

Collisional-Radiative Model for Non-Equilibrium Cesium Plasma

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非平衡セシウムプラズマに対する衝突—放射モデル

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Abstract

Macroscopic natures of the non-equilibrium cesium plasma are described by a collisional-radiative model, in which the excited atoms play a substantial role. Up to this time, only the resonance state 6P has been considered for the excited state in the collisional-radiative model because the probability of the excitation to the resonance state is very high in comparison with that to the other states. In the present work, the collisional-radiative model including seven excited states is considered and it is shown how the inclusion of the excited state other than the resonance one leads to a satisfactory description for the plasma properties. It is confirmed that the number of the excited state should be increased as the temperature and the pressure decrease.

§1. Introduction

Since cesium atom has the lowest ionization energy and large excitation cross sections, a cesium plasma with high electron density, high electrical conductivity and high efficiency of light emission can be produced easily. There are many engineering applications utilizing alkali metal vapor plasma, for example, MHD generators using thermally ionized gases, thermionic converters using surface ionization, ion propulsion and high pressure discharge lamps. In MHD generators, a nonequilibrium plasma produced by applying external electric field to the thermally ionized plasma has been used in order to increase the electrical conductivity.¹⁻³⁾ Here, the nonequilibrium plasma is defined as the one in which the electron temperature is always higher than those of the neutral atom and ion. In thermionic converters, a great deal of interest has been directed on the arc mode operation, in which the space charge is neutralized.⁴⁾ In discharge lamps, both thermal and electric field ionizations play the part for generating charged particles. The plasma in most alkali vapor plasma devices are regarded as the nonequilibrium plasma. Macroscopic quantities of such plasmas described by collisional and radiative processes.

The simplest model for describing plasma properties is the model of local thermal equilibrium,⁵⁾ in which details of elementary processes are not concerned. This model may be used effectively for the study of arc discharge at a relatively high pressure. For a very low density, the corona model, in which the dominant processes are collisional ionization and recombination, or collisional excitation and radiation is valid. For the intermediate region between the region in which the corona model is valid and the region in which the local thermal equilibrium model is valid, the collisional-radiative model has been proposed by Bates.⁶⁾ Inoue et al.⁷⁾ have applied the collisional-radiative model to cesium plasma. They considered the steady

state in which the elementary processes such as collisional ionization, excitation, recombination, radiation and diffusion are balanced as a whole, and the momentum and the energy are conserved.

In cesium plasma, it has been known that not the direct ionization by collision of electron with the atom in the ground state, but the stepwise ionization by collision of electron with the excited atom is a dominant process for the generation of the charged particle.⁸⁾ When an alternating electric field with small amplitude is applied to a d. c discharge plasma, the plasma impedance defined as the ratio of the a. c voltage to the a. c current can be measured. The expression for the plasma impedance was derived from the macroscopic transport equations representing the continuity of the particle and the conservation of the momentum and the energy for all constituents and thus it was confirmed that the elementary processes involving the excited atom contribute significantly to the macroscopic properties of the plasma.⁹⁾ Noting such an important role of the excited atom, a new technique for producing cesium plasma has been developed, in which the excited atoms are produced by irradiation of the resonance radiation from the outside and then ionized through the collision process.¹⁰⁾

Though it has been recognized that in a cesium plasma the excited atoms play an important role, the excited atoms except ones of the resonance state 6P have been neglected in the collisional-radiative model,⁷⁾ because the excitation cross section to 6P state is larger than those to the other excited states. However, there remains a question whether the excited atom in the other states affects the macroscopic properties of the plasma or not. In this paper, we will consider the collisional-radiative model including seven excited states and show how the inclusion of the excited state other the resonance one leads to a satisfactory description for the plasma properties depending on the pressure and the temperature. It is confirmed that the number of the excited state should be increased as the pressure and the temperature decrease.

§2. Collisional and Radiative Processes in Nonequilibrium Plasma

It is assumed that cesium atom is in one of nine energy states, that is, the ground state 6S, the excited state 6P, 5D, 7S, 7P, 6D, 8S and 8P and the free state. We indicate the excited states by the symbols 1, 2, 3, ……7 starting from the lowest excited state, while the ground state by g and the free state by i. These energy levels are shown in Fig. 1. The figures in the parentheses refer to the above symbols. The density of the atom in an energy state or that of the electron depends on the production and loss rates which are governed by the collisional and radiative processes. Therefore, the occurrence rate of the collisional and radiative processes per unit time, i. e., the collision frequency or the recombination coefficient must be expressed as a function of the temperature and the density.

Now, we will consider the various collisional and radiative processes in the nonequilibrium cesium plasma and estimate the collision frequency or the recombination coefficient.

2.1 Ionization

Both the direct ionization by electron collision with the atom in the ground state and

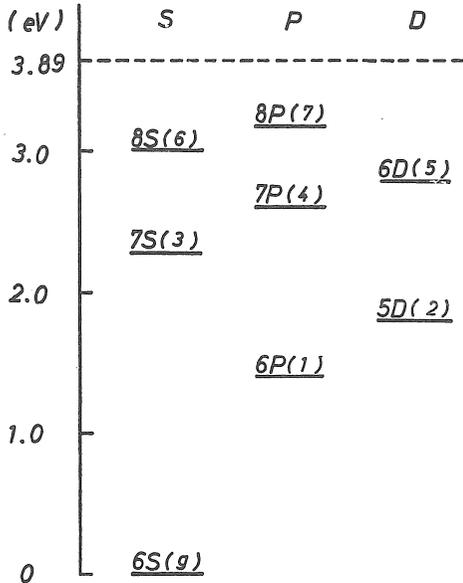


Fig.1 Energy state diagram.

the stepwise ionization by electron collision with the excited atom are considered as the mechanism for the electron-ion pair production. The number of the electron-ion pair produced by the electron collision with the atom in the ground state per unit time, that is, the corresponding ionization frequency ν_{egi} is given as follows :

$$\nu_{egi} = N_g \int_{W_i}^{\infty} f_e(W_e) \sigma_{egi}(W_e) v_e dW_e, \quad (1)$$

where σ_{egi} is the ionization cross section by the electron collision with the ground state atom, N_g the density of the ground state atom, $f_e(W_e)$ the electron energy distribution function, v_e the electron thermal velocity, W_e the

electron energy and W_i the ionization energy of the ground state atom. The ionization frequency for the stepwise ionization ν_{eji} is given by

$$\nu_{eji} = N_j \int_{W_i - W_j}^{\infty} f_e(W_e) \sigma_{eji}(W_e) v_e dW_e, \quad (2)$$

where N_j is the density of the j th excited atom, σ_{eji} the stepwise ionization cross section by electron collision with the j th excited atom, W_j the excitation energy corresponding to the j th state and the subscript j refers to a numeral ' from 1 to 7. As to the ionization process, there is another process in which the molecular ion is produced by collision between the excited atoms. Since the cross section of this process is less than 0.2 \AA^2 ,¹¹⁾ this process can be neglected in an ordinary cesium plasma.

2.2 Excitation

The excitation frequency to the j th state by the electron collision with the neutral atom is given by

$$\nu_{egj} = N_g \int_{W_j}^{\infty} f_e(W_e) \sigma_{egj}(W_e) v_e dW_e, \quad (3)$$

where σ_{egj} is the excitation cross section to the j th state.

In order to estimate these collision frequencies, it is necessary to know the value of the cross section as a function of the electron energy. However, the knowledge of these cross sections is lacking except for those of the cross sections of the direct ionization,¹²⁾ the excitation¹³⁾ to the resonance state 6P and the stepwise ionization¹⁴⁾ of the 6P state. Therefore, we have calculated the ionization and excitation cross sections using the model proposed by Gryzinski¹⁵⁾ who estimated the cross section by treating the inelastic collision as the classical collision between a free electron and the outermost bound electron of the atom. The expression for the cross section for

an optically allowed transition given by Gryzinski is expressed as follows :

$$\sigma_{egj}(W_e) = \sigma(W_j, W_i, W_e) - \sigma(W_{j+1}, W_i, W_e), \quad (4)$$

where
$$\sigma(W_j, W_i, W_e) = \frac{\sigma_0}{W_i^2} \frac{W_i}{W_e} \left(\frac{W_e}{W_e + W_i} \right)^{3/2} \times$$

$$\left[\frac{W_j}{W_i} + \frac{2}{3} \left(1 - \frac{W_j}{W_e} \right) \ln \left\{ 2.7 + \left(\frac{W_e - W_j}{W_i} \right)^{1/2} \right\} \right] \times$$

$$\left(1 - \frac{W_j}{W_e} \right) \frac{1 + W_i / (W_i + W_j)}{1}, \quad (5)$$

and
$$\sigma_0 = 6.56 \times 10^{-14} \text{ (eV}^2 \text{ cm}^2 \text{)}, \quad (6)$$

The collisional excitation cross section for an optically forbidden transition is given by

$$\sigma_{egj}(W_e) = \frac{\sigma_0}{W_j^2} \frac{W_{j+1} - W_j}{W_j} g_{\text{exch}} \quad (7)$$

Where
$$g_{\text{exch}} = \begin{cases} \frac{W_j^2}{(W_e + W_i)(W_e + W_i - W_j)} \frac{W_j(W_e - W_j)}{W_i(W_{j+1} - W_j)}, & W_e < W_{j+1}, \\ \frac{W_j^2}{(W_e + W_i)(W_e + W_i - W_j)} \frac{W_j}{W_e + W_i - W_{j+1}}, & W_e > W_{j+1}, \end{cases} \quad (8)$$

The ionization cross section for Gryzinski model is given by

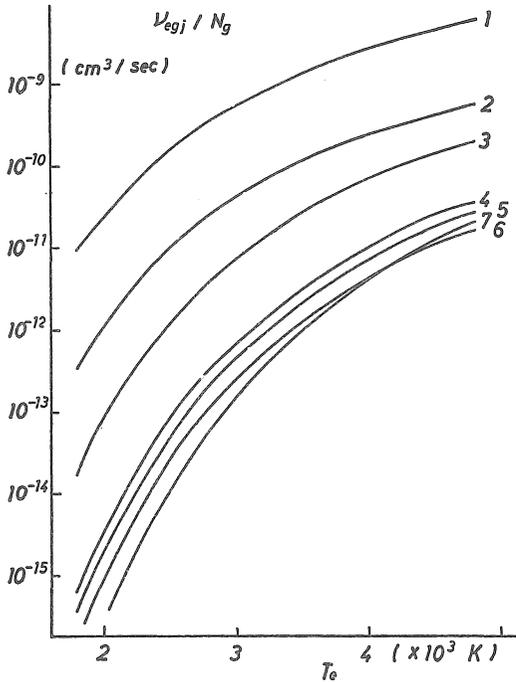


Fig. 2 Collisional excitation probabilities calculated by Gryzinski model.

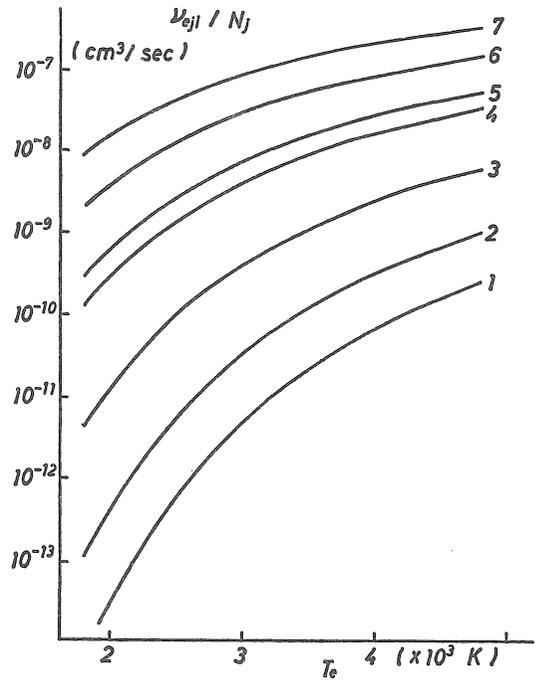


Fig. 3 Collisional ionization probabilities calculated by Gryzinski model.

$$\sigma_{eji}(W_e) = \sigma(W_i - W_j, W_i - W_j, W_e) \quad (9)$$

The collisional excitation probabilities calculated using the Gryzinski model, that is, the excitation frequencies divided by the density of the ground state atom ν_{egj}/N_g are shown in Fig. 2 as a function of the electron temperature, where a Maxwell-Boltzmann distribution for the electrons is assumed. Moreover, the collisional ionization probabilities calculated using the Gryzinski model, that is, the ionization frequencies divided by the population of the excited state ν_{eji}/N_j are shown in Fig. 3.

2.3 Recombination

In cesium plasma, the three body, the radiative and the dissociative recombination processes are the possible recombination processes. Although the dissociative recombination coefficient is not smaller than those of other processes, it is insignificant because the density of the molecular ion is smaller than that of the atomic ion as discussed previously.

The three body recombination coefficient can be expressed in terms of the ionization cross section using the principle of detailed balance. That is to say, in thermal equilibrium the rate of the ionization by the electron collision with the atom in an energy state is equal to the rate of the recombination to the energy state by three body collision between two electrons and ion :

$$N_e \nu_{eji} = N_e^2 N_i \beta_j, \quad (10)$$

where β_j is the three body recombination coefficient. In thermal equilibrium, the population of the excited state is given by the Boltzmann distribution :

$$N_j = N_g \frac{g_j}{g_g} \exp(-W_j/kT_e), \quad (11)$$

and the electron density is given by the Saha equation :

$$\frac{N_e N_i}{N_g} = \left(\frac{2\pi m_e k T_e}{h^2} \right)^{3/2} \exp(-W_i/kT_e), \quad (12)$$

where g_j and g_g are the statistical weight of the excited state j and the ground state g respectively, k the Boltzmann constant, h the planck constant and m_e the electron mass. Substituting eqs. (11) and (12) into eq.(10), we obtain the three body recombination coefficient as follows:

$$\beta_j = \frac{\nu_{eji}}{N_j} \frac{g_j}{g_g} \left(\frac{h^2}{2\pi m_e k T_e} \right)^{3/2} \exp\left(\frac{W_i - W_j}{k T_e}\right), \quad (13)$$

The above recombination coefficient was estimated for the case of thermal equilibrium, but eq. (13) is valid for the nonequilibrium system because the recombination cross section is independent of whether the system is in thermal equilibrium or not.

The radiative recombination coefficients calculated by Norcross and Stone¹⁶⁾ using the quantum defect method are listed in Table 1. The radiative recombination coefficients for arbitrary temperature which is not shown in the table is estimated by

Level	T_e (k)	$\alpha_j (T_e) \times 10^{15} \text{ cm}^3/\text{sec}$				
		1800	2200	2600	3000	3400
6S		1.34	1.15	1.01	0.89	0.797
6P		137	124	114	106	99.9
5D		322	293	272	255	241
7S		0.333	0.278	0.239	0.208	0.184
7P		43.8	39.6	36.4	33.9	31.8
6D		65.3	60.1	56.1	53.0	50.4
8S		0.198	0.165	0.140	0.122	0.108

Table 1 Radiative recombination coefficients.

interpolation using the numerical values shown in the table.

2.4 De-excitation

The excited atoms are depopulated by de-excitation by the electron collision, the natural decay by emission and the step ionization from this excited state. The de-excitation cross section through the electron collision is expressed in terms of the the excitation cross section using the principle of detailed balance in the same manner as for the case of the three body recombination coefficient

$$\sigma_{ejg}(W_e) = \frac{g_g}{g_j} \frac{W_e + W_j}{W_e} \sigma_{ejg}(W_e + W_j), \quad (14)$$

where σ_{ejg} is the de-excitation cross section.

Einstein transition probability from the jth state to the kth state by emission is related to the oscillator strength f_{jk} by

$$A_{jk} = \frac{2\pi e^2 g_j}{\epsilon_0 m_e c \lambda_{jk}^2 g_k} f_{jk} \quad (15)$$

where λ_{jk} is the wave length of the light emitted by the transition from the jth state to the kth state, c the light speed and e the electron charge. The oscillator strengths f_{jk} and the transition probabilities calculated by eq.(15) are listed in Table 2.

In optically thick plasma the self-absorption occurs, that is, the light emitted by the atom is partly absorbed by the atom. Therefore the rate of natural decay per unit time is given by the product of Einstein transition probability and the transmission probability P_{jk} which represents the degree of the self-absorption. Since the density of the excited atom is smaller than that of the ground state, the self-absorption except for the transition to the ground state is very small. Hence, we assume that the transmission probability of the light for the transition to the excited state is equal to 1, The transmission probabilities of the light for the transition to the ground state are taken from the paper of Oshitani et al..¹⁸⁾

j	k	λ_{jk} (Å)	f_{jk}	A_{jk}
6P _{1/2}	6S _{1/2}	8944	3.94×10^{-1}	3.27×10^7
6P _{3/2}	"	8521	8.14×10^{-1}	3.73×10^7
5D _{3/2}	6P _{1/2}	30100	2.51×10^{-1}	9.21×10^5
5D _{3/2}	6P _{3/2}	36139	2.11×10^{-2}	1.07×10^5
5D _{5/2}	"	34892	2.04×10^{-1}	7.48×10^5
7S _{1/2}	6P _{1/2}	13589	1.71×10^{-1}	6.18×10^6
"	6P _{3/2}	14695	2.08×10^{-1}	1.28×10^7
7P _{1/2}	6S _{1/2}	4593	2.84×10^{-3}	8.95×10^5
7P _{3/2}	"	4555	1.74×10^{-2}	2.79×10^6
7P _{1/2}	7S _{1/2}	30958	5.56×10^{-1}	3.86×10^6
7P _{3/2}	"	29316	1.11×10^0	4.31×10^6
6D _{3/2}	6P _{1/2}	8761	2.98×10^{-1}	1.29×10^7
"	6P _{3/2}	9209	3.97×10^{-2}	3.11×10^6
6D _{5/2}	6P _{1/2}	9127	3.33×10^{-1}	8.76×10^6
6D _{3/2}	7P _{1/2}	121471	3.27×10^{-1}	7.39×10^4
"	7P _{3/2}	155707	3.20×10^{-2}	8.79×10^3
6D _{5/2}	7P _{1/2}	115449	3.09×10^{-1}	5.15×10^4
8S _{1/2}	6P _{1/2}	7609	2.02×10^{-2}	2.33×10^6
"	6P _{3/2}	7944	2.04×10^{-2}	4.03×10^6
"	7P _{1/2}	39192	2.97×10^{-1}	1.29×10^6
"	7P _{3/2}	42185	3.33×10^{-1}	2.49×10^6
8P _{1/2}	6S _{1/2}	3888	3.17×10^{-4}	1.39×10^5
8P _{3/2}	"	3876	3.49×10^{-3}	7.74×10^5
8P _{1/2}	7S _{1/2}	13940	5.16×10^{-3}	1.77×10^5
8P _{3/2}	"	13781	2.56×10^{-2}	4.48×10^5

Table 2 Oscillator strengths and transition probabilities.

2.5 Diffusion

The radial diffusion process is one of loss processes of the charged particle. Assuming the ambipolar diffusion, the diffusion time is given by

$$\tau = \left(\frac{R}{2.4}\right)^2 \frac{e}{\mu_i k T_e} \quad (16)$$

where R is the radius of the plasma. μ_i the ion mobility, which was measured experimentally by Chanin.¹⁹⁾

§3. Rate Equations

We consider seven states for the excited state. The excited atom in a level lying above the seventh level is ionized with high probability because the energy necessary to ionize starting from this excited state is less than 0.3 eV and the corresponding ionization cross section is very large in comparison with those from seven excited levels. Therefore, it can be considered that the excited levels lying above the seventh level can be regarded as a virtual free state

In our model, the collisional transitions between the excited states are neglected because of the fact that their rates are less than the rate of the excitation by the electron collision with the ground state atom, and that the energy gaps between the excited states of cesium atom are less than 0.4 eV and therefore the collisional transition from the j th state to the k th state is almost canceled out by that from the k th state to the j th state.

The densities of the electron, ion and atom depend on the production and loss processes which are governed by collisional and radiative processes discussed in §2. Setting the rate of the production of a particle to be equal to the rate of the loss, the rate equations for the electron, the excited atom and the ground state atom are given as follows respectively,

$$\begin{aligned} N_e \nu_{eg_i} + N_e \sum_k (\nu_{ek_i} - N_e \alpha_k - N_e^2 \beta_k) - N_e / \tau &= 0, \\ N_e (\nu_{eg_j} - \nu_{ej_i} - \nu_{ej_g} + N_e \alpha_j + N_e^2 \beta_j) + \sum_{k>j} N_k A_{kj} P_{jk} - N_j \sum_{k<j} A_{jk} P_{jk} &= 0 \\ N_e \sum_k (\nu_{ek_g} - \nu_{eg_k}) + N_e (N_e \alpha_g + N_e^2 \beta_g - \nu_{eg_i} + \sum_k N_k A_{kg} P_{kg}) &= 0, \end{aligned} \quad (17)$$

where the quasi-neutrality of plasma $N_e \sim N_i$ is assumed. The initial density of cesium atom N_{g0} is related to the other densities by the following relation,

$$N_{g0} = N_e + N_g + \sum_j N_j \quad (18)$$

Since the collision frequencies and the recombination coefficients have been already given as a function of the temperature and densities, we can express the electron density as a function of the electron temperature solving simultaneously ten equations; eqs. (17) the rate equations for the electron, the six excited state atoms and the ground state atom and eq. (18) the relation between the densities.

The electron densities as a function of the electron temperature are shown in Fig. 4, in which the parameters represent the initial density of cesium atom. The broken lines represent the electron density calculated by Saha equation, eq. (12). In low temperature and low pressure region, the electron densities given by this model are less those by the Saha equation. The electron densities approach gradually to those evaluated by the Saha equation with increasing the temperature and pressure.

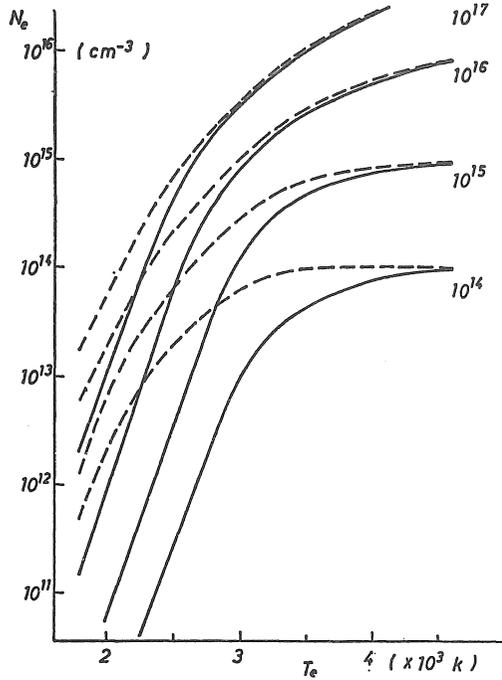


Fig.4 Electron densities as a function of the electron temperature with a parameter of the initial density.

N_g (cm^{-3})	T_e (k)	Electron densities (cm^{-3})			
		1	3	5	7
10^{14}	1800	3.4×10^8	1.2×10^9	2.4×10^9	2.7×10^9
	2400	3.8×10^{10}	8.1×10^{10}	1.2×10^{11}	1.3×10^{11}
	3000	4.2×10^{12}	5.3×10^{12}	6.0×10^{12}	6.2×10^{12}
	3600	6.3×10^{13}	6.5×10^{13}	6.7×10^{13}	6.7×10^{13}
	4200	9.2×10^{13}	9.2×10^{13}	9.2×10^{13}	9.2×10^{13}
10^{16}	1800	3.2×10^{10}	6.8×10^{10}	9.7×10^{10}	1.1×10^{11}
	2400	1.3×10^{13}	1.9×10^{13}	2.4×10^{13}	2.5×10^{13}
	3000	7.6×10^{14}	7.7×10^{14}	7.8×10^{14}	7.8×10^{14}
	3600	3.2×10^{15}	3.2×10^{15}	3.2×10^{15}	3.2×10^{15}
	4200	6.3×10^{15}	6.3×10^{15}	6.3×10^{15}	6.3×10^{15}

Table 3 Electron densities calculated by the model with different number of the excited state.

In order to illustrate how the inclusion of the excited state other than the resonance one allows us to give an accurate description of the plasma properties, we consider the model with different number of the excited state, i.e., the electron density is calculated by taking one, three, five and seven excited states into account respectively, and the results are listed in Table 3. At the high temperature and the high initial density,

the electron densities are independent on the number of the excited state considered. When the temperature and the initial density are low, the electron density decreases with the decrease of the number of the excited state.

§4. Discussion and Conclusion

We calculated the electron density of the nonequilibrium cesium plasma on the basis of the collisional-radiative model involving not only the resonance state but also seven excited states. From Fig.4, the electron densities given by our model are less than those for local thermal equilibrium when the electron temperature and initial density are low. Since the loss by diffusion and radiation increases with decreasing the temperature and the pressure, the local thermal equilibrium attained by the collision is broken and the electron density is reduced to less than that in thermal equilibrium. In fact, we estimated the electron density considering only the collision process but neglecting the diffusion and the radiation processes and obtained the same value as that given by the Saha equation for whole ranges of temperature and density.

When the electron temperature and the initial density are high, the electron density is constant independent on the number of the excited state being involved in the model (Table 3). Therefore, it is sufficient to consider only the resonance state 6P for the excited state. However, at the low temperature and the low initial density, for example, at the initial density 10^{14} cm^{-3} and the electron temperature of 1800°K , the electron density calculated by the model with seven excited states is higher than eight times that with only the resonance state for the excited state. Since the collisional excitation probability or excitation frequency corresponding to the resonance state is very high in comparison with the excitation probability corresponding to upper states as shown in Fig. 2, the population of the resonance state is very large. Inversely, the stepwise ionization probability of the excited atom in upper energy state is higher than that in lower energy state as shown in Fig. 3. As the rate of the stepwise ionization by the electron collision with the excited atom depends on the product of the excitation probability and the stepwise ionization probability, the excited atom in the upper state can not always be neglected by the reason that the excitation probability is low.

The collisional excitation cross section for an electron with an energy just above the excitation energy has very large value. If the electron temperature is low, a part of electrons with high energy in the electron energy distribution can excite an atom, and the number of electrons participating in the excitation to the upper state is not small as compared with that to the lower state. Therefore, the excited atom in the upper state can not be neglected. When the electron temperature increases, the number of electrons participating in the excitation to the lower state increases more than that to the upper state. The excited atom in the lower state plays a dominant role and the number of the excited state which should be taken into account decreases with increasing the temperature. For higher pressure, the number of the excited state is similarly small. Since collisions occur frequently, the loss by diffusion and radiation can be neglected and the system approaches the local thermal equilibrium.

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