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Discussion on Collisional Radiative Model from the Viewpoint of Ordinary Differential Equations — Stability and Transient Response

衝突輻射モデルに関する常微分方程式論的考察〜安定性と過渡応答

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In the present study, we examine a general solution to the associated linear homogeneous differential equations of the CR model, and survey the behavior of eigenvalues of the characteristic matrix, which corresponds to the reciprocal time-constant of the damping modes of the excited states to the steady state solution. It is proved that the differential equations describing the CR model are exponentially stable. The time constants of decay to the perturbation for the metastable levels are sometimes considerably long about several tens of microsecond for general glow discharge, whereas those for the excited states applied to spectroscopic observation frequently like 4p or 4p' levels are about sub microsecond, which is applicable to general diagnostics of discharge plasmas.

We often apply optical emission spectroscopy (OES) measurement to examine plasmas in many scientific fields, not only for basic science but also for practical engineering. Line intensities of the plasmas indicate number densities of corresponding upper excited states of the transition N_p (p = 1, 2, …, M in the ascending order of energy; p = 0 as a ground state), which are theoretically described by collisional-radiative (CR) model as functions of electron temperature T_e and density N_e. By applying the CR model, we can estimate the essential plasma parameters T_e and/or N_e from the excited-states populations. A large number of studies are being carried out for innovative diagnostics to determine T_e and/or N_e from OES measurement of plasmas.

Up to the present time, however, most researchers are interested in steady-state solution to the CR model. From the mathematical point of view, the governing equations of the CR model are categorized as first-order non-homogeneous linear ordinary differential equations (ODE) with constant coefficients, where unknown functions are number densities of excited states N_p:

$$\frac{\mathrm{d}\boldsymbol{N}}{\mathrm{d}t} = a\boldsymbol{N} + \boldsymbol{\delta}, \qquad (1)$$

where bold fonts denote vectors,

$$\boldsymbol{N} = \begin{pmatrix} N_1 \\ N_2 \\ \vdots \\ N_M \end{pmatrix}, \qquad \boldsymbol{\delta} = \begin{pmatrix} \delta_1 \\ \delta_2 \\ \vdots \\ \delta_M \end{pmatrix}, \qquad (2)$$

and

$$\delta_i = \alpha_i N_e^3 + \beta_i N_e^2 + C_{0i} N_e N_0. \qquad (3)$$

Symbols used above are the same as in [1]. In Eq. (1), a is an $M \times M$ square matrix, and whose components are given as follows:

$$a_{ji} = \begin{cases} N_e C_{ji} & \text{for } j < i, \quad (4) \\ N_e C_{ji} + A_{ji} & \text{for } j > i, \quad (5) \\ -N_e\left(S_i + \sum_{l=0,\neq i}^{M} C_{il}\right) - \sum_{l=0}^{i-1} A_{il} & \text{for } j = i. \quad (6) \end{cases}$$

The matrix a does not contain the time t explicitly, and consequently, Eq. (1) becomes an ODE with constant coefficients. The solution to Eq. (1) is given by the sum of the general solution of the related homogeneous equation and anyone of the particular solutions to Eq. (1). One of the simplest particular solutions to Eq. (1) is the steady-state solution. That is,

$$\overline{\boldsymbol{N}} = -a^{-1}\boldsymbol{\delta} = -a^{-1}\boldsymbol{\delta}_{\mathrm{rec}} - a^{-1}\boldsymbol{\delta}_{\mathrm{ion}}, \qquad (7)$$

where we defined

$$\boldsymbol{\delta}_{\mathrm{rec}} = \boldsymbol{\alpha} N_e^3 + \boldsymbol{\beta} N_e^2, \qquad (8)$$

$$\boldsymbol{\delta}_{\mathrm{ion}} = \boldsymbol{C}_0 N_e N_0. \qquad (9)$$

In the society of plasma spectroscopy, almost all the discussions on CR model seem to have been concentrated on the non-homogeneous solution to Eq. (1), that is, the steady-state solution, Eq. (7). The first term is referred to as the recombining component and the second one as the ionizing component. However, such discussion is valid only when the excited species reaches the steady state after relaxation time has passed for the excited-state populations. If we would like to treat the transient response of population density of excited states, or find the plasma parameters of short-time pulse discharge, we must discuss time-dependent solutions to Eq. (1). That is, when we discuss time-dependent excited kinetics, we must return to the general solutions of associate homogeneous ODE.

For this purpose, we must examine the eigenvalues λ_i of the CR matrix a as Eqs. (4) – (6). The general solutions of associate homogeneous ODE is given as follows:

$$N(t) = \sum_{i=0}^{M} C_i \exp(\lambda_i t) \xi_i \,, \qquad (10)$$

where ξ_i is the i-th eigenvector corresponding to the eigenvalue λ_i, and C_i are arbitrary constants. If there is a degeneracy in matrix a, some terms in Eq. (10) should be changed into a summation including $t \times \exp(\lambda_i t)$. This, however, is not essential in the present discussion, since we can prove that the real part of any eigenvalues is negative, and that the system is exponentially stable in terms of an ODE system by Gershgorin's theorem [2].

[Proposition] Real parts of all the eigenvalues of a square matrix a whose components are given in Eqs. (4) – (6) are negative.

[Proof] Gershgorin's theorem shows that there always exists an appropriate j that satisfies the following equation for an arbitrary eigenvalue λ_m,

$$\left|\lambda_m - a_{jj}\right| \le \sum_{\substack{k=1 \\ k\ne j}}^{M} \left|a_{jk}\right|. \qquad (11)$$

Equation (6) allows us to rewrite LHS of Eq. (11) as

$$\left|\lambda_m - a_{jj}\right| = \left|\lambda_m + N_e\left(S_j + \sum_{\substack{l=0 \\ l\ne j}}^{M} C_{jl}\right) + \sum_{l=0}^{j-1} A_{jl}\right|. \qquad (12)$$

Since each term of RHS of Eq. (11) is positive,

$$\sum_{\substack{k=1 \\ k\ne i}}^{M} \left|a_{jk}\right| = N_{\mathrm{e}} \sum_{\substack{k=1 \\ k\ne j}}^{M} C_{jk} + \sum_{k=1}^{j-1} A_{jk} \,. \qquad (13)$$

Next, let us define a positive and real parameter T_j as follows:

$$N_{\mathrm{e}}\left(S_j + \sum_{\substack{k=0 \\ k\ne j}}^{M} C_{jk}\right) + \sum_{k=0}^{j-1} A_{jk} = T_j. \qquad (14)$$

Substituting Eqs. (12) – (14) into Eq. (11), we have

$$\left|\lambda_m + T_j\right| < T_j. \qquad (15)$$

Equation (15) shows that the arbitrary eigenvalue λ_m is located within the circle whose center is located at $(-T_j)$ in the negative part on the real axis in the complex plane, and whose radius is smaller than T_j, which is real and positive. Then, the real part of the arbitrary eigevalue is concluded to be negative. This indicates that the ODE describing the CR model are exponentially stable. [Q.E.D.]

As a next step, let us examine the eigenvalues of Ar I excited-level systems of argon discharge,

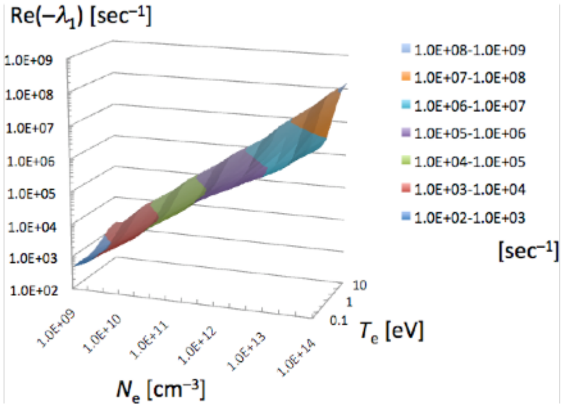

Fig. 1. Dependence of the real part of the first eigenvalue λ_1 on T_{e} and N_{e} for Ar plasma with $P = 1$ Torr and $T_{\mathrm{g}} = 500$ K.

which is often applied as main discharge species in various process engineering, as functions of electron temperature and density. Concerning numbering of the eigenvalues, we define $\lambda_1, \lambda_2, \ldots, \lambda_M$ according to the absolute value of their real part, that is, $|\mathrm{Re}(\lambda_1)| \le |\mathrm{Re}(\lambda_2)| \le \ldots \le |\mathrm{Re}(\lambda_M)|$. It indicates that the relaxation time of the excitation kinetics of the excited states is given by $|\mathrm{Re}(\lambda_1)|^{-1}$.

Figure 1 shows the dependence of $|\mathrm{Re}(\lambda_1)|$ as function of the electron temperature T_{e} and density N_{e} of Ar plasma with its discharge pressure 1 Torr and the gas temperature 500 K with Maxwellian EEDF [1]. It is found that $|\mathrm{Re}(\lambda_1)|$ is approximately proportional to the electron density N_{e}. On the other hand, we found that $|\mathrm{Re}(\lambda_1)|$ becomes about ten times larger almost stepwise at $T_{\mathrm{e}} \sim 1 - 3$ eV.

Concerning a practical application, we should be very careful about the treatment of the excited states as steady-state. However, since the eigenvector of this mode mainly concerns 4s or 4s'states, which are the metastable levels or the levels strongly coupled with them, these time constants are not practically in problem in the OES measurement, where we often apply 4p or 5p levels. For these levels, we found λ_5 has the shortest time constant, which is 1/50 times smaller than λ_1. We should examine excited states with shorter time constants when we measure rapid variation. We need further mathematical discussion about practical values or dependences of eigenvalues with respect to various plasma parameters. We also need discussion about the evolution of EEDF based upon the time-dependent Boltzmann equation.

References

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[2] G. Faussurier, C. Blancard, T. Kato and R. M. More: High Energy Density Phys., **4**, (2008) 88.