^3P\to2p^33p\,{}^3P$	3.4232[−9]	1.0897[−1]	0.899 75
$2\to3$	$2p^4\,{}^1D\to2p^4\,{}^1S$	1.1277[−7]	−3.6665[−1]	1.559 40
$2\to4$	$2p^4\,{}^1D\to2p^33s\,{}^5S^o$	5.0539[−8]	−5.2367[−1]	1.091 40
$2\to5$	$2p^4\,{}^1D\to2p^33s\,{}^3S^o$	5.8260[−8]	−2.4703[−1]	1.036 10
$2\to6$	$2p^4\,{}^1D\to2p^33p\,{}^5P$	6.4902[−13]	7.3726[−1]	0.693 80
$2\to7$	$2p^4\,{}^1D\to2p^33p\,{}^3P$	9.3345[−11]	4.3550[−1]	0.982 71
$3\to4$	$2p^4\,{}^1S\to2p^33s\,{}^5S^o$	2.3063[−7]	−2.2747[−1]	1.008 60
$3\to5$	$2p^4\,{}^1S\to2p^33s\,{}^3S^o$	2.9544[−7]	−3.0360[−1]	1.054 10
$3\to6$	$2p^4\,{}^1S\to2p^33p\,{}^5P$	5.6000[−9]	8.2482[−2]	0.912 57
$3\to7$	$2p^4\,{}^1S\to2p^33p\,{}^3P$	3.3239[−10]	5.6002[−1]	0.958 91
$4\to5$	$2p^33s\,{}^5S^o\to2p^33s\,{}^3S^o$	7.8461[−5]	−5.217[−1]	1.364 50
$4\to6$	$2p^33s\,{}^5S^o\to2p^33p\,{}^5P$	1.7770[−5]	−2.2674[−1]	1.096 10
$4\to7$	$2p^33s\,{}^5S^o\to2p^33p\,{}^3P$	6.0075[−5]	−6.285[−1]	0.831 93
$5\to6$	$2p^33s\,{}^3S^o\to2p^33p\,{}^5P$	9.2673[−4]	−7.6627[−1]	0.890 41
$5\to7$	$2p^33s\,{}^3S^o\to2p^33p\,{}^3P$	1.7781[−5]	−2.1489[−1]	1.096 30
$6\to7$	$2p^33p\,{}^5P\to2p^33p\,{}^3P$	5.0462[−2]	−1.2677	1.285 70

acceptable agreement. The $1\to7$ electron-impact excitation cross section used in this work agrees well with experimental data of Zipf [6] and with the close-coupling calculations of Smith [10].

The electron-impact excitation cross section for the $2\to3$ transition was taken from the close-coupling calculations of Henry *et al.* [13]. This cross section agrees very well with the cross sections of Lan *et al.* [14] and Thomas and Nesbet [15], and it seems to be more accurate than the cross sections obtained earlier by Smith and co-workers [16,17] and Seaton [18].

All the $|\Delta S|=0$ and $|\Delta S|=1$ (S is the atomic electronic spin quantum number) transitions other than those discussed above are calculated using the Vainshtein approach (see Ref. [19] and paper I).

The electron-impact excitation cross sections for transitions with $|\Delta S|=2$ were calculated using the semiclassical impulse approximation [20].

The rate coefficients C_{ij} for the $i\to j$ electron-impact excitation transitions discussed above were obtained assuming that the plasma electrons have a Maxwellian distribution of energy. Subsequently, the rate coefficients were fitted (with an accuracy better than 1% when $8000\lesssim T_e\lesssim15\,000$ K) to the following expression:

$$C_{ij}=B_{ij}T_e^{m_{ij}}e^{-\alpha_{ij}\beta_{ij}}\ , \tag{1}$$

where $\beta_{ij}=(E_j-E_i)/kT_e$, and E_i and E_j are the electronic energies of the levels i and j, respectively. The parameters B_{ij}, α_{ij}, and m_{ij} are given in Table I.

Since we assume that the energy distribution of free electrons is a Maxwellian distribution, the principle of detailed balance is applied to calculate the rate coefficients R_{ji} for the electron-impact $j\to i$ deexcitation,

$$R_{ji}=\frac{g_i}{g_j}e^{-\beta_{ij}}C_{ij}\ , \tag{2}$$

where g_i and g_j are the degeneracies of the atomic levels i and j, respectively.

V. PROBABILITIES OF RADIATIVE TRANSITIONS

The magnetic dipole ($M1$) and electric quadrupole ($E2$) radiation is neglected in the present model, and the only radiative transitions taken into account are the spontaneous transitions produced by the electric dipole ($E1$) (see paper I). The Einstein coefficients for the electric dipole lines are available in a number of works [21–28]. The most recent of the coefficients were averaged and the averaged values (given in Table II) are used in the present work.

TABLE II. Electric dipole ($E1$) spectral lines of oxygen atoms taken into account in the present model. λ_{ji}, f_{ij}, and A_{ji} are the line wavelength, absorption oscillator strength, and Einstein's coefficient, respectively.

$j\to i$	Transition	λ_{ji} (Å)	f_{ij}	A_{ji} (s^{-1})
$5\to1$	$2p^3\,{}^3s\,{}^3S^o\to2p^4\,{}^3P$	1303.5	1.7×10^{-2}	1.99×10^8
$6\to4$	$2p^33p\,{}^5P\to.2p^33s\,{}^5S^o$	7773.4	9.5×10^{-1}	3.5×10^7
$7\to5$	$2p^33p\,{}^3P\to2p^33s\,{}^3S^o$	8446.5	9.8×10^{-1}	3.05×10^7

VI. COLLISIONAL-RADIATIVE MODEL

The rate equations for the production of atoms excited to an ith level of the "active zone" ($4 \le i \le 7$) in an optically thin mixture of electrons, oxygen atoms, and their singly charged ions can be written as

$$\frac{\partial N_i}{\partial t} = \sum_{k<i} N_e N_k C_{ki} + \sum_{j>i} N_e N_j R_{ji} + \sum_{j>i} N_j A_{ji} - N_i \left[\sum_{j>i} N_e C_{ij} + \sum_{k<i} N_e R_{ik} + \sum_{k<i} A_{ik} \right] , \quad (3)$$

where C_{ij} is the rate coefficient for the electron-impact excitation of an atom from a lower electronic level i to an upper level j, R_{ji} is the rate coefficient for the electron-impact deexcitation of an atom from the level j to the level i, and A_{ji} is the transition probability (the Einstein coefficient) for the $j \to i$ spontaneous transition.

The population of the $i=4$, 5, 6, and 7 levels can be found from a solution of Eq. (3) assuming that N_e, T_e, and N_1 are known (the populations N_2 and N_3 can be found from the Boltzmann distribution because the levels $i=2$ and $i=3$ belong to the Boltzmann zone). However, such a procedure is not complete because it does not take into account the conservation of plasma charges [both free (ions and electrons) and those bound in atoms (an atom consists of an electron plus a singly charged ion)]—see discussion in Ref. [3] and in paper I. Therefore, Eq. (3) has to be coupled with the following equation for the (invariant) particle density (N_t) of the free and bound charges in the plasma:

$$N_t = 2N_a + N_+ + N_e \simeq 2(N_a + N_e) , \quad (4)$$

where $N_a = \sum_i N_i$ is the total density of the plasma atoms, N_+ and N_e are densities of positive ions and electrons, respectively, and where we neglected the presence of negative ions (see discussions in Ref. [3] and paper I) and assumed that the plasma is electrically neutral ($N_+ = N_e$). Under the physical conditions discussed in this work, most of the positive atomic ions are in the ground electronic states, and the presence of multiply charged ions can be ignored.

In oxygen plasmas considered here $N_a \simeq 1.1 N_1$ (see Fig. 6 of paper I). Therefore, Eq. (4) can be rewritten as

$$N_t = 2.2 N_1 + 2N_e , \quad (5)$$

and the populations of the atomic levels of the Boltzmann zone can be given as

$$N_1 = \frac{N_t + 2N_e}{2.2} , \quad (6)$$

$$N_2 = \tfrac{5}{9} N_1 e^{-(E_2 - E_1)/kT_e} , \quad (7)$$

and

$$N_3 = \tfrac{1}{9} N_1 e^{-(E_3 - E_1)/kT_e} . \quad (8)$$

If T_e, N_e, and N_t are known, then steady-state solutions of Eqs. (3) and (5)–(8) yield the steady-state populations N_4, N_5, N_6, and N_7 (of the levels $i=4$, 5, 6, and 7, respectively). The populations are in good agreement with the corresponding results obtained from the full collisional-radiative model of paper I (see Figs. 9 and 12 of paper I). Thus, the set of Eqs. (3) and (5)–(8) is an accurate representation of the kinetic processes in the steady-state oxygen plasmas considered in this work. Therefore, we use these equations below in order to determine the plasma electron density and temperature.

A solution of Eqs. (3) and (5)–(8) allows one to find four unknowns (N_4, N_5, N_6, and N_7) if N_e, T_e, and N_t are known. However, one can assume that two out of the four (N_4, N_5, N_6, and N_7) populations are known so that the plasma electron density N_e and temperature T_e can be treated as unknowns. The most natural (and convenient) choice of these two populations are the populations of the levels with $i=6$ and 7, because both levels produce strong spectral lines intensities of which can be measured directly or from some intensity ratios involving the lines. (The transition probabilities for the $6 \to 4$ and $7 \to 5$ transitions are $A_{64} = 3.5 \times 10^7$ s^{-1} and $A_{75} = 3.05 \times 10^7$ s^{-1}, respectively). The spectral line intensity I_{ji} of a $j \to i$ transition in a homogeneous plasma which is optically thin for the radiation produced in the transition is defined here as the line power radiated in all directions from a unit plasma volume,

$$I_{ji} = N_j A_{ji} h \nu_{ji} , \quad (9)$$

where, as before, A_{ji} is the transition probability, and ν_{ji} is the frequency of the emitted radiation. Thus, the populations N_6 and N_7 are related to the intensities I_{64} and I_{75} in the following way:

$$N_6 = \frac{I_{64}}{A_{64} h \nu_{64}} \quad \text{and} \quad N_7 = \frac{I_{75}}{A_{75} h \nu_{75}} . \quad (10)$$

Taking the above into account, Eq. (3) can be rewritten in the case of steady-state oxygen plasma as

$$\alpha_1 + N_5 \alpha_2 + \alpha_3 + \alpha_4 - N_4 \alpha_5 + \frac{\alpha_6}{N_e} = 0 , \quad (11)$$

$$\alpha_7 + N_4 \alpha_8 + \alpha_9 + \alpha_{10} + \frac{\alpha_{11}}{N_e} - N_5 \left[\alpha_{12} + \frac{\alpha_{13}}{N_e} \right] = 0 , \quad (12)$$

$$\alpha_{14} + N_4 \alpha_{15} + N_5 \alpha_{16} + \alpha_{17} - \alpha_{18} - \frac{\alpha_6}{N_e} = 0 , \quad (13)$$

and

$$\alpha_{20} + N_4 \alpha_{21}' + N_5 \alpha_{22} + \alpha_{23} - \alpha_{24} - \frac{\alpha_{25}}{N_e} = 0 , \quad (14)$$

where the coefficients $a_1 - a_{25}$ are given in the Appendix.

Equations (11)–(14) are the basic equations of the present model. The solution of the equations allows one to determine N_e and T_e (and N_4 and N_5) in atomic oxygen plasmas considered here if the density N_t and the intensities I_{64} and I_{75} are known. The solution of Eqs. (11)–(14) can be simplified by reducing them to the following two equations with two (N_e and T_e) unknowns:

$$(\alpha_5\alpha_{22}+\alpha_{21}\alpha_2)\left[B+\frac{\alpha_{11}}{N_e}\right]+\left[A+\frac{\alpha_{16}}{N_e}\right]\left[\alpha_{21}\left[\alpha_{12}+\frac{\alpha_{13}}{N_e}\right]+\alpha_8\alpha_{22}\right]+\left[D-\frac{\alpha_{11}}{N_e}\right]\left[\alpha_5\left[\alpha_{12}+\frac{\alpha_{13}}{N_e}\right]-\alpha_8\alpha_2\right]=0\ , \quad (15)$$

and

$$(\alpha_5\alpha_{22}+\alpha_{21}\alpha_2)\left[C-\frac{\alpha_6}{N_e}\right]+\left[A+\frac{\alpha_{16}}{N_e}\right](\alpha_{15}\alpha_{22}-\alpha_{21}\alpha_{16})-\left[D-\frac{\alpha_{11}}{N_e}\right](\alpha_{15}\alpha_2+\alpha_5\alpha_{16})+\frac{\alpha_5\alpha_{22}\alpha_{16}}{\alpha_2 N_e}(\alpha_{16}-\alpha_6)=0\ , \quad (16)$$

where

$$A=\alpha_1+\alpha_3+\alpha_4,\quad B=\alpha_7+\alpha_9+\alpha_{10}\ , \quad (17)$$

and

$$C=\alpha_{14}+\alpha_{17}-\alpha_{18},\quad D=\alpha_{20}+\alpha_{23}-\alpha_{24}\ . \quad (18)$$

The particle density N_t occurring in Eqs. (15) and (16) can be obtained from the relationship (4). The density N_a of the oxygen atoms in the parent gas (that is, a gas which contains a substantial concentration of atomic oxygen and which is to be diagnosed using the present collisional-radiative model) depends on the fraction of the atomic oxygen in the gas. This fraction is required to be not less than about 10% of all heavy particles of the parent gas. In most steady-state plasmas considered here, the fraction can be estimated from the law of mass action.

The experimental values of the intensities of the radiation produced by the $6\rightarrow4$ and $7\rightarrow5$ spontaneous transitions in oxygen atoms can be obtained from spectroscopic measurements. [The intensities I_{64} and I_{75} occurring in Eqs. (15) and (16) are, in contrary to the corresponding measured values, the intensities of radiation emitted isotropically in all directions, that is, into a solid angle 4π].

VII. APPLICATION OF THE MODEL TO NON-STEADY-STATE PLASMAS

When the population of an atomic level departs (as a result of a small change of plasma parameters) from its steady-state value, then some time (the relaxation time) is needed to reestablish steady-state population of the level. The relaxation times for each atomic level can be calculated in the way described in paper I. In the oxygen plasmas discussed here, the relaxation times for the excited atoms are very short, so that the levels $i=4$, 5, 6, and 7 of the "active zone" are, in fact, in steady state in transient plasmas where the time dependence of the plasma parameters is not very strong, and where the concentration of the atomic oxygen is substantial. Due to lack of radiative coupling (but presence of strong collisional coupling) between the two lowest metastable levels ($i=2,3$) and the ground state, the relaxation times for these levels in the plasma are also short (much shorter, for example, than the corresponding relaxation times in atomic hydrogen plasmas). Thus, the time-dependent rate equations describing populations of the atomic levels in the non-steady-state plasmas of interest here can be solved assuming that all the excited levels are in steady state, while the atomic ground level is in a transient state. Subsequently, the kinetics of the excited levels can be solved using the steady-state model of the present work, while the kinetics of the ground level must be determined from some additional, time-dependent conditions imposed on the plasma.

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APPENDIX

Coefficients occurring in Eqs. (11)–(14):

$$\alpha_1=N_1C_{14}+N_2C_{24}+N_3C_{34}\ , \quad (A1)$$

$$\alpha_2=R_{54}\ , \quad (A2)$$

$$\alpha_3=\frac{I_{64}R_{64}}{A_{64}h\nu_{64}}\ , \quad (A3)$$

$$\alpha_4=\frac{I_{75}R_{74}}{A_{75}h\nu_{75}}\ , \quad (A4)$$

$$\alpha_5=C_{45}+C_{46}+C_{47}+R_{43}+R_{42}+R_{41}\ , \quad (A5)$$

$$\alpha_6=\frac{I_{64}A_{64}}{A_{64}h\nu_{64}}\ , \quad (A6)$$

$$\alpha_7=N_1C_{15}+N_2C_{25}+N_3C_{35}\ , \quad (A7)$$

$$\alpha_8=C_{45}\ , \quad (A8)$$

$$\alpha_9=\frac{I_{64}R_{65}}{A_{64}h\nu_{64}}\ , \quad (A9)$$

$$\alpha_{10}=\frac{I_{75}}{A_{75}h\nu_{75}}R_{75}\ , \quad (A10)$$

$$\alpha_{11}=\frac{I_{75}}{A_{75}h\nu_{75}}A_{75}\ , \quad (A11)$$

$$\alpha_{12}=C_{56}+C_{57}+R_{54}+R_{53}+R_{52}+R_{51}\ , \quad (A12)$$

$$\alpha_{13}=A_{51}\ , \quad (A13)$$

$$\alpha_{14}=N_1C_{16}+N_2C_{26}+N_3C_{36}\ , \quad (A14)$$

$$\alpha_{15}=C_{46}\ , \quad (A15)$$

$$\alpha_{16}=C_{56} , \tag{A16}$$

$$\alpha_{17}=\frac{I_{75}R_{76}}{A_{75}h\nu_{75}} , \tag{A17}$$

$$\alpha_{18}=\frac{I_{64}}{A_{64}h\nu_{64}}(C_{67}+R_{65}+R_{64}+R_{63}+R_{62}+R_{61}) , \tag{A18}$$

$$\alpha_{19}=\alpha_{6} , \tag{A19}$$

$$\alpha_{20}=N_1C_{17}+N_2C_{27}+N_3C_{37} , \tag{A20}$$

$$\alpha_{21}=C_{47} , \tag{A21}$$

$$\alpha_{22}=C_{57} , \tag{A22}$$

$$\alpha_{23}=\frac{I_{64}C_{67}}{A_{64}h\nu_{64}} , \tag{A23}$$

$$\alpha_{24}=\frac{I_{75}}{A_{75}h\nu_{75}}(R_{76}+R_{75}+R_{74}+R_{73}+R_{72}+R_{71}) , \tag{A24}$$

and

$$\alpha_{25}=\alpha_{11} , \tag{A25}$$

where the rate coefficients C_{ij} and R_{ji} [Eqs. (1) and (2), respectively], are functions of the electron temperature T_e.

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