

A fitting formula for radiative cooling based on non-local thermodynamic equilibrium population from weakly-ionized air plasma

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Abstract. A fitting formula for radiative cooling with collisional-radiative population for air plasma flowfield has been developed. Population number densities are calculated from rate equations in order to evaluate the effects of nonequilibrium atomic and molecular processes. Many elementary processes are integrated to be applied to optically-thin plasmas in the number density range of $10^{12}/\text{cm}^3 \leq N \leq 10^{19}/\text{cm}^3$ and the temperature range of $300 \text{ K} \leq T \leq 40,000 \text{ K}$. Our results of the total radiative emissivity calculated from the collisional-radiative population are fitted in terms of temperature and total number density. To validate the analytic fitting formula, numerical simulation of a laser-induced blast wave propagation with the nonequilibrium radiative cooling is conducted and successfully reproduces the shock and plasma wave front time history observed by experiments. In addition, from the comparison between numerical simulations with the radiation cooling effect based on the fitting formula and those with a gray gas radiation model that assumes local thermodynamic equilibrium, we find that the displacement of the plasma front is slightly different due to the deviation of population probabilities. By using the fitting formula, we can easily and more accurately evaluate the radiative cooling effect without solving detailed collisional-radiative rate equations.

1. Introduction

In most aerospace applications of air plasma flow (hypersonic flow around a reentry body [1–4], laser-driven blast wave [5–8], and plasma processing techniques using electric discharges), plasma internal states do not achieve local thermodynamic equilibrium (LTE). It is necessary to consider nonequilibrium properties of population distribution into each plasma internal state. The nonequilibrium properties directly affect radiative emissivities and opacities, partition functions of internal energy modes, and thermal relaxations. These effects of nonequilibrium processes are still a problem that remains to be solved in those applications.

For instance, we take a gas-driven laser-propulsion system whose thrust power is obtained through interaction with a propellant gas heated by external laser beam irradiation. After a focused laser breaks down the propellant gas, a blast wave is formed by the laser-produced plasma and propagates to the projectile through the surrounding gas. The performance of a gas-driven laser propulsion system depends heavily on the blast-wave dynamics and the plasma states sustaining it. Our previous work [8] numerically simulated a laser-driven blast wave



coupled with rate equations based on a collisional-radiative (CR) model for argon plasma, in order to study the time-dependent effect of excitation and ionization processes during pulse heating. Computational results revealed that a laser-induced plasma driving a blast wave is still in ionization nonequilibrium over the pulse duration. The blast wave shell is accelerated more by the driving plasma, which is in the ionization relaxation state, because there is less radiative energy loss than indicated by plasma flow computations assuming LTE. Note that the radiation emitted from the driving plasma in the unsteady ionizing phase is weaker than that from the LTE population, since the number densities of highly-excited species are relatively lower.

As another example, we take a hypersonic flow around a space vehicle entering into the earth atmosphere. In front of the space vehicle, a strong shock wave with a high temperature layer is formed and then aerodynamic and radiative heating occurs. To protect the space vehicle from such a severe heating environment, an appropriate thermal protection system (TPS) must be equipped. For accurate prediction of the heating rates to design a proper TPS during the entry flight, the behavior of excited species has to be well known. This can be achieved through detailed description of nonequilibrium air plasma states through a CR model [1–4].

The objective of this study is to develop a curve-fitted radiative cooling formula for weakly-ionized air plasma. Population number densities are calculated from collisional-radiative rate equations in order to estimate the effects of nonequilibrium atomic and molecular processes. Our CR model consists of fifteen air species: e^- , N, N^+ , N^{2+} , O, O^+ , O^{2+} , O^- , N_2 , N_2^+ , NO, NO^+ , O_2 , O_2^+ , and O_2^- with their major electronic excited states. Many elementary processes are included and applied to doubly-ionized air plasma in a number density range of $10^{12}/\text{cm}^3 \leq N \leq 10^{19}/\text{cm}^3$ and a temperature range of $300 \text{ K} \leq T \leq 40,000 \text{ K}$. For the validation of the developed radiative cooling formula, we compute a laser-induced blast wave propagation with radiative energy loss, and compare with experimentally observed time evolution of shock wave and plasma front displacement.

2. Collisional-radiative model and radiative cooling formula

The previously developed CR code for air plasma in the optically thin case is employed to calculate nonequilibrium population densities. More detailed descriptions of our code are in the literature [9].

2.1. Atomic and molecular model

We consider that air plasma is composed of fifteen species: e^- , N, N^+ , N^{2+} , O, O^+ , O^{2+} , O^- , N_2 , N_2^+ , NO, NO^+ , O_2 , O_2^+ , and O_2^- . For atomic species, each fine structure is degenerated into terms, in addition, terms are coalesced into configurations for highly excited states. For molecular species, electronic states are included to encompass a group of major radiation bands from the infrared to the vacuum ultraviolet wavelength regions. Each rotational, vibrational, and nuclear spin states is coalesced into one electronic state after the manner of Bacri and Medani [10]. All of atomic energies and degeneracies of each electronic state and molecular spectroscopic constants are referred to Saruhashi's radiative database [11]. Our CR model comprises a total of 310 different levels [9].

2.2. Rate equations and rate constants

Number densities $N_{s,i}$ of the coalesced states i of the different chemical species s are calculated using the conservation equation:

$$\frac{dN_{s,i}}{dt} = \sum \text{populating terms} - \sum \text{depopulating terms}. \quad (1)$$

The source terms are populating and depopulating number densities per unit of time, summed over involved transition reactions. It is assumed two temperature model consist of the translation

and rotation temperature T and the vibration, electronic excitation, and electron temperature T_e to take into account of thermal nonequilibrium. Transition rate constants included in the right hand side of (1) then are expressed as a function of (T, T_e) . The number density of free electron n_e is calculated by the charge neutrality assumption for the partially ionized plasma. For the numerical time integration, we employ the backward differentiation formula (BDF) method included in the LSODE package [12]. This method is a one of the representative multi-step implicit method, and can stably solve the stiff system of ordinary differential equations.

The electron impact excitation and ionization cross sections are given by the Drawin's model [13, 14]. Rate coefficients of inverse processes, such as deexcitation and three body recombination, are derived using the detailed balance principle. For electron impact processes of diatomic molecules are calculated from Bacri's weighted-total cross section method [15], and tangible data of their rate constants are obtained from Teulet et al. [16] Backward transition rate constants obey the detailed balance in common with atomic rates. Additionally, we employ Park's rate constants for electron collisional dissociation and dissociative recombination [17]. For heavy particle impact processes, we consider fourteen chemical reactions as shown in Park's book [1]. Arrhenius parameters and the equilibrium constants for each chemical reaction are also addressed under Park's book.

Both atomic and molecular spontaneous emission coefficients $A(i, j)$ are used from Saruhashi's database [11]. We also take into account the radiative recombination rate $R_r(i, j; T_e)$ and the dielectronic recombination rate $R_d(i, j; T_e)$ of monatomic ions, using Nahar's recombination rate database [18, 19]. Furthermore, the radiative attachment rate constant for an oxygen atom ($O + e \rightarrow O^- + h\nu$) is derived from Soon and Kunc [20].

2.3. Total radiative emissivity

The total radiative emissivity Q_{rad} can be expressed by the summation of following three different electronic transition processes.

$$Q_{\text{rad}} = \sum_s J_{s,bb} + \sum_s J_{s,fb} + J_{ff}. \quad (2)$$

(i) atomic line and molecular band emission $J_{s,bb}$ is

$$J_{s,bb} = \sum_{i>j} N_{s,i} A(i, j) E_{ij}, \quad (3)$$

where E_{ij} represents energy level.

(ii) recombination radiation $J_{s,fb}$ can be expressed as [21]

$$J_{s,fb} = n_e N_{s,i} R_r(i, j; T_e) \left(I_{\infty,s} + \frac{3}{2} k T_e \right), \quad (4)$$

where $I_{\infty,s}$ represents ionization potential.

(iii) Bremsstrahlung radiation J_{ff} can be expressed as [22]

$$J_{ff} = 1.426 \times 10^{-28} G n_e^2 \bar{z} \sqrt{T_e} (1 + D), \quad (5)$$

where $\bar{z}$ represents the averaged ionization degree. The averaged Gaunt factor G and the average nonhydrogenic correction factor D are set to 1.2 and -0.15, respectively.

2.4. Appropriate conditions for local thermodynamic and Corona equilibrium

In this subsection, we refer to quasi-steady state in the CR model as the CR steady state (CRSS). CRSS is an intermediate state between LTE and Corona equilibrium (CE) [21]. In LTE or CE, the population distribution among the various excitation states $\mathcal{P}_{s,i} = N_{s,i}/N_s$ can be determined by basic principles. For LTE, $\mathcal{P}_{s,i}$ obeys the Boltzmann distribution function,

$$\mathcal{P}_{s,i}^{\text{LTE}} = \frac{N_{s,i}}{N_s} = \frac{G_i}{Q_s} \exp\left(-\frac{E_i}{kT_e}\right). \quad (6)$$

In contrast, in low-density CE plasmas, the upward excitation rate due to collisions is so low relative to the spontaneous decay rate that one can safely assume that an electron excitation to an upper level will most likely decay to the ground state before experiencing further excitation. Therefore, most chemical species are in their ground state; $\mathcal{P}_{s,i}^{\text{CE}} = N_{s,1}/N_s = 1$. Unlike LTE or CE, in CRSS the population distribution $\mathcal{P}_{s,i}^{\text{CRSS}}$ cannot be determined from these principles, and the density of a given excited species can be solved only from the whole set of rate equations. Thus, we must take into account all the processes that increase or decrease the excited species' abundance. Using CRSS is not always easy and convenient; whenever possible, it is preferable to use LTE or CE to model a plasma. It is therefore important and helpful to know temperature and density conditions for which these three models are valid.

Figure 1 indicates appropriate conditions to employ the above three models. Computations are carried out over a temperature range of 5,000 K to 40,000 K, and a total number density range of $10^{12}/\text{cm}^3$ to $10^{19}/\text{cm}^3$. Such conditions cover a range associated with many applications utilizing air plasmas (e.g., hypersonic flow around a reentry space vehicle, air-driven laser-propulsion system, electric discharges, plasma processing, and arcs in high-power electric thrusters). Figure 1 confirms that the discrepancy from LTE is large in a lower-density, higher-temperature region. We can find that the population probability $\mathcal{P}_{s,i}^{\text{CRSS}}$ does not achieve Boltzmann equilibrium when the total number density is less than or equal to $10^{16}/\text{cm}^3$. Therefore, the CRSS population must be considered when we analyze low-density plasmas.

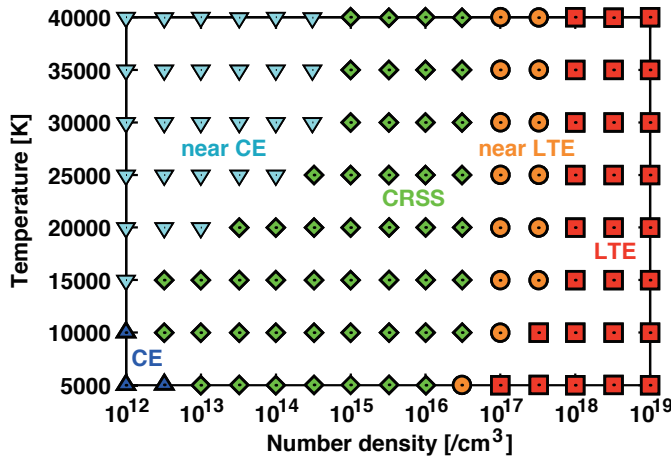


Figure 1. Summary of appropriate conditions for the air plasmas in the total number density range from $10^{12}/\text{cm}^3$ to $10^{19}/\text{cm}^3$ and the temperature range from 5,000 K to 40,000 K.

Note that, in this work, the quasi-steady state is defined as the numerical solution of the time-dependent rate equations at 1 μs with residuals of all time derivatives less than 10^{-8} . If time derivatives of number densities of the some excited states are not converged at 1 μs , the numerical time integration is continued until the residuals become lower than the threshold. Because long-lifetime excited species having a small number density are still emitting radiation

in a CE or a near CE state. On the other hand, while in a near LTE state whose internal state distributions approach Boltzmann equilibrium function in the quasi-steady state, it would be required to solve the whole set of time-dependent rate equations when evaluating unsteady properties of air plasmas more accurately down to submicrosecond scale.

2.5. Fitting formula for radiative cooling with CR population

It is very helpful to develop a radiative cooling formula for nonequilibrium flow computation, since we can easily include the radiative energy loss with low computational cost. In this section, we present a curve fitted formula for radiative cooling and results of the total radiative emissivity Q_{rad} (see (2)–(5)) calculated from the collisional-radiative population that was shown in figure 1.

Obtained results are indicated in figure 2 in the total number density range from $10^{12}/\text{cm}^3$ to $10^{19}/\text{cm}^3$ and the temperatures from 5,000 K to 40,000 K. Vertical axis is the radiative emissivity in common logarithm scale, and each curve represents number densities from $10^{12}/\text{cm}^3$ to $10^{19}/\text{cm}^3$. For all the cases, the radiative emissivities are monotonically increasing function of temperature and total number density. We have fitted radiative emissivity Q_{rad} in terms of temperature T_e and total number density N as follows,

$$\log_{10} Q_{\text{rad}}(T_e, N) = a + \frac{b}{Z} + \frac{c}{Y} + d Y + e Y^2, \quad (7)$$

where $Y = \log_{10} N$ and $Z = T_e/10000$. Fitting parameters are given by

$$a = -127.4, \quad b = -5.931, \quad c = 425.9, \quad d = 10.16, \quad e = -0.2148. \quad (8)$$

This curve fitted function means a modification to the gray gas radiation in accordance with the nonequilibrium population. By utilizing this relation, we can evaluate radiative energy loss without solving CR rate equations.

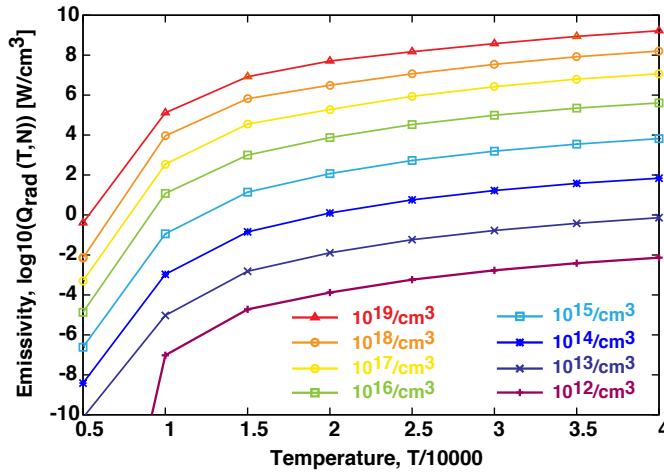


Figure 2. Total radiative emissivities computed from CR population in the total number density range from $10^{12}/\text{cm}^3$ to $10^{19}/\text{cm}^3$ and the temperature range from 5,000 K to 40,000 K.

3. Validation of the radiative cooling formula by laser-induced blast wave

Hereafter, to validate our fitting formula (7) for radiative cooling with CR population. We compute a laser-induced blast wave coupled with radiative energy loss and compare with experimentally observed shock wave and plasma front displacements [7]. The blast wave code is based on axisymmetric Navier-Stokes equations with eleven chemical species and chemical reactions including some ionization reactions. An amount of laser energy absorbed in flowfield is computed with the ray-tracing method [23]. Description of the code is summarized as follows.

3.1. Flow equations

The governing equations are the two-dimensional axisymmetric Navier-Stokes equations, which can be written as

$$\frac{\partial y \mathbf{Q}}{\partial t} + \frac{\partial y (\mathbf{F} - \mathbf{F}_v)}{\partial x} + \frac{\partial y (\mathbf{G} - \mathbf{G}_v)}{\partial y} + \mathbf{S} = y \mathbf{W}, \quad (9)$$

where $\mathbf{Q}$ is the conservative variables, $\mathbf{F}$ and $\mathbf{G}$ are the convective flux vector, $\mathbf{F}_v$ and $\mathbf{G}_v$ are the viscous flux vector, $\mathbf{S}$ is the vector from axisymmetric formulation, and $\mathbf{W}$ is the source term. The equation set (9) consists of ten species mass, momentum, total energy and vibrational, electronic excitation, and free electron energy conservation equations. The conservative variables $\mathbf{Q}$ thus becomes

$$\mathbf{Q} = (\tilde{\rho}_s, \rho_s, \rho u, \rho v, \epsilon, \epsilon_v + \epsilon_{ex} + \epsilon_{el})^t, \quad (10)$$

where a charge neutrality is assumed to omit ρ_{e-} . Here, $\tilde{\rho}_s$, ρ_s , u , v , ϵ , ϵ_v , ϵ_{ex} and ϵ_{el} are the density of elemental species s , the density of species s , the velocity in the x -direction, the velocity in the y -direction, the total energy per unit volume, the vibrational energy per unit volume and the electronic energy per unit volume, the free electron energy per unit volume. The total density ρ is given by the sum of the densities of all species. The viscosity of each species is given by a model for reacting flow developed by Blottner et al. [24]. The conductivities of translational-rotational temperature and vibrational temperature for each species are given by Eucken's relation [25]. Moreover, the viscosity and conductivity of the mixture gas are calculated using Wilke's semi-empirical mixing rule [26]. The mass diffusion coefficients are assumed to be constant for all species with a constant Schmitt number S_c of 0.5. Therefore the mass diffusion coefficients D_s for each species s can be given by $D_s = \mu / S_c \rho$, where μ is the viscosity of the mixture. We employ the reaction rate coefficients proposed in the two-temperature model of Park [27] as mentioned above. The vibrational-electronic-electron energy conservation equation accounts for the following eight physical processes.

- (i) energy transfer between the translational and vibrational energy modes
- (ii) vibrational energy source due to dissociation or recombination reactions
- (iii) energy transfer between the molecular translational and electron translational energy modes
- (iv) electron energy loss when a free electron strikes a neutral particle and frees another electron
- (v) electronic excitation energy production
- (vi) free electron energy source due to ionization reactions
- (vii) free electron energy generated by laser absorption
- (viii) energy loss by radiative cooling Q_{rad}

The governing equations are discretized with a cell-centered finite volume method. We employ the AUSM-DV upwind scheme for obtaining the numerical flux [28]. A second-order spatial accuracy is obtained by the MUSCL approach [29]. An Euler explicit method is used for the time integration, while the diagonal point implicit method [30] is employed in the time integration of source terms. Note that a more detailed description about these numerical modeling can be found in previous a paper [31].

3.2. Numerical condition

At the beginning of the simulation, we make a focal spot which has a higher electron temperature because of the lack of the breakdown processes in our code. The electron temperature of the spot is distributed in a Gaussian form with FWHM of 10 μm and the peak value of $\simeq 7,000$ K. A two-dimensional ray-tracing method is employed to find out the absorption rate by inverse bremsstrahlung of free electron. A laser pulse profile is assumed to be composed of a pulse and a tail part as indicated in figure 3. the properties of our laser are that the FWHM of

a pulse part is 40 ns, the maximum power is 24 MW, the total laser energy is 10 J, and the wavelength of laser is $10.6 \mu\text{m}$. These the properties are the same of the CO_2 laser that is employed in the experiment of laser-induced blast wave [7]. The simulation domain covers a $32 \text{ mm} \times 12 \text{ mm}$ section with 161×61 grid points as shown in figure 4. The incident laser is divided into 160 rays and is traced from the right boundary with a Gaussian distribution whose FWHM is 40 mm. The divided rays are inclined to be focused at the distance of 25 mm from the boundary. Initial temperature and density are 288.15 K and 1.225 kg/m^3 , the same as in the experimental conditions.

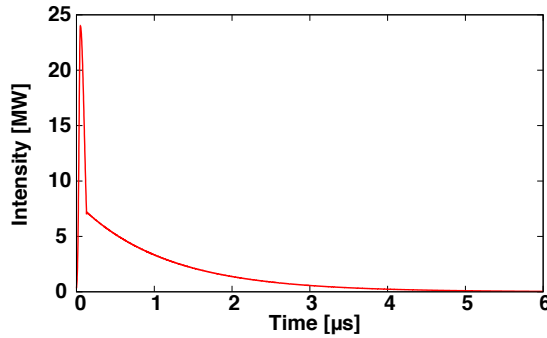


Figure 3. Incident laser profile.

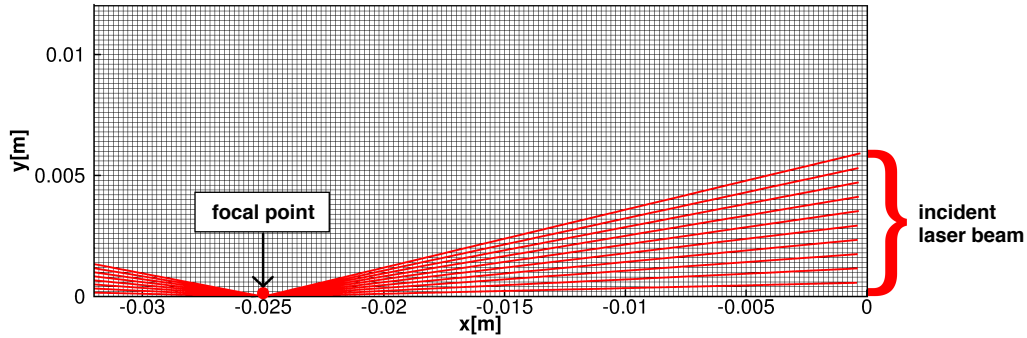


Figure 4. Computational domain and initial laser alignment.

3.3. Propagation speed of shock and ionization wave

Figures 5 (a)-(d) show the pressure (upper) and the electron number density (lower) contours at $t = 1, 2, 3,$ and $6 \mu\text{s}$ after a laser beam irradiation. Seeds of free electrons generated by the initial ionization absorb laser energy at the focal point ($x = -0.025 \text{ m}$). At first, the ionization wave propagates toward the incident direction of the laser, and the tear-drop-shaped blast wave is generated as shown in figure 5 (a). After that, the plasma region expands with a blast wave propagation, and the electron number density becomes lower. Therefore, more laser energy reaches the plasma region inside of blast wave, and the ionization wave propagates toward the direction of laser irradiation as indicated in figure 5 (b). From $t = 3 \mu\text{s}$ to $6 \mu\text{s}$, as indicated by figure 5 (c) and (d), the flowfield absorbs a little energy with low incident laser intensity as shown in figure 3. The total energy absorbed in the flowfield is 3.1 J, or 31 % of the total laser energy.

Figure 6 shows numerically and experimentally obtained results of the propagation speeds of the shock and ionization wave toward the direction of the incident laser pulse. We can find that

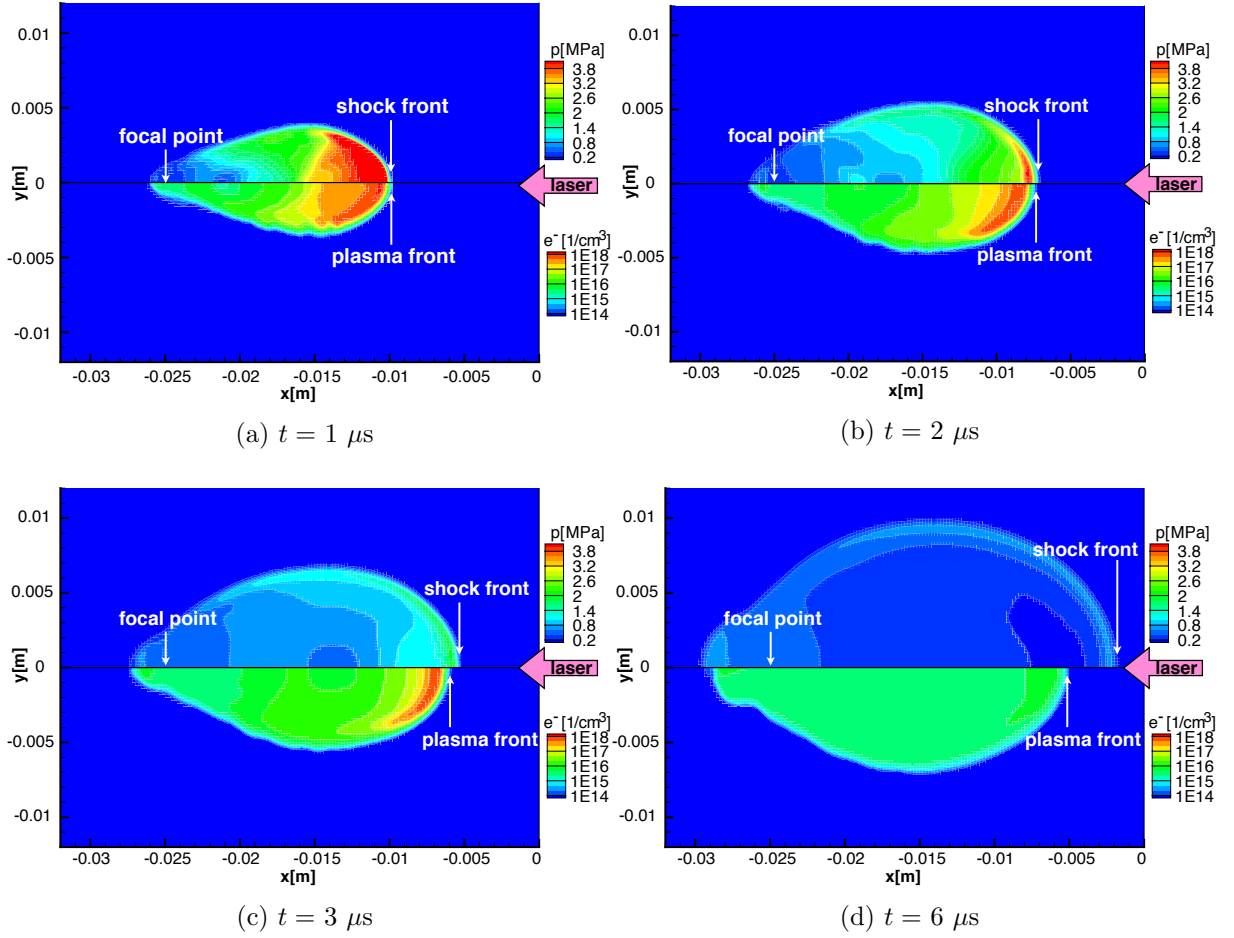


Figure 5. Time evolution of the pressure (upper) and the electron number density (lower) contours after the laser beam irradiation.

the speeds of the shock wave and the plasma front propagation agree well with experimental results except at an early stage of rapid expansion lasting until $t \simeq 0.4 \mu\text{s}$. Moreover, each transition time from the configuration of laser-supported detonation (LSD) to laser-supported combustion (LSC) is almost the same. Additionally, a flowfield simulation with a gray gas radiation is also conducted to compare with the derived radiative cooling formula. Note that the computed optical depth along the symmetric axis at $3 \mu\text{s}$ after the irradiation of the laser pulse is about 0.1. Then some reabsorption of radiation emitted from the other site in the plasma region occurs but is neglected in this work. For the gray gas radiation $Q_{\text{rad}} \equiv \kappa_p \sigma T_e^4$, the Planck mean absorption coefficient κ_p for equilibrium air plasma estimated by Taylor and Ali [32] is employed with the necessary modification of setting the values of κ_p to values ten times smaller than those of the original model, because their model is developed by assuming plasma of higher density (total number density $N \simeq 10^{19}/\text{cm}^3$) with respect to the density of the laser-induced plasma ($N \simeq 10^{18}/\text{cm}^3$). From the comparison of the non-LTE radiative cooling formula with the modified gray gas radiation, we see that the displacement of the plasma front is slightly different due to the deviation of population probabilities in each electronic excitation state, since the air density is high enough to achieve LTE as indicated in figure 1. The gray gas radiation model derived from LTE population could overestimate the radiative energy loss; therefore, we should use the curve fitted formula for radiative cooling with CR population to

make reliable computational predictions.

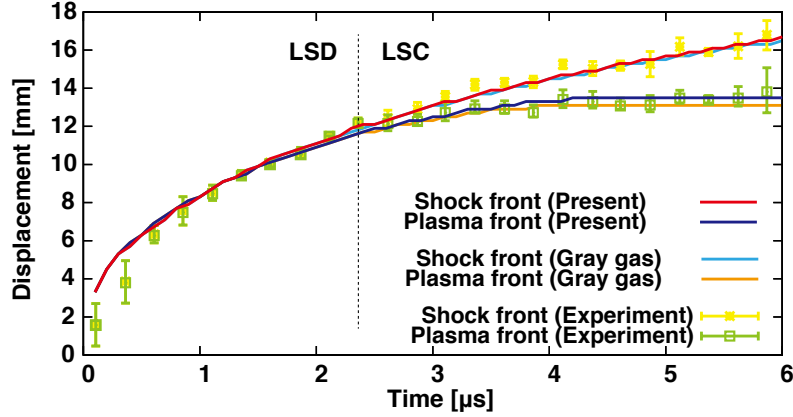


Figure 6. Comparison of numerically and experimentally obtained $x-t$ diagrams of the shock and plasma front toward the direction of the incident laser pulse.

4. Conclusion

A fitting formula for radiative cooling with CR population for air plasma flowfield has been developed to estimate the radiative cooling rate with higher accuracy and lower computational cost. Population number densities were calculated from CR rate equations in order to evaluate the effects of nonequilibrium atomic and molecular processes. The CR model consists of fifteen species: e^- , N, N^+ , N^{2+} , O, O^+ , O^{2+} , O^- , N_2 , N_2^+ , NO, NO^+ , O_2 , O_2^+ , and O_2^- with their major electronic excited states. Many elementary processes are integrated and applied to optically-thin plasmas in the number density range of $10^{12}/\text{cm}^3 \leq N \leq 10^{19}/\text{cm}^3$ and the temperature range of $300 \text{ K} \leq T \leq 40,000 \text{ K}$.

We have investigated the appropriate conditions for CRSS, LTE, and CE plasmas in the total number density range from $10^{12}/\text{cm}^3$ to $10^{19}/\text{cm}^3$ and at temperatures in the range from 5,000 K to 40,000 K. The results demonstrate that the LTE assumption is valid when the total number density exceeds $10^{17}/\text{cm}^3$. We should employ the CRSS model to analyze low-density plasmas in engineering applications.

Our results of the total radiative emissivity calculated from the CR population are fitted in terms of temperature and total number density. To validate this fitted radiation cooling formula, we have also computed a laser-induced blast wave propagation with radiative energy loss, and compared with experimentally observed shock and plasma front. We could fairly reproduce the thermochemical nonequilibrium flowfield for the blast wave induced by a pulse laser heating. From the comparison of the fitted radiative cooling formula with the corrected gray gas radiation assuming LTE, the displacement of the plasma front was slightly different due to the deviation of population probabilities in each electronic excitation state. The radiative cooling formula with CR model is an improvement on the gray gas radiation with nonequilibrium properties of population densities. Therefore, we can easily and appropriately evaluate the radiative energy loss by utilizing this fitting formula without solving detailed CR rate equations.

Acknowledgments

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