Transport properties in a two-temperature plasma: Theory and application

V. Rat,[1] P. André,[2] J. Aubreton,[1] M. F. Elchinger,[1] P. Fauchais,[1] and A. Lefort[2]
[1]SPCTS, University of Limoges, 123 avenue A. Thomas, 87060 Limoges Cedex, France
[2]LAEPT, Blaise Pascal University, 24 avenue des Landais, 63177 Aubière Cedex, France
(Received 24 October 2000; revised manuscript received 17 January 2001; published 23 July 2001)

An alternate derivation of transport properties in a two-temperature plasma has been performed. Indeed, recent works have shown that the simplified theory of transport properties out of thermal equilibrium introduced by Devoto and then Bonnefoi, very often used in two-temperature modeling, is questionable and particularly does not work when calculating the combined diffusion coefficients of Murphy. Thus, in this paper, transport properties are derived without Bonnefoi's assumptions in a nonreactive two-temperature plasma, assuming chemical equilibrium is achieved. The electron kinetic temperature T_e is supposed to be different from that of heavy species T_h. Only elastic processes are considered in a collision-dominated plasma. The resolution of Boltzmann's equation, thanks to the Chapman-Enskog method, is used to calculate transport coefficients from sets of linear equations. The solution of these systems allows transport coefficients to be written as linear combinations of collision integrals, which take into account the interaction potential for a collision between two particles. These linear combinations are derived by extending the definition and the calculation of bracket integrals introduced by Chapman *et al.* to the thermal nonequilibrium case. The obtained results are rigorously the same as those of Hirschfelder *et al.* at thermal equilibrium. The derivation of diffusion velocity and heat flux shows the contribution of a new gradient, that of the temperature ratio $\theta = T_e/T_h$. An application is presented for a two-temperature argon plasma. First, it is shown that the two-temperature linear combinations of collision integrals are drastically modified with respect to equilibrium. Secondly, the two-temperature simplified theory of transport coefficients of Devoto and Bonnefoi underestimates the electron thermal conductivity with respect to the accurate value at $T_e = 20\,000$ K. Lastly, contrary to the simplified theory of transport coefficients, the diffusion coefficients satisfy the symmetry conditions. An example is given at $T_e = 6000$ K for different values of θ for the diffusion coefficient between electrons and heavy species $D_{e\text{-Ar}}$ as well as for that between argon atoms and argon ions $D_{\text{Ar-Ar}^+}$.

DOI: 10.1103/PhysRevE.64.026409 PACS number(s): 52.25.−b

I. INTRODUCTION

The wide variety of experimental devices that generate thermal plasmas, namely high-intensity arcs, thermal rf discharges, or microwave discharges [1], has allowed the development, for the past 30 years, of numerous applications, such as deposition, cutting, welding, surface modifications, heating (ladles or tundishes), extractive metallurgy, waste destruction, etc. [2,3]. In spite of the great strides in the development of plasma processes, the growth of these technologies, as stated by Pfender for plasma spraying [4], has been "relatively slow for two reasons: there is still a lack of a solid engineering base in terms of control, reproductibility and optimization of thermal processes; and there is only a limited range of applications that appear to be economically viable, based on present technology." That is why, since the 1960s, many efforts have been devoted to modeling that, backed by measurements, have not only enhanced our knowledge base but also have made and should make essential contributions towards removing sources of the previously mentioned obstacles that hamper the growth of this technology. To simulate the plasma flow within the current-carrying area or the plasma jet, knowledge of thermodynamic and transport properties is a prerequisite. However, the reliability of these simulations is strongly dependent on the accuracy of these data, especially the transport coefficients being used.

The theory of transport properties of nonreactive gases at thermal equilibrium is based on the resolution of Boltzmann's equation due to the Chapman-Enskog method. The distribution function of different species is assumed to be a Maxwellian distribution function perturbed by a first-order perturbation function. The latter is developed in the form of a series of Sonine polynomials, and is used to express, according to the chosen approximation order, the transport coefficients as determinants depending on collision integrals taking into account the interaction potential between two colliding species.

Since the 1960s, thermal and electrical conductivities, and viscosity, have been widely calculated at thermal equilibrium for pure or binary nonreactive plasma-forming gases (see, for example, [5–10]) and more recently for reactive gases [11]. Such equilibrium calculations have allowed a better understanding of the plasma flow neglecting diffusion phenomena. The difficulty with diffusion is the number of coefficients to be considered: $\frac{1}{2}K(K-1)$ ordinary diffusion coefficients D_{ij} and $(K-1)$ thermal diffusion coefficients D_1^T, K being the number of species to be considered in the plasma. For example, in a binary mixture of N_2 and H_2, at least 10 species have to be considered. Diffusion was taken into account only in the 1990s thanks to the combined diffusion coefficients of Murphy [12]. He has used the properties of the diffusion coefficients, $D_{ij} = D_{ji}$ and $D_j^T = -D_j^T$, in a binary mixture to describe the diffusive mixing of two nonreactive ionized gases A and B at thermal equilibrium, and he defined combined diffusion coefficients $\overline{D_{AB}^x}$, $\overline{D_{AB}^T}$, and $\overline{D_{AB}^p}$, respectively, due to concentration, temperature, and pressure gradients (depending on D_{ij} and D_i^T). This method has allowed him to explain demixing processes in an Ar-H_2 free-burning arc [13,14]. Similar calculations were also performed for an Ar-N_2 free-burning arc [15], showing that demixing causes a significant increase in the nitrogen mass fraction in the central region out to a radius of 1 mm.

However, during the past two decades, with the growing development of spectroscopic or laser scattering techniques for plasma diagnostics, it has been shown that in thermal plasmas used in the different processes, nonequilibrium is more the rule than the exception. The electron kinetic temperature T_e can be different from that of heavy species T_h because of, on the one hand, the huge mass difference between these two kinds of particles and, on the other hand, the high mobility of electrons in an electric field compared to that of heavy species. The fraction of kinetic energy exchanged during a collision between an electron and a heavy species is so weak that an equilibrium state with two temperatures can be defined when collisions are not too numerous. In such a case, the two-temperature plasma composition and the related thermodynamic functions can be obtained [16–20].

For example, Farmer *et al.* [21] have presented evidence for departures from local thermodynamical equilibrium (LTE) in the region near the cathode in an argon free-burning arc. Haidar [22] has also noted this situation as well as Tanaka *et al.* [23]. According to these authors, deviations from LTE are due to the cold gas, which penetrates by the Maecker effect into the arc plasma. Bouaziz *et al.* [24] have also shown the departure from LTE in the vicinity of the anode of an argon-transferred arc for cutting or welding applications. In general, in the dc arc electrode vicinity, nonequilibrium always prevails [25]. André *et al.* [26] have shown by spectroscopic analysis that nonequilibrium also exists in an inductively coupled plasma torch working at less than a *kW* supplied power level with argon as a plasma-forming gas. Chen *et al.* [27] have put the emphasis on the diffusion of electrons from the core of a plasma jet to its fringes, resulting in a strong departure from LTE in this region. To conclude from the few presented examples, many authors have highlighted departures from LTE in very different experimental conditions.

To predict mass, energy, and momentum transfer in plasmas out of thermal equilibrium, experimental studies are usually combined with modeling. Nevertheless, the numerical data, which give two-temperature transport coefficients (excluding diffusion), used to solve the mass, energy, and momentum equations, are very rare. Indeed, in 1981, Chen *et al.* [28] have recognized the lack in the literature of two-temperature reliable transport coefficients and used approximate data for a two-temperature modeling of the anode contraction in an atmospheric pressure argon plasma. Fifteen years later, Charrada *et al.* [29] calculated electrical and thermal conductivities and viscosity from data given by Hirschfelder *et al.* [5] at thermal equilibrium that they have adapted to the two-temperature modeling of a mercury high-pressure plasma. Jenista *et al.* [30], developing a two-temperature modeling of the anode region in high-current electric arcs, have used the expressions of Spitzer-Härm [31], giving the electrical and thermal conductivities, as well as the thermal binary diffusion coefficients. However, these formulas have been derived at thermal equilibrium, in the 1950s, for a fully ionized plasma [32]. In his very recent nonequilibrium modeling of transferred arcs, Haidar [33] has used the two-temperature transport coefficients of Devoto [34,35]. Also very recently, Gonzales *et al.* [36] have presented a two-temperature modeling high-voltage SF6 circuit breaker using also transport coefficients calculated from Devoto's work. Thus, many models use either very crude approximations of two-temperature transport coefficients, often adapted from equilibrium calculations, or the only available two-temperature theory of transport properties, which is that of Devoto [34,35].

Devoto [34,35] has derived the transport coefficients in a two-temperature plasma starting from a simplified kinetic approach. He has assumed that the distribution function of heavy species is weakly affected by electrons during collisions so that the change, during collision, of the first-order perturbation function of heavy species is much smaller than that of electrons. He has then obtained simplified expressions for the different fluxes of mass, momentum, and energy, which has allowed us to separate the calculation of transport coefficients of electrons and heavy species. However, Bonnefoi [37,38] has shown that the definition given by Devoto of the vector $\vec{d}_i$ including the driving forces for diffusion did not check the closure relationship $\Sigma_{i=1}^{N}\vec{d}_i=\vec{0}$ in the description of the two-temperature plasma. He has developed an adapted formalism to this alternate definition, but stays in the simplified kinetic approach. It has to be noted that the transport coefficients of Devoto have been applied at thermal equilibrium as well as out of thermal equilibrium since the assumption of the large mass difference between electrons and heavy particles always stands. Indeed, transport coefficients of electrons have only to be affected by the electron temperature T_e, whereas those of heavy species by heavy species have to be affected by the temperature T_h because of the uncoupling between electrons and heavy species. The ease of use of Devoto's formulas explains why so many models use his theory. It seems that Devoto's assumptions are valid at thermal equilibrium since a good agreement is observed when comparing thermal conductivity and viscosity coefficient calculated from the complete expressions and those from the simplified ones [35].

Nevertheless, as soon as diffusion is taken into account in a two-temperature model, it has been shown [39] that the application of combined diffusion coefficients as defined by Murphy at equilibrium [12] and calculated using Devoto's simplified theory leads to unphysical results even when T_e tends towards T_h. When using the simplified theory of transport coefficients of Devoto, the symmetry relationships $\overline{D_{AB}^x}=\overline{D_{BA}^x}$ and $\overline{D_{AB}^T}=-\overline{D_{BA}^T}$ are not fulfilled when the ionization degree exceeds 10% [39]. This simplified theory is therefore not adapted to account for diffusion in a multitemperature plasma.

This paper is aimed at deriving diffusion coefficients, thermal conductivity, and viscosity coefficients in a two-temperature plasma, that is, when the kinetic temperature of electrons T_e is different from that of heavy species T_h, without Devoto's assumptions. The developed theory demonstrates, at least for argon, for which calculations have been performed, that Devoto's theory underestimates the thermal conductivity. Two-temperature diffusion coefficients, which to our knowledge no theory provides with accuracy, will allow us to derive in particular two-temperature combined diffusion coefficients used to describe demixing in free-burning arcs [13], which is neglected by most two-temperature models. Indeed, Murphy *et al.* [40], comparing spectroscopic measurement and an equilibrium numerical model that takes into account diffusion, have shown that two-temperature demixing cannot be a negligible process. The numerical model underestimates by almost 50% the hydrogen mass fraction with respect to measurements on the axis at 1 mm below the cathode in a region where departures from LTE prevail. Murphy *et al.* [40] attribute this discrepancy in part to transport coefficients that should be calculated in nonequilibrium conditions. It has to be noted that Jansen [41] has taken into account diffusion processes based on Stefan-Maxwell equations in a two-temperature plasma [42] in a numerical simulation model for a hydrogen-cascaded arc plasma. However, in this hydrodynamic approach, the linear system of equations for diffusive mass fluxes with respect to the mass-averaged velocity of the mixture is not solved exactly and an effective binary diffusion approximation is used. Kolesnikov *et al.* [43,44] have presented an accurate self-consistent set of Stefan-Maxwell equations for multicomponent diffusion in a two-temperature plasma, but, unfortunately, no numerical result has been displayed to our knowledge. Moreover, when defining the combined diffusion coefficients, Murphy has used diffusion coefficients obtained from the resolution of Boltzmann's equation rather than those resulting from the hydrodynamic approach.

The presented derivation is based on the resolution of Boltzmann's equation of each species in a nonreactive mixture due to the Chapman-Enskog method [6]. It is assumed that the electron temperature T_e is higher than that of heavy species T_h, their difference being characterized by their ratio $\theta = T_e/T_h$.

The chemical state of the plasma is frozen (no chemical reaction between the different species) and the chemical equilibrium is supposed to be achieved. The composition of the two-temperature plasma is supposed to be known even though the disagreement between authors [17,45–51] dealing with the generalized Saha equation, the Gibbs free-energy minimization, or the calculation kinetic method of composition highlights the difficulty of defining the "equilibrium state" of a two-temperature plasma.

Moreover, the energy transfers are limited to elastic collisions in a collision-dominated plasma. The "elastic" Knudsen number is much smaller than unity, regardless of the considered species in the mixture. Radiation is not considered.

First, we will present the resolution of Boltzmann's equation due to the Chapman-Enskog method adapted to nonequilibrium plasma and leading to the introduction of sets of linear equations allowing us to calculate the distribution functions. The derivation of diffusion velocity, heat flux, and tensor of pressure will be performed. It will be shown that the derivation of diffusion velocity as well as that of heat flux introduces a new gradient due to the temperature ratio $\theta = T_e/T_h$ and that two-temperature diffusion coefficients between electrons and heavy species can be defined contrary to the simplified theory of Devoto.

The results of derivations of bracket integrals, indispensable for the calculation of transport coefficients out of thermal equilibrium, will also be presented, as well as the definition of collision integrals introducing a collision effective temperature T^*_{ij} between electrons and heavy species.

To validate this theory, an application is given for a two-temperature argon plasma, often used in experimental devices as shown previously. Accurate two-temperature transport coefficients are calculated. Important discrepancies between our results and those of the simplified theory of Devoto are obtained, showing then that the latter cannot be applied in two-temperature plasma.

Appendix A defines mathematical notations used in the following expansions. Appendix B gives more details about the calculations of bracket integrals.

II. RESOLUTION OF THE BOLTZMANN'S EQUATION

A. Preliminary definitions

If t is the time, m_i, $\vec{r}$, and $\vec{v}_i$ are, respectively, the mass, the space vector, and the velocity of a particle of the ith species, $\vec{F}_i$ is the external force acting on it, and $f_i(\vec{r},\vec{v}_i,t)$ is the distribution function of this species, the following definitions are adopted.

The index 1 is for electrons,

$$n_i = \int f_i(\vec{r},\vec{v}_i,t)\, d\vec{v}_i, \tag{2.1}$$

n_i being the number density of the ith species;

$$\overline{\vec{v}_i} = \frac{1}{n_i}\int \vec{v}_i f_i(\vec{r},\vec{v}_i,t)\, d\vec{v}_i, \tag{2.2}$$

$\overline{\vec{v}_i}$ being the mean velocity of the ith species.

If the mixture contains N different species, the mass average velocity $\vec{v}_0$ is defined as follows:

$$\vec{v}_0 = \frac{1}{\rho}\sum_{i=1}^{N} n_i m_i \overline{\vec{v}_i}, \tag{2.3}$$

ρ being the density of the plasma, $\rho = \Sigma_{i=1}^{N}\rho_i$, with $\rho_i = n_i m_i$ the density of the ith species.

The peculiar and the reduced velocities defined for the temperature T_i are, respectively, written as

$$\vec{V}_i = \vec{v}_i - \vec{v}_0, \tag{2.4}$$

$$\vec{W}_i = \left(\frac{m_i}{2kT_i}\right)^{1/2}\vec{V}_i. \tag{2.5}$$

The total pressure p of the plasma, assumed to be a perfect gas, is defined according to Dalton's law, where k is Boltzmann's constant,

$$p=\sum_{i=1}^{N} p_i, \tag{2.6}$$

where

$$p_1=n_1 k T_e, \tag{2.7}$$

$$p_i=n_i k T_h \quad (i\geqslant 2). \tag{2.8}$$

p_1 and p_i $(i\geqslant 2)$ are, respectively, the partial pressures of electrons and heavy species. The following operator is also introduced:

$$\frac{D}{Dt}=\frac{\partial}{\partial t}+\vec{v}_i\cdot\vec{\nabla}+\frac{\vec{F}_i}{m_i}\cdot\vec{\nabla}_{\vec{v}_i}. \tag{2.9}$$

In many of the following developments, the temperature T_i will be associated to the ith species. However, in the realistic case of a two-temperature plasma, $T_i=T_e$ if $i=1$ and $T_i=T_h$ if $i\geqslant 2$.

B. Resolution of Boltzmann's equation

The resolution of Boltzmann's equation is based on the Chapman-Enskog expansion [6].

1. Subdivision of Boltzmann's equation

The distribution function f_i of the ith species is the solution of the integrodifferential equation of Boltzmann [5]:

$$\frac{Df_i}{Dt}=\sum_{j=1}^{N}\int\int\int (f_i' f_j' - f_i f_j) g b\, db\, d\varepsilon\, d\vec{v}_j. \tag{2.10}$$

f_i' is the distribution function after collision of the ith species, g is the relative velocity of the species i and j, and b and ε are, respectively, the impact parameter and the incidence azimuthal angle.

The mixture can be characterized by the knowledge of the unknowns n_i, $\vec{v}_0$, T_e, and T_h by solving the equations of continuity, momentum, and energy obtained from Boltzmann's equation. These equations of change are derived according to the chosen approximation order for the distribution function of different species. When inserting a Maxwellian distribution function, which corresponds to the zero-order approximation, into the equations of change, the obtained Euler equations can be solved and each flux of molecular properties vanishes. If the first-order approximation of the distribution function is introduced into the equations of change, the Navier-Stokes equations are obtained. This system then requires the calculation of transport coefficients to be solved. The equations of change out of thermal equilibrium have been derived in Ref. [37].

Thus, it will be assumed that the zero-order approximation function is Maxwellian at T_e for electrons and T_h for heavy species. The distribution function of the ith species, the solution of Eq. (2.10), is approximated by a Maxwellian distribution function $f_i^{(0)}$ perturbed by Φ_i such as $|\Phi_i|\ll 1$, which is the first-order perturbation function of the ith species with the temperature T_i:

$$f_i=f_i^{(0)}(1+\Phi_i). \tag{2.11}$$

Inserting Eq. (2.11) into Eq. (2.10), it can be shown that

$$\frac{Df_i^{(0)}}{Dt}=I_i^{(0)}+\sum_{j=1}^{N}\int\int\int f_i^{(0)} f_j^{(0)}[(\Phi_i'+\Phi_j')K_i-\Phi_i-\Phi_j] \times g b\, db\, d\varepsilon\, d\vec{v}_j. \tag{2.12}$$

According to the Chapman-Enskog method, whose the assumptions are recalled, for instance, in Ref. [52], the Maxwellian $f_i^{(0)}$ and the unknowns Φ_i are assumed to vary slowly in space and time over a distance of a mean free path and over the time of a mean free flight. This assumption justifies, in a first approximation, neglecting the derivatives of Φ_i as well as of products of Φ_i with derivatives of $f_i^{(0)}$ on the left-hand side of Eq. (2.12).

$I_i^{(0)}$ represents the zero-order approximation of Chapman-Enskog's expansion and does not vanish for two colliding particles with different temperatures:

$$I_i^{(0)}=\sum_{j=1}^{N}\int\int\int (f_i'^{(0)} f_j'^{(0)} - f_i^{(0)} f_j^{(0)}) g b\, db\, d\varepsilon\, d\vec{v}_j. \tag{2.13}$$

$I_i^{(0)}$ will be introduced into the calculation of the left-hand side of Eq. (2.12).

A term $K_i(W_i,\theta_{ij})$, taking into account the thermal nonequilibrium when electrons and heavy species collide, has been introduced as follows:

$$f_i'^{(0)} f_j'^{(0)} = f_i^{(0)} f_j^{(0)} K_i(W_i,\theta_{ij}) \quad \forall \ \ i,j\in[1;N], \tag{2.14}$$

where

$$K_i(W_i,\theta_{ij})=\exp(-(W_i'^2-W_i^2)(1-\theta_{ij})) \tag{2.15}$$

with

$$\theta_{ij}=\frac{T_i}{T_j} \tag{2.16}$$

and W_i' is the reduced velocity after collision.

However, it has to be noted that $K_i=K_j$ and, of course, when two species with the same temperature collide, $K_i=1$. The introduction of K_i allows the definition of bracket integrals to be generalized out of thermal equilibrium.

The calculation of $Df_i^{(0)}/Dt$ and $Df_i^{(0)}/Dt$ $(i\geqslant 2)$ is obtained using the equations of change [37].

For electrons, it is shown that

$$\frac{Df_1^{(0)}}{Dt}=f_1^{(0)}\Bigg[(W_1^2-\tfrac{5}{2})\vec{V}_1\cdot\vec{\nabla}\ln T_e+2\vec{W}_1^{\mathrm{o}}\vec{W}_1:\vec{\nabla}\vec{v}_0+\frac{Q_1^{(0)}}{n_1kT_e}(\tfrac{2}{3}W_1^2-1)+\frac{\vec{V}_1}{n_1kT_e}\cdot\left(\frac{\rho_1}{\rho}\sum_{j=1}^{N}n_j\vec{F}_j-n_1\vec{F}_1-\frac{\rho_1}{\rho}\vec{\nabla}p+\vec{\nabla}p_1\right)\Bigg].\tag{2.17}$$

For heavy species ($\forall i\geqslant 2$), it is shown that

$$\frac{Df_i^{(0)}}{Dt}=f_i^{(0)}\Bigg[(W_i^2-\tfrac{5}{2})\vec{V}_i\cdot\vec{\nabla}\ln T_h+2\vec{W}_i^{\mathrm{o}}\vec{W}_i:\vec{\nabla}\vec{v}_0-\frac{Q_1^{(0)}}{(n-n_1)kT_h}(\tfrac{2}{3}W_i^2-1)+\frac{\vec{V}_i}{n_ikT_h}\cdot\left(\frac{\rho_i}{\rho}\sum_{j=1}^{N}n_j\vec{F}_j-n_i\vec{F}_i-\frac{\rho_i}{\rho}\vec{\nabla}p+\vec{\nabla}p_i\right)\Bigg],\tag{2.18}$$

where

$$Q_1^{(0)}=\sum_{j=2}^{N}\int\int\int\int\tfrac{1}{2}m_1\times(V_1'^2-V_1^2)f_1^{(0)}f_j^{(0)}gb\,db\,d\varepsilon\,d\vec{v}_i d\vec{v}_j.\tag{2.19}$$

$Q_1^{(0)}$ corresponds to the exchanged kinetic energy between electrons and heavy species during collisions. It can be calculated using variable changes similar to those given in Appendix B. Neglecting the terms such as m_1/m_i ($i\geqslant 2$), it can be shown that

$$Q_1^{(0)}=4kn_1(T_h-T_e)\left(\frac{8kT_e}{\pi m_1}\right)^{1/2}\sum_{j=2}^{N}n_j\frac{m_1}{m_j}\overline{Q}_{1j}^{(1,1)},\tag{2.20}$$

$\overline{Q}_{1j}^{(1,1)}$ being a collision integral defined in [37]. $\vec{W}_i^{\mathrm{o}}\vec{W}_i$ is defined in Appendix A.

However, Eqs. (2.17) and (2.18) are modified as follows to take into account the coupling between electrons and heavy species.

It is assumed that the thermal nonequilibrium defined by the ratio $\theta=T_e/T_h$ depends on space vector $\vec{r}$, that is, $\theta=\theta(\vec{r})$.

Hence,

$$\vec{\nabla}\ln T_e=\vec{\nabla}\ln T_h+\vec{\nabla}\ln\theta.\tag{2.21}$$

Moreover, the total pressure p is written as

$$p=n_1kT_e+\sum_{i=2}^{N}n_ikT_h\tag{2.22}$$

or

$$kT_h=\frac{p}{n[1+x_1(\theta-1)]}\tag{2.23}$$

with

$$n=\sum_{i=1}^{N}n_i.\tag{2.24}$$

x_i is the molar fraction of the species i. The molar fractions obviously check

$$\sum_{i=1}^{N}x_i=1.\tag{2.25}$$

So, the partial pressure of electrons p_1 is written as

$$p_1=n_1k\theta T_h=\frac{x_1\theta p}{1+x_1(\theta-1)}.\tag{2.26}$$

Thus, it can be shown that

$$\vec{\nabla}p_1=\frac{\theta p}{D^2}\vec{\nabla}x_1+\frac{x_1\theta}{D}\vec{\nabla}p+\frac{x_1p(1-x_1)}{D^2}\vec{\nabla}\theta,\tag{2.27}$$

where

$$D=1+x_1(\theta-1).\tag{2.28}$$

The partial pressure p_i ($i\geqslant 2$) of the ith heavy species is also written as

$$p_i=n_ikT_h=\frac{x_ip}{1+x_1(\theta-1)},\tag{2.29}$$

therefore

$$\vec{\nabla}p_i=\frac{p}{D}\vec{\nabla}x_i-\frac{x_ip(\theta-1)}{D^2}\vec{\nabla}x_1+\frac{x_i}{D}\vec{\nabla}p-\frac{x_ix_1p}{D^2}\vec{\nabla}\theta.\tag{2.30}$$

Using Eq. (2.21) and substituting Eqs. (2.27) and (2.30), respectively, into Eqs. (2.17) and (2.18), we get the following using $I_i^{(0)}$. For electrons,

$$\frac{Df_1^{(0)}}{Dt}-I_1^{(0)}=f_1^{(0)}\Bigg[(W_1^2-\tfrac{5}{2})(\vec{V}_1\cdot\vec{\nabla}\ln T_h)+2\vec{W}_1^{\mathrm{o}}\vec{W}_1:\vec{\nabla}\vec{v}_0+\frac{\vec{V}_1}{n_1kT_e}\cdot\vec{d}_1+\frac{g_1}{n_1kT_h}\vec{V}_1\cdot\vec{\nabla}\ln\theta+(W_1^2-\tfrac{5}{2})\vec{V}_1\cdot\vec{\nabla}\ln\theta\Bigg]+Q_1^{(0)}\left[\frac{f_1^{(0)}}{n_1kT_e}(\tfrac{2}{3}W_1^2-1)-\frac{I_1^{(0)}}{Q_1^{(0)}}\right],\tag{2.31}$$

where

$$\vec{d}_1=\frac{\rho_1}{\rho}\sum_{j=1}^{N}n_j\vec{F}_j-n_1\vec{F}_1+\left(\frac{x_1\theta}{D}-\frac{\rho_1}{\rho}\right)\vec{\nabla}p+\frac{\theta p}{D^2}\vec{\nabla}x_1\tag{2.32}$$

and

$$g_1=\frac{x_1 p(1-x_1)}{D^2}. \quad (2.33)$$

For heavy species ($i\geqslant 2$),

$$\frac{Df_i^{(0)}}{Dt}-I_i^{(0)}=f_i^{(0)}\Bigg[(W_i^2-\tfrac{5}{2})(\vec{V}_i\cdot\vec{\nabla}\ln T_h)+2\vec{W}_i^{\circ}\vec{W}_i:\vec{\nabla}\vec{v}_0+\frac{\vec{V}_i}{n_ikT_h}\cdot\vec{d}_i+\frac{\theta g_i}{n_ikT_h}\vec{V}_i\cdot\vec{\nabla}\ln\theta\Bigg]+Q_1^{(0)}\times\Bigg[\frac{f_i^{(0)}}{(n-n_1)kT_h}(1-\tfrac{2}{3}W_i^2)-\frac{I_i^{(0)}}{Q_1^{(0)}}\Bigg], \quad (2.34)$$

where

$$\vec{d}_i=\frac{\rho_i}{\rho}\sum_{j=1}^{N}n_j\vec{F}_j-n_i\vec{F}_i+\left(\frac{x_i}{D}-\frac{\rho_i}{\rho}\right)\vec{\nabla}p+\frac{p}{D}\vec{\nabla}x_i-\frac{x_i(\theta-1)p}{D^2}\vec{\nabla}x_1 \quad (2.35)$$

and

$$g_i=-\frac{x_ix_1p}{D^2}. \quad (2.36)$$

With these definitions, it is checked that

$$\sum_{i=1}^{N}\vec{d}_i=\vec{0} \quad (2.37)$$

and

$$\sum_{i=1}^{N}g_i=0. \quad (2.38)$$

The transport terms, Eqs. (2.31) and (2.34), show that transport phenomena are due to a new gradient $\nabla\ln\theta$, which characterizes the temperature difference between electrons and heavy species, the heavy species temperature gradient, the velocity gradient, external forces (forced diffusion), the concentration and pressure gradients, and a term acting on the hydrostatic pressure to the first-order approximation of Sonine polynomials. The introduction of $\nabla\ln\theta$ allows the exchange of transport properties (mass, momentum, and energy) between the subsystems consisting of electrons and heavy species, which are not isolated from each other.

From a calculation point of view, the introduction of $\nabla\ln\theta$, allows us to consider the coupling between electrons and heavy species in the resolution of systems of linear equations, which give the transport coefficients.

2. Presumed form of the first-order perturbation function

According to relationships (2.31) and (2.34), a general form of $\Phi_i(\forall i)$ in Eq. (2.12) can be presumed such that

$$\Phi_i=-\vec{A}_i\cdot\vec{\nabla}\ln T_h-\vec{\vec{B}}_i:\vec{\nabla}\vec{v}_0+\sum_{j=1}^{N}\vec{C}_i^j\cdot\vec{d}_j+D_iQ_1^{(0)}+\sum_{j=1}^{N}\vec{E}_i^j\cdot g_j\vec{\nabla}\ln\theta-\vec{F}_i\cdot\vec{\nabla}\ln\theta. \quad (2.39)$$

The previous unknowns Φ_i are replaced by the new ones: $\vec{A}_i$, $\vec{\vec{B}}_i$, $\vec{C}_i^j$, D_i, $\vec{E}_i^j$, and $\vec{F}_i$. It has to be noted that $\vec{A}_i$, $\vec{C}_i^j$, $\vec{E}_i^j$, and $\vec{F}_i$ are vectors, $\vec{\vec{B}}_i$ are second-order tensors, and D_i are scalars.

Substituting Eq. (2.39) into Eq. (2.12) and identifying the terms in factors of $\vec{\nabla}\ln T_h$, $\vec{\nabla}\vec{v}_0$, $\vec{d}_j$, $Q_1^{(0)}$, $g_j\vec{\nabla}\ln\theta$, and $\vec{\nabla}\ln\theta$ in the left- and right-hand side due to Eqs. (2.31) and (2.34), a general formulation can be defined as follows:

$$\overline{\overline{R_i^{(h,k)}}}=\sum_{j=1}^{N}\int\int\int f_i^{(0)}f_j^{(0)}\big((\overline{\overline{T_i'^{(h,k)}}}+\overline{\overline{T_j'^{(h,k)}}})K_i(W_i,\theta_{ij})-\overline{\overline{T_i^{(h,k)}}}-\overline{\overline{T_j^{(h,k)}}}\big)gb\,db\,d\varepsilon\,d\vec{v}_j, \quad (2.40)$$

TABLE I. Definition of $\overline{\overline{R_1^{(h,k)}}}$, $\overline{\overline{R_i^{(h,k)}}}$, and $\overline{\overline{T_i^{(h,k)}}}$.

$\overline{\overline{R_1^{(h,k)}}}$	$\overline{\overline{R_i^{(h,k)}}}$ ($i\geqslant 2$)	$\overline{\overline{T_i^{(h,k)}}}$
$f_1^{(0)}(\frac{5}{2}-W_1^2)\vec{V}_1$	$f_i^{(0)}(\frac{5}{2}-W_i^2)\vec{V}_i$	$\vec{A}_i$
$-2f_1^{(0)}\vec{W}_1^{\circ}\vec{W}_1$	$-2f_i^{(0)}\vec{W}_i^{\circ}\vec{W}_i$	$\vec{\vec{B}}_i$
$f_1^{(0)}\frac{(\delta_{1h}-\delta_{1k})\vec{V}_1}{n_1kT_e}$	$f_i^{(0)}\frac{(\delta_{ih}-\delta_{ik})\vec{V}_i}{n_ikT_h}$	$\vec{C}_i^{hk}$
$\frac{f_1^{(0)}}{n_1kT_e}(\frac{2}{3}W_1^2-1)-\frac{I_1^{(0)}}{Q_1^{(0)}}$	$-\frac{f_i^{(0)}}{(n-n_1)kT_h}(\frac{2}{3}W_i^2-1)-\frac{I_i^{(0)}}{Q_1^{(0)}}$	D_i
$f_1^{(0)}\frac{(\delta_{1h}-\delta_{1k})\vec{V}_1}{n_1kT_h}$	$f_i^{(0)}\frac{\theta(\delta_{ih}-\delta_{ik})\vec{V}_i}{n_ikT_h}$	$\vec{E}_i^{hk}$
$f_1^{(0)}(\frac{5}{2}-W_1^2)\vec{V}_1$	0	$\vec{F}_i$

TABLE II. Definition of n, W_i, and $t_{ip}^{(h,k)}$. $\vec{U}$ is the second-order unity tensor.

$\overline{\overline{T_i^{(h,k)}}}$	n	$\overline{\overline{W}}_i$	$t_{ip}^{(h,k)}$
$\vec{A}_i$	$\frac{3}{2}$	$\vec{W}_i$	a_{ip}
$\vec{B}_i$	$\frac{5}{2}$	$\vec{W}_i^{\mathrm{o}}\vec{W}_i=\vec{W}_i\vec{W}_i-\frac{1}{3}W_i^2\vec{U}$	b_{ip}
$\vec{C}_i^{hk}$	$\frac{3}{2}$	$\vec{W}_i$	c_{ip}^{hk}
D_i	$\frac{1}{2}$	1	d_{ip}
$\vec{E}_i^{hk}$	$\frac{3}{2}$	$\vec{W}_i$	e_{ip}^{hk}
$\vec{F}_i$	$\frac{3}{2}$	$\vec{W}_i$	f_{ip}

where $\overline{\overline{T_i^{(h,k)}}}$ is a tensor, a vector, or a scalar. Table I gives the correspondence between Eqs. (2.40) and (2.12) after the previous substitutions. The unknown $\vec{C}_i^h$ and $\vec{E}_i^h$ have been replaced, respectively, as shown in [5], by $\vec{C}_i^h-\vec{C}_i^k$ or $\vec{C}_i^{hk}$ and $\vec{E}_i^{hk}$ taking into account Eqs. (2.37) and (2.38). The indexes h and k vary from 1 to N.

δ_{ij} are Kronecker symbols defined such as $\delta_{ij}=1$ if $i=j$, and $\delta_{ij}=0$ otherwise.

3. Auxiliary conditions

It is assumed that the unknowns $\overline{\overline{T_i^{(h,k)}}}$ are expressed as linear combination of Sonine polynomials $S_n^p(W_i^2)$ defined in [6] (ξ is the order of the expansion):

$$\overline{\overline{T_i^{(h,k)}}}=\overline{\overline{W}}_i\sum_{p=0}^{\xi-1} t_{ip}^{(h,k)}S_n^p(W_i^2), \tag{2.41}$$

where $\overline{\overline{W}}_i, t_{ip}^{(h,k)}$, and n are defined in Table II.

Auxiliary conditions have to be taken into account [6]:

$$\int f_i^{(0)}\Phi_i d\vec{v}_i=0, \tag{2.42}$$

$$\sum_{i=1}^{N} m_i\int \vec{v}_i f_i^{(0)}\Phi_i d\vec{v}_i=0, \tag{2.43}$$

$$\frac{1}{2}\sum_{i=1}^{N} m_i\int (\vec{v}_i-\vec{v}_0)^2 f_i^{(0)}\Phi_i d\vec{v}_i=0. \tag{2.44}$$

As a result, several constraints on the different unknowns are introduced according to Eq. (2.39):

$$\sum_{j=1}^{N} n_j(m_jT_j)^{1/2}t_{j0}^{(h,k)}=0, \tag{2.45}$$

where $t_{j0}^{(h,k)}$ is equal to a_{j0}, c_{j0}^{hk}, e_{j0}^{hk}, and f_{j0}. This constraint (2.45) takes into account the fact that the sum of mass fluxes has to vanish in the mixture. Moreover, we also get

$$d_{i0}=0, \quad \forall i, \tag{2.46}$$

$$\sum_{j=1}^{N} n_jT_jd_{j1}=0. \tag{2.47}$$

4. Introduction of systems of linear equations

Multiplying Eq. (2.40) by $\overline{\overline{W}}_iS_n^m(W_i^2)$, integrating over $\vec{v}_i$, and defining $R_{im}^{(h,k)}$ as

$$R_{im}^{(h,k)}=\int (\overline{\overline{R_i^{(h,k)}}}:\overline{\overline{W}}_i)S_n^m(W_i^2)d\vec{v}_i, \tag{2.48}$$

sets of linear equations for each unknown are introduced taking into account the previous constraints:

$$\sum_{j=1}^{N}\sum_{p=0}^{\xi-1} q_{ij}^{mp}t_{jp}^{(h,k)}=-r_{im}^{(h,k)}, \tag{2.49}$$

where

$$r_{im}^{(h,k)}=\left(\frac{2\pi m_i}{kT_i}\right)^{1/2}R_{im}^{(h,k)}, \tag{2.50}$$

$$q_{ij}^{mp}=\left(\frac{2\pi m_i}{kT_i}\right)^{1/2}\tilde{Q}_{ij}^{mp}, \tag{2.51}$$

and

$$\tilde{Q}_{ij}^{mp}=\begin{cases} Q_{ij}^{mp}-\dfrac{n_j(m_jT_j)^{1/2}}{n_i(m_iT_i)^{1/2}}Q_{ii}^{mp}\delta_{m0}\delta_{p0} & \text{if } t_{jp}^{hk}=a_{jp},\ c_{jp}^{hk},\ e_{jp}^{hk},\ \text{or } f_{jp} \\ Q_{ij}^{mp} & \text{if } t_{jp}^{hk}=b_{jp} \\ Q_{ij}^{mp}-\dfrac{n_jT_j}{n_iT_i}Q_{ii}^{mp}\delta_{m0}\delta_{p1} & \text{if } t_{jp}^{hk}=d_{jp} \end{cases}$$

with

$$Q_{ij}^{mp}=\sum_{\ell=1}^{N} n_in_\ell\{\delta_{ij}[\overline{\overline{W}}_iS_n^m(W_i^2);\overline{\overline{W}}_iS_n^p(W_i^2)]_{i\ell}+\delta_{j\ell}[\overline{\overline{W}}_iS_n^m(W_i^2);\overline{\overline{W}}_\ell S_n^p(W_\ell^2)]_{i\ell}\}. \tag{2.52}$$

Q_{ij}^{mp} are expressed as functions of bracket integrals that are generalized out of thermal equilibrium as follows. They are introduced with notations similar to those of Chapman *et al.* [6],

TABLE III. Calculation results of $r_{1m}^{(h,k)}$ and $r_{im}^{(h,k)}$.

t_{ip}	$r_{1m}^{(h,k)}$	$r_{im}^{(h,k)}$ $(i\geqslant 2)$
a_{ip}	$\frac{15}{2}\pi^{1/2}n_1\delta_{1m}$	$\frac{15}{2}\pi^{1/2}n_i\delta_{1m}$
b_{ip}	$-5n_1\left(\frac{2\pi m_1}{kT_e}\right)^{1/2}\delta_{0m}$	$-5n_i\left(\frac{2\pi m_i}{kT_h}\right)^{1/2}\delta_{0m}$
c_{ip}^{hk}	$3\pi^{1/2}\frac{(\delta_{1h}-\delta_{1k})\delta_{0m}}{kT_e}$	$3\pi^{1/2}\frac{(\delta_{ih}-\delta_{ik})\delta_{0m}}{kT_h}$
d_{ip}	$-\frac{(2\pi m_1)^{1/2}\delta_{1m}}{(kT_e)^{3/2}}-\frac{J_{1m}^{(0)}}{Q_1^{(0)}}\left(\frac{2\pi m_1}{kT_e}\right)^{1/2}$	$\frac{(2\pi m_i)^{1/2}n_i\delta_{1m}}{(n-n_1)(kT_h)^{3/2}}-\frac{J_{im}^{(0)}}{Q_1^{(0)}}\left(\frac{2\pi m_i}{kT_h}\right)^{1/2}$
e_{ip}^{hk}	$3\pi^{1/2}\frac{(\delta_{1h}-\delta_{1k})\delta_{0m}}{kT_h}$	$3\pi^{1/2}\frac{\theta(\delta_{ih}-\delta_{ik})\delta_{0m}}{kT_h}$
f_{ip}	$\frac{15}{2}\pi^{1/2}n_1\delta_{1m}$	0

$$[\overline{\overline{W}}_iS_n^m(W_i^2);\overline{\overline{W}}_iS_n^p(W_i^2)]_{ij}=\frac{1}{n_in_j}\int\int\int\int f_i^{(0)}f_j^{(0)}(\overline{\overline{W}}_iS_n^p(W_i^2)-\overline{\overline{W}}_i'S_n^p(W_i'^2)K_i(W_i,\theta_{ij})):\overline{\overline{W}}_iS_n^m(W_i^2)gb\,db\,d\varepsilon\,d\vec{v}_id\vec{v}_j, \tag{2.53}$$

$$[\overline{\overline{W}}_iS_n^m(W_i^2);\overline{\overline{W}}_jS_n^p(W_j^2)]_{ij}=\frac{1}{n_in_j}\int\int\int\int f_i^{(0)}f_j^{(0)}(\overline{\overline{W}}_jS_n^p(W_j^2)-\overline{\overline{W}}_j'S_n^p(W_j'^2)K_i(W_i,\theta_{ij})):\overline{\overline{W}}_iS_n^m(W_i^2)gb\,db\,d\varepsilon\,d\vec{v}_id\vec{v}_j. \tag{2.54}$$

For a collision between two particles belonging to the same species, $i=j$, $T_i=T_j$, and $K_i=1$, the definition is the same as that of Chapman *et al.* [6],

$$[\overline{\overline{W}}_iS_n^m(W_i^2);\overline{\overline{W}}_iS_n^p(W_i^2)]_i=\frac{1}{n_i^2}\int\int\int\int f_i^{(0)}f^{(0)}(\overline{\overline{W}}_iS_n^p(W_i^2)+\overline{\overline{W}}S_n^p(W^2)-\overline{\overline{W_i'}}S_n^p(W_i'^2)$$
$$-\overline{\overline{W'}}S_m^p(W'^2)):\overline{\overline{W}}_iS_n^m(W_i^2)gb\,db\,d\varepsilon\,d\vec{v}_id\vec{v}. \tag{2.55}$$

It has to be noted that the symmetry by changing functions in the bracket integral expressions is not conserved because of the presence of the term $K_i(W_i,\theta_{ij})$ when two species with different temperatures collide, that is, for two functions F_i and H_j,

$$[F_i;H_j]_{ij}\neq[H_j;F_i]_{ij}. \tag{2.56}$$

The values of $r_{im}^{(h,k)}$ are given in Table III.

$J_{im}^{(0)}$ is defined as

$$J_{im}^{(0)}=\int I_i^{(0)}S_{1/2}^m(W_i^2)d\vec{v}_i. \tag{2.57}$$

$J_{im}^{(0)}$ can be easily calculated following the expansions shown in Appendix B; its result is given therein.

III. TRANSPORT COEFFICIENTS

The diffusion velocity, heat flux, and pressure tensor are introduced in the usual form due to the first-order perturbation function but taking into account the temperature ratio gradient. This new contribution will allow us to define new transport coefficients.

A. Diffusion coefficients in a multitemperature plasma

The diffusion velocity is given by

$$\bar{\vec{V}}_i=\frac{1}{n_i}\int\vec{V}_if_id\vec{V}_i. \tag{3.1}$$

Inserting $f_i=f_i^{(0)}(1+\Phi_i)$ due to Eq. (2.39) into Eq. (3.1), we get

$$\bar{\vec{V}}_i=\frac{n}{n_i\rho kT_i}\sum_{j=1}^{N}m_j(D_{ij}\vec{d}_j+D_{ij}^{\theta}g_j\vec{\nabla}\ln\theta)-\frac{D_i^T}{n_im_i}\vec{\nabla}\ln T_h$$
$$-\frac{D_i^{\theta*}}{n_im_i}\vec{\nabla}\ln\theta, \tag{3.2}$$

where

$$D_{ij}=\frac{n_i\rho kT_i}{nm_j}\left(\frac{kT_i}{2m_i}\right)^{1/2}c_{i0}^{ji}, \tag{3.3}$$

$$D_{ij}^{\theta}=\frac{n_i\rho kT_i}{nm_j}\left(\frac{kT_i}{2m_i}\right)^{1/2}e_{i0}^{ji}, \tag{3.4}$$

$$D_i^T=n_im_i\left(\frac{kT_i}{2m_i}\right)^{1/2}a_{i0}, \tag{3.5}$$

and

$$D_i^{\theta *}=n_im_i\left(\frac{kT_i}{2m_i}\right)^{1/2}f_{i0} \tag{3.6}$$

with, of course, $T_i=T_e$ if $i=1$ and $T_i=T_h$ if $i\geqslant 2$. The different unknowns are determined by solving sets of linear equations defined in the preceding section.

The coefficients D_{ij} are the two-temperature ordinary diffusion coefficients, which the simplified theory of Devoto does not provide, and D_i^T are the two-temperature thermal diffusion coefficients. New thermal nonequilibrium diffusion coefficients D_{ij}^{θ} and $D_i^{\theta *}$ are introduced taking into account the temperature difference between electrons and heavy species and corresponding to diffusion due to the gradient of the temperature ratio $\theta=T_e/T_h$. It has to be noted that the diffusion velocity of electrons depends on the heavy species gradient. In fact, T_h can be considered as a reference temperature, the temperature difference T_e and T_h being completed by the introduction of $D_i^{\theta *}$ (see Table I). The diffusion coefficient D_{ij}^{θ} traduces a mass transfer in the mixture, which tends to eliminate the temperature difference between electrons and heavy species. Moreover, Eq. (3.2) can be expressed as a function of $\vec{\nabla}\ln T_e$ using the relation (2.21).

B. Translational thermal conductivity in a multitemperature plasma

The heat flux is given by

$$\vec{q}=\tfrac{1}{2}\sum_{j=1}^{N} m_j\int V_j^2\vec{V}_jf_jd\vec{V}_j. \tag{3.7}$$

Inserting the form of f_i, that is, $f_i=f_i^{(0)}(1+\Phi_i)$ due to Eq. (2.39), into Eq. (3.7), the heat flux is then given by

$$\vec{q}=\sum_{i=1}^{N}\left\{\tfrac{5}{2}kT_in_i\overline{\vec{V}_i}-\kappa_i'\vec{\nabla}T_h-\kappa_i'^{\,\theta}T_i\vec{\nabla}\ln\theta-\sum_{j=1}^{N}\frac{\kappa_{ij}^{D}}{n_jm_j}\vec{d}_j\right\}, \tag{3.8}$$

where

$$\kappa_i'=-\tfrac{5}{4}k\frac{T_i}{T_h}n_i\left(\frac{2kT_i}{m_i}\right)^{1/2}a_{i1}, \tag{3.9}$$

$$\kappa_i'^{\,\theta}=-\tfrac{5}{4}kn_i\left(\frac{2kT_i}{m_i}\right)^{1/2}\left(f_{i1}-\sum_{j=1}^{N}e_{i1}^{ji}g_j\right), \tag{3.10}$$

$$\kappa_{ji}^{D}=\tfrac{5}{4}n_in_jm_jkT_i\left(\frac{2kT_i}{m_i}\right)^{1/2}c_{i1}^{ji} \tag{3.11}$$

with $T_i=T_e$ if $i=1$ and $T_i=T_h$ if $i\geqslant 2$. κ_i' is the translational thermal conductivity of the ith species.

An alternate contribution due to the thermal nonequilibrium appears in Eq. (3.7) and its corresponding thermal conductivity κ_i^{θ} as shown in the derivation of diffusion velocity. The calculation is quite different from that performed at equilibrium because of the asymmetry of calculations introduced by the presence of different temperatures. It has to be noted that, in the expression of thermal conductivity of electrons κ_1', the term $\theta=T_e/T_h$ takes into account the fact that their heat flux is calculated with respect to the temperature gradient of heavy species $\vec{\nabla}T_h$.

The true translational thermal conductivity can also be defined by introducing the specific enthalpy h_i of the ith species [53] such that $\rho_ih_i=\frac{5}{2}n_ikT_i$. It is shown that the heat flux can be written as

$$\vec{q}=\sum_{i=1}^{N}\left(h_i-\frac{\rho k}{n}\sum_{j=1}^{N}\frac{\Lambda_jE_{ji}T_i}{n_jm_jm_i}\right)\rho_i\overline{\vec{V}_i}-\sum_{i=1}^{N}(\kappa_i\vec{\nabla}T_h+\kappa_j^{\theta}T_i\vec{\nabla}\ln\theta), \tag{3.12}$$

where

$$\Lambda_i=\sum_{j=1}^{N}\kappa_{ji}^{D}, \tag{3.13}$$

$$\kappa_i=\kappa_i'+\frac{\rho k}{n}\sum_{j,k=1}^{N}\frac{\kappa_{ij}^{D}T_kE_{jk}}{m_kn_jm_jT_h}D_k^T, \tag{3.14}$$

$$\kappa_i^{\theta}=\kappa_i'^{\,\theta}+\frac{\rho k}{n}\sum_{j,k=1}^{N}\frac{\kappa_{ij}^{D}E_{jk}n_kT_k}{n_jm_jT_i}D_k^{\theta}, \tag{3.15}$$

and

$$D_i^{\theta}=\frac{D_i^{\theta *}}{n_im_i}-\frac{n}{n_i\rho kT_i}\sum_{j=1}^{N}m_jD_{ij}^{\theta}g_j \tag{3.16}$$

with $T_i=T_e$ if $i=1$ and $T_i=T_h$ if $i\geqslant 2$.

E_{ij} is the element of the inverse of the matrix where the general element is $D_{ji}m_i$ as defined in [53]. κ_i and κ_i^{θ} are, respectively, the true translational conductivity due to the temperature gradient of heavy species and thermal nonequilibrium between electrons and heavy species. The thermal conductivity due to the nonequilibrium thermal κ_i^{θ} highlights the energy transfer between the two subsystems consisting of electrons and heavy species, which are not independent anymore. This energy transfer tends to remove the temperature difference between both species in the mixture.

C. Viscosity coefficient in a multitemperature plasma

The pressure tensor $\overleftrightarrow{p}$ is given by

$$\overleftrightarrow{p}=\sum_{i=1}^{N} m_i \int \vec{V}_i\vec{V}_i f_i d\vec{V}_i . \qquad (3.17)$$

Inserting $f_i=f_i^{(0)}(1+\Phi_i)$ due to Eq. (2.39) into Eq. (3.17), the tensor of pressure is given by

$$\overleftrightarrow{p}=\sum_{i=1}^{N} [n_i k T_i(1-d_{i1}Q_1^{(0)})\overleftrightarrow{U}-2\mu_i\overleftrightarrow{S}]. \qquad (3.18)$$

The viscosity coefficient μ_i associated to the ith species is

$$\mu_i=\tfrac{1}{2}n_i k T_i b_{i0} \qquad (3.19)$$

with $T_i=T_e$ if $i=1$ and $T_i=T_h$ if $i\geqslant 2$.

The tensor $\overleftrightarrow{S}$ has the components

$$S_{ij}=\frac{1}{2}\left(\frac{\delta v_{0i}}{\delta x_j}+\frac{\delta v_{0j}}{\delta x_i}\right)-\frac{1}{3}\delta_{ij}\vec{\nabla}\cdot\vec{v}_o .$$

It has to be noted that the hydrostatic pressure $p=\Sigma_{i=1}^{N} n_i k T_i$ defined at the zero-order approximation of the distribution function is perturbed by d_{il} obtained from the second-order approximation of Sonine polynomials.

IV. CALCULATION OF BRACKET INTEGRALS IN A MULTITEMPERATURE PLASMA

This section is devoted to the calculation of bracket integrals such as $[\overline{\overline{W}}_i S_n^q(W_i^2);\overline{\overline{W}}_j S_n^p(W_j^2)]_{ij}$ and $[\overline{\overline{W}}_j S_n^q(W_i^2);\overline{\overline{W}}_i S_n^p(W_i^2)]_{ij}$ defined in Sec. II B 4, where $\overline{\overline{W}}$ and n are, respectively and successively, $\vec{W}$ and $\frac{3}{2}$, $\vec{W}^{\mathrm{o}}\vec{W}$ and $\frac{5}{2}$. The calculation of the bracket integral in which $\overline{\overline{W}}$ and n are, respectively, 1 and $\frac{1}{2}$ is not presented because it is not directly used in the calculation of transport coefficients. Its derivation can be easily obtained following Appendix B. Only the results are presented below, whereas Appendix B gives more details about the derivation of $[\vec{W}_i S_{3/2}^q(W_i^2);\vec{W}_j S_{3/2}^p(W_j^2)]_{ij}$. In Sec. VI A, comments are given about computed results for an electron–heavy-species collision.

Moreover, the expressions of bracket integrals corresponding to the collision electrons-electrons or heavy species–heavy species are the same as at thermal equilibrium, but with the correct corresponding temperatures.

A. Calculation of the bracket integral $[\vec{W}_i S_{3/2}^q(W_i^2);\vec{W}_j S_{3/2}^p(W_j^2)]_{ij}$

Following notations given in Appendix B, where reduced masses are defined as

$$M_{i1}=\frac{m_i T_j}{m_i T_j+m_j T_i}, \qquad (4.1)$$

$$M_{i2}=\frac{m_j T_j}{m_i T_i+m_j T_j}, \qquad (4.2)$$

it is shown in Appendix B that

$$[\vec{W}_i S_{3/2}^q(W_i^2);\vec{W}_j S_{3/2}^p(W_j^2)]_{ij}=8\,\frac{(m_i+m_j)^3}{(m_i m_j)^{3/2}}\,\theta_{ij}^{3/2}[1+M_{i1}(\theta_{ij}^2-1)]^{1/2}M_{i1}^{p+2}M_{i2}^{q+2}\sum_{qpr'\ell'} A_{qpr'\ell'}\Omega_{ij}^{(\ell',r')}, \qquad (4.3)$$

where $\Omega_{ij}^{(l',r')}$ is the collision integral taking into account the interaction potential between the species i and j. Their definition is given in Sec. V. $A_{qpr'p'}$ are numbers depending on m_i, m_j, and $\theta_{ij}=T_i/T_j$. They are obtained from the following expression:

$$\sum_{qpr'\ell'} A_{qpr'\ell'}\gamma^{2r'}\cos^{\ell'}\chi=\sum \gamma^{2r}(r+\tfrac{3}{2}+\gamma^2\{B_{ij}-A_{ij}\cos\chi\})(-1)^{j+i}[2(1-\cos\chi)]^{k-j}$$
$$\times(2\cos\chi+\theta_{ij}-1)^i(\theta_{ij}-1)^{2r+\ell-2k-m-i}(\theta_{ij}+1)^{\ell-m}M_{i1}^{r+\ell-k-m}M_{i2}^{r-k-i}$$
$$\times\frac{(r+\frac{3}{2}+\ell)_\ell(j+m)!}{j!(k-j)!i!(r-k-i)!m!(\ell-m)!(j+m-n)!n!}, \qquad (4.4)$$

where

$$\eta_{ij}=\frac{m_i}{m_j},$$

$$\alpha_{ij}=\frac{m_iT_j+m_jT_i}{m_iT_i+m_jT_j},$$

$$A_{ij}=M_{i2}(1-\theta_{ij})+\alpha_{ij},$$

$$B_{ij}=M_{i2}\eta_{ij}(1-\theta_{ij}).$$

The term rq denotes the product of the q factors r, $r-1,\ldots,r-q+1$. The sum of the left-hand side of expression (4.4) is independent of that of the right-hand side. The sums have to be developed in accordance with the summation conditions given below.

First, for fixed values of q and p, the double sum on the left-hand side of Eq. (4.4) is developed over r' and ℓ'. This expansion will be the same in expression (4.3). Then, the multiple sum of the right-hand side of expression (4.4) is developed over r, k, j, i, ℓ, and m. Lastly, the coefficients in front of the products $\gamma^{2a}\cos^b\chi$ on the right-hand side are identified with the A_{qpab} numbers on the left-hand side of Eq. (4.4) and are introduced into Eq. (4.3).

The summation conditions are the following. For the left-hand side of Eq. (4.4),

$$0\leq r'\leq p+q+1,$$

$$0\leq \ell'\leq q+1.$$

For the right-hand side of Eq. (4.4),

$$0\leq j\leq k\leq r\leq p+q,$$

$$0\leq i\leq r-k,$$

$$\ell=p+q+j-2k-i,$$

$$0\leq m\leq \ell,$$

$$0\leq k+i+\ell-q\leq j+m,$$

$$n=k+i+\ell-q.$$

It has to be noticed that when $\theta_{ij}=1$ in the final expansion, our results lead strictly to those obtained by [5,6].

B. Calculation of the bracket integral $[\vec{W}_iS^q_{3/2}(W_i^2);\vec{W}_iS^p_{3/2}(W_i^2)]_{ij}$

Following a calculation similar to the previous one, it can be easily shown that

$$[\vec{W}_iS^q_{3/2}(W_i^2);\vec{W}_iS^p_{3/2}(W_i^2)]_{ij}=8\,\frac{(m_i+m_j)^3}{(m_im_j)^{3/2}}\,\theta_{ij}^{3/2}(M_{i1}M_{i2})^{3/2}\sum_{qpr'\ell'}A'_{qpr'\ell'}\Omega_{ij}^{(\ell',r')},\tag{4.5}$$

where $A'_{qpr'\ell'}$ is a number depending on m_i, m_j, and $\theta_{ij}=T_i/T_j$, which is obtained from the following:

$$\sum_{qpr'\ell'}A'_{qpr'\ell'}\gamma^{2r'}\cos^{\ell'}\chi=\sum\,[M_{i1}(r+\tfrac{3}{2})+\gamma^2\{A_{ij}(1+\cos\chi)-M_{i2}\}]\gamma^{2r}(\theta_{ij}+1)^n(\theta_{ij}-1)^{2r+j-i-2k}M_{i2}^r(1$$
$$-M_{i1})^mM_{i1}^{r+j-k-i}(2M_{i1}-1)^{\ell-m}(2\cos\chi+\theta_{ij}-1)^{j-i-n}[2-2M_{i1}+M_{i1}(\theta_{ij}^2-1)$$
$$+2M_{i1}\theta_{ij}\cos\chi]^{k-j}\frac{(-1)^j(r+\frac{3}{2}+\ell)_\ell(m+i)!}{(r-k)!(k-j)!i!n!(j-i-n)!m!(\ell-m)!u!(m+i-u)!},\tag{4.6}$$

where

$$A_{ij}=M_{i2}+M_{i1}M_{i2}(\theta_{ij}-1).$$

The summation conditions are the following. For the left-hand side of Eq. (4.6),

$$0\leq r'\leq p+q+1,$$

$$0\leq \ell'\leq q+1.$$

For the right-hand side of Eq. (4.6),

$$0\leq i\leq j\leq k\leq r\leq p+q,$$

$$0\leq n\leq j-i,$$

$$0\leq 2k+2\ell-j-p-q\leq \ell,$$

$$0\leq k+\ell-n-q\leq 2k+2\ell+i-j-p-q,$$

$$u=k+\ell-n-q,$$

$$m=2k+2\ell-j-p-q.$$

C. Calculation of the bracket integral $[\vec{W}_i^o\vec{W}_iS^q_{5/2}(W_i^2);\vec{W}_j^o\vec{W}_jS^p_{5/2}(W_j^2)]_{ij}$

It can be shown that

$$[\vec{W}_i^{\circ}\vec{W}_i S_{5/2}^{q}(W_i^2);\vec{W}_j^{\circ}\vec{W}_j S_{5/2}^{p}(W_j^2)]_{ij}=\frac{16}{3}\frac{(m_i+m_j)^3}{(m_i m_j)^{3/2}}\theta_{ij}^{3/2}M_{i1}^{p+(5/2)}M_{i2}^{q+(5/2)}\times[1+M_{i1}(\theta_{ij}^2-1)]\sum_{qpr'\ell'}B_{qpr'\ell'}\Omega_{ij}^{(\ell',r')}. \tag{4.7}$$

Coefficients $B_{qpr'\ell'}$ are obtained due to

$$\sum_{qpr'\ell'}B_{qpr'\ell'}\gamma^{2r'}\cos^{\ell'}\chi=\sum\gamma^{2r}\Bigg[(r+\tfrac{3}{2})(r+\tfrac{5}{2})+(2r+5)\gamma^2(B_{ij}-A_{ij}\cos\chi)+\gamma^4\times\left((B_{ij}-A_{ij}\cos\chi)^2-\frac{A_{ij}^2}{2}(1-\cos^2\chi)\right)\Bigg](-1)^{j+i}[2(1-\cos\chi)]^{k-j}(2\cos\chi+\theta_{ij}-1)^i\times(\theta_{ij}-1)^{2r+\ell-2k-m-i}(\theta_{ij}+1)^{\ell-m}M_{i1}^{r+\ell-k-m}M_{i2}^{r-k-j}\times\frac{(r+\frac{5}{2}+\ell)_\ell(j+m)!}{j!(k-j)!i!(r-k-i)!m!(\ell-m)!(j+m-n)!n!}, \tag{4.8}$$

where

$$\eta_{ij}=\frac{m_i}{m_j},\quad \alpha_{ij}=\frac{m_iT_j+m_jT_i}{m_iT_i+m_jT_j},$$

$$A_{ij}=M_{i2}(1-\theta_{ij})+\alpha_{ij},$$

$$B_{ij}=M_{i2}\eta_{ij}(1-\theta_{ij}).$$

The summation conditions are the following. For the left-hand side of Eq. (4.8),

$$0\leqslant r'\leqslant p+q+2,$$

$$0\leqslant \ell'\leqslant q+2.$$

For the right-hand side of Eq. (4.8),

$$0\leqslant j\leqslant k\leqslant r\leqslant p+q,$$

$$0\leqslant i\leqslant r-k,$$

$$\ell=p+q+j-2k-i,$$

$$0\leqslant m\leqslant \ell,$$

$$0\leqslant k+i+\ell-q\leqslant j+m,$$

$$n=k+i+\ell-q.$$

D. Calculation of the bracket integral $[\vec{W}_i^{\circ}\vec{W}_i S_{5/2}^{q}(W_i^2);\vec{W}_i^{\circ}\vec{W}_i S_{5/2}^{p}(W_i^2)]_{ij}$

It can be shown that

$$[\vec{W}_i^{\circ}\vec{W}_i S_{5/2}^{q}(W_i^2);\vec{W}_j^{\circ}\vec{W}_j S_{5/2}^{p}(W_j^2)]_{ij}=\frac{16}{3}\frac{(m_i+m_j)^3}{(m_i m_j)^{3/2}}\theta_{ij}^{3/2}(M_{i1}M_{i2})^{3/2}\sum_{qpr'\ell'}B'_{qpr'\ell'}\Omega_{ij}^{(\ell',r')}. \tag{4.9}$$

Coefficients $B'_{pqr'\ell'}$ are obtained from

$$\sum_{qpr'\ell'}B'_{qpr'\ell'}\gamma^{2r'}\cos^{\ell'}\chi=\sum\Bigg\{\Bigg[(r+\tfrac{3}{2})(r+\tfrac{5}{2})M_{i1}^2+(2r+5)\gamma^2M_{i1}[A_{ij}(1+\cos\chi)-M_{i2}]+\gamma^4\bigg([A_{ij}(1+\cos\chi)-M_{i2}]^2-\frac{A_{ij}^2}{2}(1-\cos^2\chi)\bigg)\Bigg]\gamma^{2r}(\theta_{ij}+1)^n(\theta_{ij}-1)^{2r+j-i-2k}M_{i2}^r(1-M_{i1})^mM_{i1}^{r+j-k-i}(2M_{i1}-1)^{\ell-m}\times(2\cos\chi+\theta_{ij}-1)^{j-i-n}[2-2M_{i1}+M_{i1}(\theta_{ij}^2-1)+2M_{i1}\theta_{ij}\cos\chi]^{k-j}\times\frac{(-1)^j(r+\frac{5}{2}+\ell)_\ell(m+i)!}{(r-k)!(k-j)!i!n!(j-i-n)!m!(\ell-m)!u!(m+i-u)!}\Bigg\}, \tag{4.10}$$

where

TABLE IV. Calculation of $A_{00r\ell'}$ for different values of θ with a mass ratio $m_j/m_i=10^{-5}$.

θ	1.00	1.25	1.50	2.00	3.00
A_{0011}	−1.00	−1.25	−1.50	−2.00	−3.00

$$A_{ij}=M_{i2}+M_{i1}M_{i2}(\theta_{ij}-1).$$

The summation conditions are the following. For the left-hand side of Eq. (4.10),

$$0\leqslant r'\leqslant p+q+2,$$

$$0\leqslant \ell'\leqslant q+2.$$

For the right-hand side of Eq. (4.10),

$$0\leqslant i\leqslant j\leqslant k\leqslant r\leqslant p+q,$$

$$0\leqslant n\leqslant j-i,$$

$$0\leqslant 2k+2\ell-j-p-q\leqslant \ell,$$

$$0\leqslant k+\ell-n-q\leqslant 2k+2\ell+i-j-p-q,$$

$$u=k+\ell-n-q,$$

$$m=2k+2\ell-j-p-q.$$

V. COLLISION INTEGRAL

The definition of the collision integral is the same as that given by Chapman and Cowling [5],

$$\phi_{ij}^{(\ell')}=\int_0^{\infty}(1-\cos^{\ell'}\chi)gb\,db, \tag{5.1}$$

$$\Omega_{ij}^{(\ell',r')}=\pi^{1/2}\int_0^{\infty}e^{-\gamma^2}\gamma^{2(r'+1)}\phi_{ij}^{(\ell')}d\gamma, \tag{5.2}$$

χ being the deflection angle.

$\phi_{ij}^{(\ell')}$ and $\Omega_{ij}^{(\ell',r')}$ are, respectively, the transport cross section and the collision integral. It has to be noted that during a collision between two particles with different temperatures, the variable changes (B5) and (B6) given in Appendix B imply the introduction of an effective temperature of collision T_{ij}^{*} such that

TABLE V. Calculation of $A_{01r\ell'}$ and $A_{10r\ell'}$ for different values of θ with a mass ratio $m_j/m_i=10^{-5}$.

θ	1.00	1.25	1.50	2.00	3.00
A_{0111}	−2.50	−3.12	−3.75	−5.00	−7.50
A_{1011}	−2.50	−1.00	0.00	1.25	2.50
A_{0121}	1.00	1.25	1.50	2.00	3.00
A_{1021}	1.00	1.40	2.00	3.50	7.00
A_{0122}	0.00	0.00	0.00	0.00	0.00
A_{1022}	0.00	−0.50	−1.00	−2.00	−4.00

TABLE VI. Calculation of $A_{11r\ell'}$ for different values of θ with a mass ratio $m_j/m_i=10^{-5}$.

θ	1.00	1.25	1.50	2.00	3.00
A_{1111}	−13.75	−8.50	−5.00	−0.63	−3.75
A_{1121}	5.00	3.30	3.00	4.50	11.00
A_{1122}	2.00	0.75	−0.50	−3.00	−8.00
A_{1131}	−1.00	−1.40	−2.00	−3.50	−7.00
A_{1132}	0.00	0.50	1.00	2.00	4.00

$$T_{ij}^{*}=\left[\frac{1}{m_i+m_j}\left(\frac{m_i}{T_j}+\frac{m_j}{T_i}\right)\right]^{-1}. \tag{5.3}$$

However, Hirschfelder has defined a transport cross section $Q_{ij}^{(\ell')}$ such that

$$Q_{ij}^{(\ell')}=\frac{2\pi}{g}\phi_{ij}^{(\ell')}, \tag{5.4}$$

that is,

$$Q_{ij}^{(\ell')}=2\pi\int_0^{+\infty}(1-\cos^{\ell'}\chi)b\,db. \tag{5.5}$$

Expressing Eq. (5.2) with the variable γ (defined in Appendix B), $\gamma^2=\mu_{ij}g^2/2kT_{ij}^{*}$,

$$\Omega_{ij}^{(\ell'',r')}=\left(\frac{kT_{ij}^{*}}{2\pi\mu_{ij}}\right)^{1/2}\int_0^{+\infty}e^{-\gamma^2}\gamma^{2r'+3}Q_{ij}^{(\ell'')}d\gamma, \tag{5.6}$$

$\mu_{ij}=m_im_j/(m_i+m_j)$ being a reduced mass.

VI. APPLICATION TO A TWO-TEMPERATURE ARGON PLASMA

In this section, an application is presented for a two-temperature argon plasma. First, computed results of bracket integrals show clearly the discrepancies with respect to the equilibrium. Transport coefficients are then calculated for ionized argon plasma out of thermal equilibrium.

A. Comments on the results of bracket integrals in a two-temperature plasma

Some computed results of the calculation of expressions (4.4) and (4.8) are presented below for different values of (q,p) for a mass ratio $m_j/m_i=10^{-5}$, which corresponds to an electron–heavy-species collision. The behavior of $A_{qpr'\ell'}$ and $B_{qpr'\ell'}$ is given in Tables IV–VIII as a function of thermal nonequilibrium θ. It is to be noted that, in this case, m_j is the mass of an electron. Consequently, θ_{ij} is calculated such that $\theta_{ij}=1/\theta$. The thermal nonequilibrium has been chosen to vary from 1 to 3 because experimental studies (for example dealing with a free-burning arc [23] or an inductively coupled plasma torch [26]), close to the atmospheric pressure, have shown that θ does not exceed 3. First, it is

TABLE VII. Calculation of $B_{00r\ell'}$ for different values of θ with a mass ratio $m_j/m_i=10^{-5}$.

θ	1.00	1.25	1.50	2.00	3.00
B_{0011}	−5.00	−6.25	−7.50	−10.00	−15.00
B_{0021}	0.00	0.63	−1.50	−4.00	−12.00
B_{0022}	1.50	2.34	3.37	6.00	13.49

observed that our results strictly lead to those presented in [5] at thermal equilibrium ($\theta=1$) in each calculation (see Tables IV–VII).

Moreover, Table V shows that a asymmetry in $A_{01r'\ell'}$ and $A_{10r'\ell'}$ arises when calculating for $\theta\neq 1$ as predicted in Sec. II B 4.

Tables V, VI, VII, and VIII show, respectively, the introduction of new coefficients A_{0122} and A_{1022}, A_{1132}, B_{1021}, and B_{1031} and B_{1033}, which are not present at thermal equilibrium. As a result, the corresponding collision integrals $\Omega_{ij}^{(\ell',r')}$ will have to be taken into account in the calculation of transport coefficients out of thermal equilibrium. Thus, it is shown that the previous linear combinations are significantly modified in comparison with equilibrium.

Besides, it has to be underlined that the results presented in these tables are different if bracket integrals are calculated with a mass ratio $m_i/m_j=10^{-5}$, except at thermal equilibrium, because of the asymmetry previously dealt with. In this case, it is observed that coefficients of linear combinations do not significantly vary. Indeed, when an electron and a heavy species collide, the latter is much less affected by collision than the electron. Thus, when $m_i/m_j=10^{-5}$, m_j represents the mass of a heavy species. As a result, it is expected that, according to the definition of bracket integrals [expression (2.54)], the results of the calculation of the latter are slightly modified regardless of the applied thermal nonequilibrium θ.

Similar behaviors of $A'_{qpr\ell'}$ and $B'_{qpr\ell'}$, computed from expressions (4.6) and (4.10), are observed when varying θ. In this case, these coefficients depend on the mass ratio m_i/m_j at thermal equilibrium.

B. Two-temperature transport coefficients in an argon plasma

An application of transport coefficients derived in the previous sections is presented for a two-temperature argon plasma. Calculations are performed at atmospheric pressure and the two-temperature plasma composition is obtained using the Saha equation for ionization of Van de Sanden *et al.* [45]. Electron Ar and Ar^+ species have been considered. Transport coefficients are calculated in the fourth approximation ($\xi=4$) for different values of θ.

TABLE VIII. Calculation of $B_{10r\ell'}$ for different values of θ with a mass ratio $m_j/m_i=10^{-5}$.

θ	1.00	1.25	1.50	2.00	3.00
B_{1011}	−17.50	−10.50	−5.83	0.00	5.83
B_{1021}	7.00	8.41	11.66	21.00	44.32
B_{1022}	5.25	1.75	−1.75	−8.75	−22.74
B_{1031}	0.00	0.36	1.08	4.00	16.65
B_{1032}	−1.50	−2.69	−4.75	−11.50	−35.58
B_{1033}	0.00	0.94	2.25	6.00	18.00

TABLE IX. Electron thermal conductivity for different values of θ calculated at $T_e=20\,000$ K.

θ	1.00	1.30	2.00	3.00
κ_e (W m^{-1} K^{-1})	2.58	2.71	2.95	3.20
$\kappa_e^{\mathrm{Devoto}}$ (W m^{-1} K^{-1}) [34,37]	2.58	2.38	2.00	1.85

Table IX presents a comparison of the electron thermal conductivity calculated from the expression (3.14) and from expressions given in [37] for different values of the nonequilibrium parameter θ and for $T_e=20\,000$ K. The total thermal conductivity is not displayed because the contribution of electrons is more prevalent when calculating the thermal conductivity at that temperature. It has to be noted that the electron thermal conductivity κ_e has been calculated with respect to the electron temperature gradient to be compared to that of Devoto, so that expression (3.14) has to divided by θ.

First, the presented theory gives the same results as other authors [1,7] at thermal equilibrium (when $\theta=1.00$). Moreover, it has been checked that, at thermal equilibrium, the results obtained from [37] are the same as those of the complete calculation, which confirms that the simplified theory of the transport coefficient is valid at thermal equilibrium.

However, the simplified theory of Devoto, modified by Bonnefoi, underestimates the electron thermal conductivity when nonequilibrium conditions are applied. The discrepancy reaches more than 40% for $\theta=3.00$ (see Table IX). It can be observed that the electron thermal conductivity of Devoto decreases with θ. It has been checked that, when calculating κ_e with $\theta=1.00$ but with a nonequilibrium composition, κ_e increases with respect to the equilibrium value.

Table X shows two-temperature ordinary diffusion coefficients calculated at $T_e=6000$ K for different values of θ. This temperature has been chosen because ionization is no longer negligible and the ionization of argon atoms is not completed, which allows us to observe the behavior of diffusion coefficients implying electron Ar and Ar^+ species.

First, the obtained equilibrium values ($\theta=1.00$) for $D_{e\text{-Ar}}$ are in good agreement with the admitted values since it has been checked that the value of electrical conductivity, which is a function of $D_{e\text{-Ar}}$ and $D_{e\text{-Ar}^+}$ with $D_{e\text{-Ar}}\approx D_{e\text{-Ar}^+}$, is that obtained at thermal equilibrium [53].

TABLE X. Ordinary diffusion coefficients for different values of θ calculated at $T_e=6000$ K.

θ	1.00	1.30	2.00	3.00
$D_{e\text{-Ar}}$ (m^2 s^{-1})	7.73	5.92	3.84	2.55
$D_{\text{Ar-Ar}^+}$ (10^{-4} m^2 s^{-1})	2.58	2.38	2.00	1.85

It is observed that two-temperature diffusion coefficients decrease with θ. The diffusion coefficient $D_{\text{Ar-Ar}^+}$ being a function of θ only through the composition, the latter contributes to decrease the diffusion coefficients. The same observation can be made for $D_{e\text{-Ar}}$, but the influence θ through the bracket integrals emphasizes this tendency. A more detailed analysis of transport coefficients over a wide range of temperature will be performed in a future paper.

The consistent calculation of the electron thermal conductivity κ_e shows that the calculation of thermal diffusion D_i^T as well as that of ordinary diffusion coefficients D_{ij}, through the inverse matrix element E_{ji}, are consistent [see relation (3.14)]. Is has been checked that the two-temperature diffusion coefficients satisfy symmetry conditions through Eq. (2.45). This calculation also shows that, contrary to a commonly accepted idea, the transport coefficients have to be computed keeping the coupling between electrons and heavy species in nonequilibrium conditions, which is not the case of the commonly used approach of Devoto [34,35].

VII. CONCLUSION

An alternate derivation of transport properties in a two-temperature plasma has been performed. Recent works have shown that the simplified theory of transport properties out of thermal equilibrium introduced by Devoto and then modified by Bonnefoi and neglecting the coupling between electrons and heavy species leads to unphysical results. Thus, two-temperature transport coefficients have been derived starting from Boltzmann's equation. The latter is solved by using the well-known Chapman-Enskog method, which has been adapted to thermal nonequilibrium plasmas. It has been assumed that the distribution function of species, the solution of Boltzmann's equation, is a Maxwellian, at T_e for electrons and T_h for heavy species, perturbed by a slowly time- and space-dependent first-order perturbation function.

The zero-order approximation of the subdivision of Boltzmann's equation using the Chapman-Enskog method does not vanish for two colliding species with different temperatures, that is, for electrons and heavy species. Consequently, the corresponding expression has been included in the calculation of the first-order perturbation function. The latter has been shown to be a function of the temperature gradient of heavy species (or of electrons), which can be considered as a reference temperature gradient, and a gradient of temperature ratio $\theta = T_e/T_h$. The introduction of the gradient $\vec{\nabla}\theta$ allows us to define separate systems of linear equations for the calculation of transport coefficients and to maintain the coupling between electrons and heavy species in the resolution of these systems, which was not the case in the simplified theory of Devoto. To solve them, the calculation of bracket integrals, introduced by Chapman and Cowling, has been generalized out of thermal equilibrium. These bracket integrals are significantly modified in comparison with those at thermal equilibrium. This is due, on the one hand, to the coefficients of these linear combinations, which are then drastically changed following the applied thermal nonequilibrium, and, on the other hand, to the introduction of collision integrals with indexes (r', ℓ') higher than those at thermal equilibrium. Moreover, the derivation of diffusion velocity shows that nonequilibrium diffusion coefficients characterizing the temperature difference in a two-temperature plasma $[\vec{\nabla}\theta = \vec{\nabla}(T_e/T_h)]$ have to be introduced. The translational thermal conductivity is also altered compared to previous calculations.

An application is presented for a two-temperature argon plasma commonly used in experimental devices. Calculated two-temperature transport coefficients show that the developed theory is consistent. First, the model in which $T_e = T_h$ checks equilibrium results given in the literature. Secondly, the simplified theory of transport coefficients, very often used in modeling, underestimates at $T_e = 20\,000$ K the electron thermal conductivity up to 40% compared to the accurate value for a nonequilibrium parameter $\theta = 3.00$. Lastly, it is shown that two-temperature diffusion coefficients, satisfying symmetry rules, can be calculated between electrons and heavy species, contrary to what happens when using the simplified theory of transport coefficients of Devoto, and when $\theta = 1.00$ have values currently admitted at equilibrium.

The derivation developed here will allow a physically rigorous treatment of transport properties in nonequilibrium plasmas. Particularly, it will allow the combined diffusion coefficient method of Murphy to be applied to nonequilibrium plasmas and describe demixing in non-LTE regions such as those observed close to the electrodes in dc arcs or in the fringes of an arc column or a plasma jet.

APPENDIX A

Consider two vectors $\vec{a}$ and $\vec{b}$ with respective components (a_x, a_y, a_z) and (b_x, b_y, b_z). The product of these vectors $\vec{a}\vec{b}$ is written

$$\vec{a}\vec{b} = \begin{pmatrix} a_x b_x & a_x b_y & a_x b_z \\ a_y b_x & a_y b_y & a_y b_z \\ a_z b_x & a_z b_y & a_z b_z \end{pmatrix}.$$

$\vec{a}\vec{b}$ is a second-order tensor.

Let $\overleftrightarrow{W}$ be a second-order tensor [6],

$$\mathring{\overleftrightarrow{W}} = \overleftrightarrow{W} - \tfrac{1}{3}(w_{xx} + w_{yy} + w_{zz})\overleftrightarrow{U}, \tag{A1}$$

where w_{xx}, w_{yy}, and w_{zz} are components of tensor $\overleftrightarrow{W}$.

The double product between two second-order tensors $\overleftrightarrow{W}$ and $\overleftrightarrow{W}'$ is written as

$$\overleftrightarrow{W} : \overleftrightarrow{W}' = \sum_{\alpha\beta} w_{\alpha\beta} w'_{\alpha\beta}, \tag{A2}$$

$w_{\alpha\beta}$ and $w'_{\alpha\beta}$ being, respectively, components of $\overleftrightarrow{W}$ and $\overleftrightarrow{W}'$.

APPENDIX B

This appendix is devoted to the calculation of the bracket integral $[\vec{W}_i S_{3/2}^q(W_i^2); \vec{W}_j S_{3/2}^p(W_j^2)]_{ij}$ and that of $J_{im}^{(0)}$. The derivation of the other bracket integrals is similar to the following.

1. Encounter relationships

An isolated system, consisting in two colliding particles i and j, is considered. It is assumed, for the sake of generality, that the kinetic temperatures of the species i and j are, respectively, T_i and T_j.

$\vec{V}_i$, $\vec{V}_j$, $\vec{G}_0$, and $\vec{g}$ are, respectively, the peculiar velocities of species i and j, the velocity of their mass center, and their relative velocity.

The classical encounter relationships are the following:

$$\overline{V_i}=\vec{G}_0-\frac{m_j}{m_i+m_j}\vec{g}, \tag{B1}$$

$$\vec{V}_j=\vec{G}_0+\frac{m_i}{m_i+m_j}\vec{g}, \tag{B2}$$

where $\vec{g}=\vec{v}_j-\vec{v}_i=\vec{V}_j-\vec{V}_i$.

The reduced velocities of species i and j are

$$\vec{W}_i=\left(\frac{m_i}{2kT_i}\right)^{1/2}\vec{V}_i, \tag{B3}$$

$$\vec{W}_j=\left(\frac{m_j}{2kT_j}\right)^{1/2}\vec{V}_j. \tag{B4}$$

The following variable change introduced by Devoto [34] is used:

$$\vec{\Gamma}_0=\left(\frac{m_i}{2kT_i}+\frac{m_j}{2kT_j}\right)^{1/2}\vec{G}_0, \tag{B5}$$

$$\vec{\gamma}=\left[\frac{m_im_j}{(m_i+m_j)^2}\left(\frac{m_i}{2kT_j}+\frac{m_j}{2kT_i}\right)\right]^{1/2}\vec{g}. \tag{B6}$$

During a collision between a heavy particle and a light one, the collision mass center is almost that of the heavy particle. $\vec{\Gamma}_0$ is therefore mainly a function of T_j if $m_i \ll m_j$. However, since the velocity of the light particle is much higher than that of the heavy one in the previous assumption, $\vec{\gamma}$ is mainly a function of T_i. Inserting this variable change into Eqs. (B1) and (B2), we get

$$\vec{W}_i=M_{i1}^{1/2}\vec{\Gamma}_0-M_{i2}^{1/2}\vec{\gamma}, \tag{B7}$$

$$\vec{W}_j=M_{j1}^{1/2}\vec{\Gamma}_0+M_{j2}^{1/2}\vec{\gamma}, \tag{B8}$$

where reduced masses have been defined:

$$M_{i1}=\frac{m_iT_j}{m_iT_j+m_jT_i}, \tag{B9}$$

$$M_{i2}=\frac{m_jT_j}{m_iT_i+m_jT_j}, \tag{B10}$$

$$M_{j1}=\frac{m_jT_i}{m_iT_j+m_jT_i}, \tag{B11}$$

$$M_{j2}=\frac{m_iT_i}{m_iT_i+m_jT_j}. \tag{B12}$$

It has to be noted that if $\vec{W}_i'$ and $\vec{\gamma}'$ are, respectively, $\vec{W}_i$ and $\vec{\gamma}$ after collision, it follows that

$$\vec{W}_i'=M_{i1}^{1/2}\vec{\Gamma}_0-M_{i2}^{1/2}\vec{\gamma}' \tag{B13}$$

and [6]

$$\gamma=\gamma', \tag{B14}$$

$$\vec{\gamma}\cdot\vec{\gamma}'=\gamma^2\cos\chi. \tag{B15}$$

2. Calculation of $[\vec{W}_iS_{3/2}^q(W_i^2);\vec{W}_jS_{3/2}^p(W_j^2)]_{ij}$

This bracket integral called I_{ij} is defined in Sec. II B 4 by

$$I_{ij}=\frac{1}{n_in_j}\int\int\int f_i^{(0)}f_j^{(0)}[\vec{W}_jS_{3/2}^p(W_j^2)-\vec{W}_j'K_i(W_i,\theta_{ij})S_{3/2}^p(W_j'^2)]\cdot\vec{W}_iS_{3/2}^q(W_i^2)\times gb\,db\,d\varepsilon\,d\vec{v}_id\vec{v}_j. \tag{B16}$$

According to the definition of Sonine polynomials [6], I_{ij} is the coefficient of s^pt^q in the expansion of Π_{ij} defined as

$$\Pi_{ij}=\frac{1}{n_in_j}(1-s)^{-5/2}(1-t)^{-5/2}\int\int\int\int f_i^{(0)}f_j^{(0)}(\vec{W}_je^{-SW_j^2}-\vec{W}_j'K_ie^{-SW_j'^2})\cdot\vec{W}_ie^{-TW_i^2}gb\,db\,d\epsilon\,d\vec{v}_id\vec{v}_j, \tag{B17}$$

where

$$S=\frac{s}{1-s}\quad\text{and}\quad T=\frac{t}{1-t}.$$

After performing the variable changes (B5) and (B6) and inserting the Maxwellian distribution functions defined as

$$f_i^{(0)}=n_i\left(\frac{m_i}{2\pi kT_i}\right)^{3/2}\exp(-W_i^2), \tag{B18}$$

$$f_j^{(0)}=n_j\left(\frac{m_j}{2\pi kT_j}\right)^{3/2}\exp(-W_j^2) \tag{B19}$$

into Eq. (B17), we get

$$\Pi_{ij}=(1-s)^{-5/2}(1-t)^{-5/2}\pi^{-3}\frac{(m_i+m_j)^3}{(m_im_j)^{3/2}}(M_{i1}M_{i2})^{3/2}\theta_{ij}^{3/2}$$
$$\times\int\int\int\int e^{-W_i^2-W_j^2}(\vec{W}_j e^{-SW_j^2}$$
$$-\vec{W}_j' e^{-SW_j'^2-(1-\theta_{ij})(W_i'^2-W_i^2)})\cdot\vec{W}_i e^{-TW_i^2}$$
$$\times g b db\, d\epsilon\, d\vec{\Gamma}_0 d\vec{\gamma}, \tag{B20}$$

where K_i has been replaced by its value

$$K_i(W_i,\theta_{ij})=\exp[-(1-\theta_{ij})(W_i'^2-W_i^2)]. \tag{B21}$$

As shown in [6], let $H_{ij}(\chi)$ be such that

$$H_{ij}(\chi)=\int \exp[-W_i^2-W_j^2-SW_j'^2-TW_i^2$$
$$-(1-\theta_{ij})(W_i'^2-W_i^2)]\vec{W}_j'\cdot\vec{W}_i d\vec{\Gamma}_0. \tag{B22}$$

Thus, $H_{ij}(0)$ is the limit function of $H_{ij}(\chi)$ when the species i and j do not collide:

$$H_{ij}(0)=\int \exp(-W_i^2-W_j^2-SW_j^2-TW_i^2)\vec{W}_j\cdot\vec{W}_i d\vec{\Gamma}_0. \tag{B23}$$

Equation (B17) can be written as follows:

$$\Pi_{ij}=(1-s)^{-5/2}(1-t)^{-5/2}\pi^{-3}\frac{(m_i+m_j)^3}{(m_im_j)^{3/2}}(M_{i1}M_{i2})^{3/2}\theta_{ij}^{3/2}$$
$$\times\int\int\int[H_{ij}(0)-H_{ij}(\chi)]gb\, db\, d\epsilon\, d\vec{\gamma}. \tag{B24}$$

Several relationships useful for the following expansion are defined below:

$$\theta_{ij}=\frac{T_i}{T_j},\qquad \eta_{ij}=\frac{m_i}{m_j},$$

$$\left(\frac{M_{i1}M_{i2}M_{j2}}{M_{j1}}\right)^{1/2}\theta_{ij}=M_{j2}, \tag{B25}$$

$$\left(\frac{M_{i1}M_{i2}M_{j2}}{M_{j1}}\right)^{1/2}=M_{i2}\eta_{ij},\quad \left(\frac{M_{i2}M_{j1}M_{j2}}{M_{i1}}\right)^{1/2}=M_{i2}\theta_{ij},$$

$$M_{i1}M_{i2}\theta_{ij}^2=M_{j1}M_{j2}, \tag{B26}$$

$$M_{j2}=M_{i1}+M_{i1}M_{i2}(\theta_{ij}^2-1), \tag{B27}$$

$$M_{j1}=M_{i2}+M_{i1}M_{i2}(\theta_{ij}^2-1), \tag{B28}$$

$$1-M_{i1}M_{i2}-M_{i2}M_{j1}=M_{i1}M_{i2}(\theta_{ij}^2+1), \tag{B29}$$

$$a_{ij}=1+SM_{j1}+TM_{i1}, \tag{B30}$$

$$a_{ji}=1+SM_{j2}+TM_{i2}, \tag{B31}$$

$$S^*=1-\theta_{ij}-\theta_{ij}S, \tag{B32}$$

$$b_{ij}=a_{ji}-\frac{M_{i1}M_{i2}}{a_{ij}}(S^{*2}+T^2+2S^*T\cos\chi), \tag{B33}$$

$$\vec{W}_i=M_{i1}^{1/2}\vec{\Gamma}_0-M_{i2}^{1/2}\vec{\gamma}, \tag{B34}$$

$$\vec{W}_j'=M_{j1}^{1/2}\vec{\Gamma}_0+M_{j2}^{1/2}\vec{\gamma}'. \tag{B35}$$

Using the relations (B30), (B31), (B32), (B34), and (B35), it can be shown that

$$H_{ij}(\chi)=\int \exp[-a_{ij}\Gamma_0^2-a_{ji}\gamma^2+2(M_{i1}M_{i2})^{1/2}\vec{\Gamma}_0\cdot(S^*\vec{\gamma}'$$
$$+T\vec{\gamma})]\vec{W}_j'\cdot\vec{W}_i d\vec{\Gamma}_0. \tag{B36}$$

If, instead of choosing K_i for the calculation of Eq. (B22), K_j is used, the same result is obtained.

Let $\vec{X}$ be such that [6]

$$\vec{X}=\vec{\Gamma}_0-\frac{(M_{i1}M_{i2})^{1/2}}{a_{ij}}(S^*\vec{\gamma}'+T\vec{\gamma}). \tag{B37}$$

The $H_{ij}(\chi)$ function is written with this variable change:

$$H_{ij}(\chi)=\int \exp(-a_{ij}X^2-b_{ij}\gamma^2)\vec{W}_j'\cdot\vec{W}_i d\vec{X}. \tag{B38}$$

The scalar product $\vec{W}_j'\cdot\vec{W}_i$ is calculated with Eqs. (B34) and (B35) and integration relationships given in Chapman and Cowling [6], so that

$$H_{ij}(\chi)=\pi^{3/2}e^{-b_{ij}\gamma^2}a_{ij}^{-5/2}(M_{i1}M_{j1})^{1/2}[\tfrac{3}{2}+\gamma^2$$
$$\times(1-b_{ij}+B_{ij}-A_{ij}\cos\chi)] \tag{B39}$$

with

$$A_{ij}=M_{i2}(1-\theta_{ij})+\alpha_{ij}, \tag{B40}$$

$$\alpha_{ij}=\frac{m_iT_j+m_jT_i}{m_iT_i+m_jT_j}, \tag{B41}$$

$$B_{ij}=M_{i2}\eta_{ij}(1-\theta_{ij}). \tag{B42}$$

So, using the fact that

$$e^{-b_{ij}\gamma^2}=e^{-\gamma^2}e^{(1-b_{ij})\gamma^2}=e^{-\gamma^2}\sum_{r=0}^{\infty}\frac{(1-b_{ij})^r}{r!}\gamma^{2r}, \tag{B43}$$

we get

$$H_{ij}(\chi)=\pi^{3/2}e^{-\gamma^2}a_{ij}^{-5/2}(M_{i1}M_{j1})^{1/2}\sum_{r=0}^{\infty}(r+\tfrac{3}{2}+\gamma^2$$
$$\times\{B_{ij}-A_{ij}\cos\chi\})\frac{(1-b_{ij})^r}{r!}\gamma^{2r}. \tag{B44}$$

After having calculated $1-b_{ij}$ thanks to its definition, it follows that

$$(1-s)^{-5/2}(1-t)^{-5/2}\pi^{-3/2}(M_{i1}M_{j1})^{-1/2}H_{ij}(\chi)$$
$$=e^{-\gamma^2}\sum_{r=0}^{\infty}\left\{\frac{\gamma^{2r}}{r!}(r+\tfrac{3}{2}+\gamma^2\{B_{ij}-A_{ij}\cos\chi\})\right.$$
$$\left.\times\frac{\{-sM_{i1}-tM_{i2}[1+M_{i1}(\theta_{ij}-1)(2\cos\chi+\theta_{ij}-1)]+2stM_{i1}M_{i2}(1-\cos\chi)+M_{i1}M_{i2}(1-\theta_{ij})^2\}}{\{1-sM_{i1}-tM_{i2}[1+M_{i1}(\theta_{ij}^2-1)]\}^{r+(5/2)}}\right\}. \quad (B45)$$

Equation (B45) can be written as a series such that

$$(1-s)^{-5/2}(1-t)^{-5/2}\pi^{-3/2}(M_{i1}M_{j1})^{-1/2}H_{ij}(\chi)$$
$$=e^{-\gamma^2}\sum_{qpr'\ell'}A_{qpr'\ell'}\gamma^{2r'}\cos^{\ell'}\chi(M_{i1}s)^p(M_{i2}t)^q, \quad (B46)$$

where $A_{qpr'\ell'}$ is a number depending on m_i, m_j, T_i, and T_j.

The remaining calculation is not worth presenting because it is similar to that shown in [6]. Thus, only the final results are given below.

$$[\vec{W}_iS_{3/2}^q(W_i^2);\vec{W}_jS_{3/2}^p(W_j^2)]_{ij}$$
$$=8\frac{(m_i+m_j)^3}{(m_im_j)^{3/2}}\theta_{ij}^{3/2}[1+M_{i1}(\theta_{ij}^2$$
$$-1)]^{1/2}M_{i1}^{p+2}M_{i2}^{q+2}\sum_{qpr'\ell'}A_{qpr'\ell'}\Omega_{ij}^{(\ell',r')} \quad (B47)$$

with

$$\phi_{ij}^{(\ell')}=\int_0^{\infty}(1-\cos^{\ell'}\chi)gb\,db, \quad (B48)$$

$$\Omega_{ij}^{(\ell',r')}=\pi^{1/2}\int_0^{\infty}e^{-\gamma^2}\gamma^{2(r'+1)}\phi_{ij}^{(\ell')}d\gamma. \quad (B49)$$

$\phi_{ij}^{(\ell')}$ and $\Omega_{ij}^{(\ell',r')}$ are, respectively, the transport cross section and the collision integral defined by Chapman *et al.* [6].

The $A_{qpr'\ell'}$ numbers are obtained by using successively Newton binomial decompositions of expressions (B45). Thus, it can be shown using Eq. (B46) that

$$\sum_{qpr'\ell'}A_{qpr'\ell'}\gamma^{2r'}\cos^{\ell'}\chi$$
$$=\sum\gamma^{2r}(r+\tfrac{3}{2}+\gamma^2\{B_{ij}-A_{ij}\cos\chi\})(-1)^{j+i}$$
$$\times[2(1-\cos\chi)]^{k-j}\times(2\cos\chi+\theta_{ij}-1)^i$$
$$\times(\theta_{ij}-1)^{2r+\ell-2k-m-i}(\theta_{ij}+1)^{\ell-m}M_{i1}^{r+\ell-k-m}M_{i2}^{r-k-i}$$
$$\times\frac{(r+\frac{3}{2}+\ell)_{\ell}(j+m)!}{j!(k-j)!i!(r-k-i)!m!(\ell-m)!(j+m-n)!n!}, \quad (B50)$$

where

$$\eta_{ij}=\frac{m_i}{m_j},\quad \alpha_{ij}=\frac{m_iT_j+m_jT_i}{m_iT_i+m_jT_j},$$
$$A_{ij}=M_{i2}(1-\theta_{ij})+\alpha_{ij},$$
$$B_{ij}=M_{i2}\eta_{ij}(1-\theta_{ij}).$$

The summation conditions are the following. For the left-hand side of Eq. (B51),

$$0\le r'\le p+q+1,$$
$$0\le\ell'\le q+1.$$

For the right-hand side of Eq. (B51),

$$0\le j\le k\le r\le p+q,$$
$$0\le i\le r-k,$$
$$\ell=p+q+j-2k-i,$$
$$0\le m\le\ell,$$
$$0\le k+i+\ell-q\le j+m,$$
$$n=k+i+\ell-q.$$

3. Calculation of $J_{im}^{(0)}$

It has been shown that

$$J_{im}^{(0)}=\int I_i^{(0)}S_{1/2}^m(W_i^2)d\vec{v}_i, \quad (B51)$$

where

$$I_i^{(0)}=\sum_{j=1}^{N}\int\int\int(f_i'^{(0)}f_j'^{(0)}-f_i^{(0)}f_j^{(0)})gb\,db\,d\epsilon\,d\vec{v}_j. \quad (B52)$$

Note that the calculation has been performed for two colliding particles i and j with different temperatures. Following the previous expansions, it can be shown that

$$J_{im}^{(0)}=-\sum_{j=1}^{N}8n_in_j\frac{(m_i+m_j)^3}{(m_im_j)^{3/2}}(M_{i1}M_{i2})^{3/2}\theta_{ij}^{3/2}$$
$$\times\sum_{mr'\ell'}D_{mr'\ell'}\Omega_{ij}^{(\ell',r')}. \quad (B53)$$

$D_{mr'\ell'}$ are numbers depending on m_i, m_j, T_i, and T_j and are obtained using the following expression:

$$\sum_{mr'\ell'} D_{mr'\ell'} \gamma^{2r'} \cos^{\ell'} \chi$$

$$= \sum_{rk\ell} \frac{\gamma^{2r}}{(r-k)!k!\ell!} [-M_{i2} - M_{i1}M_{i2}(1-\theta_{ij})^2$$

$$+ 2M_{i1}M_{i2}(1-\theta_{ij})\cos\chi]^k [M_{i1}M_{i2}(1-\theta_{ij})^2]^{r-k}$$

$$\times (1-M_{i1})^{\ell} (r + \tfrac{1}{2} + \ell)_{\ell}. \quad \text{(B54)}$$

The conditions of summation are the following. For the left-hand side of Eq. (B54),

$$r', \ell' \leqslant m.$$

For the right-hand side of Eq. (B54),

$$0 \leqslant k \leqslant r \leqslant m,$$

$$0 \leqslant \ell \leqslant m,$$

$$\ell = m - k.$$

[1] M. I. Boulos, P. Fauchais, and E. Pfender, *Thermal Plasmas: Fundamentals and Application* (Plenum, New York, 1994), Vol. 1.
[2] P. Fauchais and A. Vardelle, IEEE Trans. Plasma Sci. **25**, 1258 (1997).
[3] E. Pfender, Plasma Chem. Plasma Process. **19**, 1 (1999).
[4] E. Pfender, J. Thermal Spray Technol. **6**, 126 (1997).
[5] J. O. Hirschfelder, C. F. Curtis, and R. B. Bird, *Molecular Theory of Gases and Liquids*, 2nd ed. (Wiley, New York, 1964).
[6] S. Chapman and T. G. Cowling, *The Mathematical Theory of Non-Uniform Gases*, 3rd ed. (Cambridge University Press, Cambridge, England, 1970).
[7] A. B. Murphy and C. J. Arundell, Plasma Chem. Plasma Process. **14**, 451 (1994).
[8] A. B. Murphy, Plasma Chem. Plasma Process. **15**, 279 (1995).
[9] M. Capitelli, R. Celiberto, and C. Gorse, Plasma Chem. Plasma Process. **16**, 267 (1996).
[10] A. B. Murphy, IEEE Trans. Plasma Sci. **25**, 809 (1997).
[11] B. V. Alexeev, A. Chikhaoui, and I. T. Grushin, Phys. Rev. E **49**, 2809 (1994).
[12] A. B. Murphy, Phys. Rev. E **48**, 3594 (1993).
[13] A. B. Murphy, Phys. Rev. E **55**, 7473 (1997).
[14] S. C. Snyder, A. B. Murphy, D. L. Hofeldt, and L. D. Reynolds, Phys. Rev. E **52**, 2999 (1995).
[15] A. B. Murphy, J. Phys. D **29**, 1922 (1996).
[16] P. André, IEEE Trans. Plasma Sci. **23**, 453 (1995).
[17] P. André, M. Abbaoui, A. Lefort, and M. J. Parizet, Plasma Chem. Plasma Process. **16**, 379 (1996).
[18] J. Aubreton, M. F. Elchinger, and P. Fauchais, Plasma Chem. Plasma Process. **18**, 1 (1998).
[19] P. Fauchais, M. F. Elchinger, and J. Aubreton, J. High. Temp. Mater. Process. **4**, 21 (2000).
[20] V. Rat, P. André, J. Aubreton, M. F. Elchinger, P. Fauchaisr, and A. Lefort, J. Phys. D **34**, 1 (2001).
[21] A. J. D. Farmer and G. N. Haddad, J. Phys. D **21**, 432 (1988).
[22] J. Haidar, J. Phys. D **28**, 2494 (1995).
[23] Manabu Tanaka and Masao Ushio, J. Phys. D **32**, 1153 (1999).
[24] M. Bouaziz, A. Gleizes, and M. Razafinimanana, J. Appl. Phys. **84**, 4128 (1998).
[25] J. Heberlein, Ann. N. Y. Acad. Sci. **891**, 14 (2000).
[26] P. André, J. Ondet, R. Pellet, and A. Lefort, J. Phys. D **30**, 2043 (1997).
[27] W. L. T. Chen, J. Herberlein, and E. Pfender, *Diagnostics of Non-equilibrium Situations in Thermal Plasma Spray Jets*, in *Thermal Spray: Advances in Coating Technology*, edited by C. C. Berndt (ASM Int., Ohio, 1992), pp. 327–336.
[28] D. M. Chen and E. Pfender, IEEE Trans. Plasma Sci. **9**, 265 (1981).
[29] K. Charrada, G. Zissis, and Aubes, J. Phys. D **29**, 2432 (1996).
[30] J. Jenista, J. V. R. Herberlein, and E. Pfender, IEEE Trans. Plasma Sci. **25**, 883 (1997).
[31] M. Mitchner and C. H. Kruger, *Partially Ionized Gases* (Wiley, New York, 1973).
[32] L. Spitzer and R. Härm, Phys. Rev. **89**, 977 (1952).
[33] J. Haidar, J. Phys. D **32**, 263 (1999).
[34] R. S. Devoto, Ph.D. thesis, Stanford University (1965).
[35] R. S. Devoto, Phys. Fluids **10**, 2105 (1967).
[36] J. J. Gonzales, R. Girard, and A. Gleizes, J. Phys. D **33**, 2759 (2000).
[37] C. Bonnefoi, state thesis, University of Limoges, France (1983).
[38] J. Aubreton, C. Bonnefoi, and J. M. Mexmain, Rev. Phys. Appl. **21**, 365 (1986).
[39] V. Rat, J. Aubreton, M. F. Elchinger, and P. Fauchais, Plasma Chem. Plasma Process. **21**, 355 (2001).
[40] A. B. Murphy and K. Hiraoka, J. Phys. D **33**, 2183 (2000).
[41] G. Jansen, Ph.D. thesis, Eindhoven University of Technology (2000).
[42] J. D. Ramshaw, J. Non-Equilib. Thermodyn. **18**, 121 (1993).
[43] A. F. Kolesnikov and G. A. Tirskii, in *Proceedings: Models in the Mechanics of Continuum Media* (Institute of Theoretical and Applied Mechanics, Siberian Branch of the Russian Academy of Science, Novosibirsk, 1979), p. 114.
[44] A. F. Kolesnikov, Technical note 196, Von Karman Institute for Fluid Dynamics, St Genesius-Rode, Belgium (1999).
[45] I. Prigogine, Bull. Cl. Sci., Acad. R. Belg. **26**, 53 (1940).
[46] A. V. Potapov, High Temp. **4**, 55 (1966).
[47] E. Richley and D. T. Tuma, J. Appl. Phys. **53**, 8537 (1982).
[48] A. Morro and M. Romeo, J. Plasma Phys. **39**, 41 (1988).
[49] M. C. M. van de Sanden, P. P. J. M. Schram, A. G. Peeters, J.

A. M. van der Mullen, and G. M. W. Kroesen, Phys. Rev. A **40**, 5273 (1989).

[50] D. Giordano and M. Capitelli, J. Thermophys. Heat Transfer **9**, 803 (1995).

[51] Xi Chen and Peng Han, J. Phys. D **32**, 1711 (1999).

[52] F. R. W. Mc Court, J. J. M. Beenakker, W. E. Köhler, and I. Kuščer, *Nonequilibrium Phenomena in Polyatomic Gases* (Clarendon, Oxford, 1990), Vol. 1.

[53] R. S. Devoto, Phys. Fluids **9**, 1230 (1966).