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Discussion on the transport processes in electrons with non-Maxwellian energy distribution function in partially-ionized plasmas

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Abstract.

In a previous work [1], we have developed a non-linear moment model for electrons that self-consistently captures non-Maxwellian electron energy distribution function (EEDF) effects. The model does not rely in the local approximation and the transport coefficients are calculated by expanding the distribution function into Hermite polynomials and by taking moments of the Boltzmann equation, including the collision operator for elastic and inelastic collisions with arbitrary cross sections. This model captures the classical Fick's, Fourier's, and Ohm's law as well as Soret, Dufour, and Peltier effects. In addition, novel non-local transport phenomena appear as a result of spatial gradients of the kurtosis of the distribution function. In this paper, we discuss on the transport effects by analyzing two collisional models: constant collision frequency and constant cross section. We estimate the order of magnitude of the transport processes in non-equilibrium electrons by analyzing the Langmuir probe measurements of a low-pressure argon inductively-coupled discharge. The results show that, under these conditions, the transport produced by the spatial gradients in the kurtosis of the distribution function produces a heat-flux contribution that is of the same order of magnitude as the Fourier and Dufour's effects. These transport effects are beyond the local field or the electron gradient expansions, commonly used in the low-temperature plasma modelling.

1. Introduction

Understanding the transport of mass, momentum, and energy in multicomponent plasmas is a fundamental problem that concerns all areas of plasma physics, from low-temperature plasmas [2, 3, 4] and magnetically-confined fusion plasmas [5, 6] to space plasmas [7]. In this paper, we will focus on the collisional transport of electrons in partially-ionized plasmas. In particular, we study the transport processes of electrons under non-equilibrium conditions where the electron energy distribution function (EEDF) is not Maxwellian. This loss of thermal equilibrium under collisional

conditions can be a result of the collisions with the other species (elastic and inelastic), spatial gradients or the presence of an electromagnetic field. Measurements of non-Maxwellian EEDFs are found in gas discharges [8], in the core distribution function of the solar wind [9, 10, 11] and observed in kinetic simulations of the scrape-off layer of tokamaks [12].

The transport of electrons with non-Maxwellian EEDFs is a long-lived problem in gas discharges and swarms [13, 14]. In order to develop fluid models that describe the transport of non-equilibrium electrons, a common approach is to compute the transport coefficients from the solution of the spatially-homogeneous Boltzmann equation in the “two-term” approximation [15, 14]. This approach is usually referred to as “local-field approximation” as it assumes a local equilibrium between the local electric field and the collisions. The transport fluxes are usually expanded in series of the density gradient [4] or, alternatively, by including an ansatz that includes the mean energy gradient [15, 14]. In addition to swarms and gas discharges, the two-term approximation has also been used to study the transport coefficients in fully-ionized heliospheric plasmas. In these plasmas, the EEDFs are also measured as non-Maxwellian with populated tails, usually well-fitted with Kappa distributions [16]. The influence of the suprathermal particles on the transport coefficients has been studied by Husidic et al. [17] by considering a fully-ionized plasma and a Krook-type collisional operator.

Alternatively, the collisional transport in multi-component plasmas has been traditionally studied with Grad’s method [18] as explained in seminal works by Braginskii [19], Balescu [20] and Zhdanov [21] or more recently by Hunana et al. [22], Simakov [23], and Raghunathan et al. [24]. The Grad’s method considers velocity moments of the kinetic equation by expanding the distribution function into series of Hermite polynomials. In most of these works [19, 20, 21, 23, 24], the energy distribution function is considered to be Maxwellian and only perturbations in the first and second spherical harmonics components of the distribution function in the velocity space are considered.

In a recent work [1], we have developed an electron fluid model based on a regularized Grad’s method that includes the perturbations in the energy distribution function. The model considers electron-electron, electron-ion and electron-neutral elastic collisions as well as inelastic and ionization collisions. As opposed to the “local-field approximation”, the model does not rely in the solution of a spatially-homogenous Boltzmann solver nor it expands the transport fluxes into series of the density gradient. Instead, the model is based on the resolution of the scalar and vectorial moments of the Boltzmann equation up to the contracted fourth-order moment. As a result, the transport coefficients are self-consistently calculated with the Chapman-Cowling collision integrals as well as novel transport fluxes appear as a result of non-local transport phenomena.

In this paper, we first summarize the set of regularized 9M equations. After, we present a discussion on the transport phenomena. The general equations can be simplified and obtain analytical expressions, for two collision models: for a constant

collision frequency and a constant cross section. Finally, we estimate the order of magnitude of the transport fluxes in an ICP argon discharge at 5 mTorr.

2. Summary of the derivation of the model

We now proceed to briefly describe the derivation of the transport model of the regularized moment equations. A detailed derivation can be found in Ref. [1].

2.1. Kinetic equation

We consider the electron kinetic equation in a partially-ionized plasma that reads as follows:

$$\frac{\partial f_{\mathbf{e}}}{\partial t} + \mathbf{v} \cdot \nabla f_{\mathbf{e}} - \frac{e\mathbf{E}}{m_{\mathbf{e}}} \cdot \nabla_{\mathbf{v}} f_{\mathbf{e}} = \sum_{\alpha}^{\mathbf{i}, \mathbf{g}, \mathbf{e}} \left. \frac{\delta f_{\mathbf{e}}}{\delta t} \right|_{\mathbf{e}\alpha}^{\mathbf{el}} + \left. \frac{\delta f_{\mathbf{e}}}{\delta t} \right|_{\mathbf{e}\mathbf{g}}^{\mathbf{exc}} + \left. \frac{\delta f_{\mathbf{e}}}{\delta t} \right|_{\mathbf{e}\mathbf{g}}^{\mathbf{iz}}. \quad (1)$$

Here, the subscripts \mathbf{e} , \mathbf{i} , and \mathbf{g} stands for the electron, ion, and gas species (where the excited states are not tracked), respectively. The collision terms include elastic collisions, including electron-electron, electron-ion, and electron-neutral, with a Boltzmann collision operator; inelastic collisions with a Wang Chan-Uhlenbeck [25]; the inelastic collisions with a simplified Giovangigli collision operator [26] that considers the recombination reaction to be negligible, as usually done in low-temperature plasmas, e.g., see Robson and Ness [27]. The ions and neutrals are assumed to be at rest and in thermodynamic equilibrium at a different temperature from the electrons.

2.2. Hermitian expansion and the two-term approximation

In Grad's method [18], the distribution function is approximated by a Maxwellian distribution function with a polynomial perturbation,

$$f_{\mathbf{e}}(\mathbf{x}, \mathbf{c}_{\mathbf{e}}, t) = n_{\mathbf{e}} \left(\frac{m_{\mathbf{e}}}{2\pi e T_{\mathbf{e}}} \right)^{3/2} e^{-\frac{m_{\mathbf{e}} \mathbf{c}_{\mathbf{e}}^2}{2e T_{\mathbf{e}}}} [1 + \chi_{\mathbf{e}}(\mathbf{c}_{\mathbf{e}})], \quad \text{with } \mathbf{c}_{\mathbf{e}} = \mathbf{v} - \mathbf{u}_{\mathbf{e}}. \quad (2)$$

The perturbation is a truncated series of Hermite polynomials [20],

$$\chi_{\mathbf{e}}(\mathbf{c}_{\mathbf{e}}) = \sum_{n=0}^N h^{(2n)} H^{(2n)}(\mathbf{c}_{\mathbf{e}}) + \sum_{n=0}^M h_r^{(2n+1)} H_r^{(2n+1)}(\mathbf{c}_{\mathbf{e}}) + \sum_{n=0}^P h_{rs}^{(2n+2)} H_{rs}^{(2n+2)}(\mathbf{c}_{\mathbf{e}}) + \dots \quad (3)$$

The superscript stands for the order of the polynomial whereas the subscript refers to the tensorial rank. N , M , P ... are the number of polynomials considered in the truncation.

The irreducible Hermite polynomials can be written as a function of the Laguerre-Sonine polynomials and spherical harmonics,

$$\chi_{\mathbf{e}}(\mathbf{c}_{\mathbf{e}}) = \sum_{n=0}^N b_n^0 L_n^{1/2}(c_{\mathbf{e}}) + \sum_{n=0}^M \sum_{m=-1}^1 b_n^1 L_n^{3/2}(c_{\mathbf{e}}) c_{\mathbf{e}} Y_1^m + \sum_{n=0}^P \sum_{m=-2}^2 b_n^2 L_n^{5/2}(c_{\mathbf{e}}) c_{\mathbf{e}}^2 Y_2^m + \dots$$

Here, $L_n^{l+1/2}$ are the Laguerre-Sonine polynomials and Y_l^m are the spherical harmonics. Written in this form, we can identify that the expansion in the polynomials $H^{(2n)}$ are responsible for the isotropic part of the distribution function, $H_r^{(2n+1)}$ for the first anisotropy, and $H_{rs}^{(2n+2)}$ for the second anisotropy. In conclusion, the reader can see the equivalence between the Grad's and the polynomial expansion that is used in multi-term Boltzmann solvers for low-temperature plasmas and swarms [4, 27, 28]. For this reason, in the present paper, we consider a distribution function with the same perturbation as in the two-term approximation, i.e., scalar $h^{(2n)}$ and vectorial moments $h_r^{(2n+1)}$. As in the two-term expansion, no tensorial polynomials $H_{rs}^{(2n+2)}$ are taken into account in this theory. In this work, we consider moments up to the fourth moment, i.e., $N = 2$, $M = 1$ (the so-called 9M equations).

The set of equations studies the evolution of nine fields: density, velocity vector, energy, heat-flux vector and contracted (trace) fourth-moment. The moment variables are computed from the distribution function as

$$\begin{aligned} n_\epsilon &= \int f_\epsilon d\mathbf{v}, & m_\epsilon n_\epsilon u_{\epsilon i} &= \int m_\epsilon v_i f_\epsilon d\mathbf{v}, & p_\epsilon &= \frac{1}{3} \int m_\epsilon c_\epsilon^2 f_\epsilon d\mathbf{v}, \\ q_{\epsilon i} &= \frac{1}{2} \int m_\epsilon c_\epsilon^2 c_{\epsilon i} f_\epsilon d\mathbf{v}, & \text{and } p_\epsilon^{(4)} &= \frac{1}{2} \int m_\epsilon c_\epsilon^4 f_\epsilon d\mathbf{v}. \end{aligned} \quad (4)$$

We use the normalized contracted fourth-moment that measures the deviations of the fourth-order moment from a Maxwellian, i.e., excess kurtosis of the distribution function,

$$\Delta_\epsilon = \frac{p_\epsilon^{(4)} - \frac{15}{2} \frac{n_\epsilon}{e} T_\epsilon^2}{m_\epsilon} \frac{15 n_\epsilon e^2 T_\epsilon^2}{2 m_\epsilon}. \quad (5)$$

With these definitions and Grad's non-equilibrium distribution function, the moment evolution equations as well as the collisional exchange terms can be exactly computed. We refer the interested reader to Ref. [1] where the 9-M set of moment equations is presented.

The Grad's non-equilibrium distribution function for this choice of moments can be decomposed into an isotropic part of the distribution function and an anisotropic part along the first spherical harmonic, as follows:

$$f_\epsilon^{(9M)}(\mathbf{x}, \mathbf{v}_\epsilon, t) = n_\epsilon \left(\frac{m_\epsilon}{2\pi e T_\epsilon} \right)^{3/2} e^{-\frac{m_\epsilon v_\epsilon^2}{2eT_\epsilon}} \left[1 + \chi_{isot}^{(9M)}(v_\epsilon) + \chi_{anisot}^{(9M)}(\mathbf{v}_\epsilon) \right] \quad (6)$$

where

$$\chi_{isot}^{(9M)}(v_\epsilon) = \sum_{n=0}^2 h^{(2n)} H^{(2n)}(v_\epsilon) = \left(\frac{15}{8} - \frac{5\beta_\epsilon}{2} v_\epsilon^2 + \frac{\beta_\epsilon^2}{2} v_\epsilon^4 \right) \Delta_\epsilon \quad (7)$$

and

$$\chi_{anisot}^{(9M)}(\mathbf{v}_\epsilon) = \sum_{n=0}^1 h_r^{(2n+1)} H_r^{(2n+1)}(\mathbf{v}_\epsilon) = 2\beta_\epsilon u_{\epsilon i} v_{\epsilon i} + \frac{8\beta_\epsilon^2}{5m_\epsilon n_\epsilon} q_{\epsilon i} v_{\epsilon i} \left(\beta_\epsilon v_\epsilon^2 - \frac{5}{2} \right). \quad (8)$$

Here, $\beta_\epsilon = m_\epsilon / (2eT_\epsilon)$, and we have assumed $u_\epsilon \ll \sqrt{eT_\epsilon/m_\epsilon}$, as explained in [1].

The reader can note that the 9-M distribution function can be written as in the two-term Boltzmann expansion as:

$$f_{\epsilon}^{(9M)}(\mathbf{x}, \mathbf{v}_{\epsilon}, t) = f_{\epsilon}^0(\mathbf{x}, v_{\epsilon}, t) + \frac{\mathbf{v}_{\epsilon}}{v_{\epsilon}} \cdot \mathbf{f}_{\epsilon}^1(\mathbf{x}, v_{\epsilon}, t) \quad (9)$$

with

$$f_{\epsilon}^0(\mathbf{x}, v_{\epsilon}, t) = n_{\epsilon} \left(\frac{m_{\epsilon}}{2\pi e T_{\epsilon}} \right)^{3/2} e^{-\frac{m_{\epsilon} v_{\epsilon}^2}{2e T_{\epsilon}}} \left\{ 1 + \left(\frac{15}{8} - \frac{5\beta_{\epsilon}}{2} v_{\epsilon}^2 + \frac{\beta_{\epsilon}^2}{2} v_{\epsilon}^4 \right) \Delta_{\epsilon} \right\}, \quad (10)$$

$$\mathbf{f}_{\epsilon}^1(\mathbf{x}, v_{\epsilon}, t) = n_{\epsilon} v_{\epsilon} \left(\frac{m_{\epsilon}}{2\pi e T_{\epsilon}} \right)^{3/2} e^{-\frac{m_{\epsilon} v_{\epsilon}^2}{2e T_{\epsilon}}} \left\{ 2\beta_{\epsilon} \mathbf{u}_{\epsilon} + \frac{8\beta_{\epsilon}^2}{5m_{\epsilon} n_{\epsilon}} \left(\beta_{\epsilon} v_{\epsilon}^2 - \frac{5}{2} \right) \mathbf{q}_{\epsilon} \right\}. \quad (11)$$

2.3. Regularization of equations

The original system of Grad's moment equations has no parabolic terms in the equations, which can lead to spurious discontinuities. For this reason, different authors have proposed the regularization (or parabolization) of the Grad's equations [29, 30, 31] by introducing a Chapman-Enskog expansion of the moment variables in series of the Knudsen number.

Following the methodology proposed in previous regularized models, the study of electrons in a partially-ionized plasma allows for additional considerations in the Chapman-Enskog expansion. In particular, the electron-to-atom mass ratio can be included in the asymptotic analysis [32], as well as the different Knudsen numbers (corresponding to the different collision interactions). As a result, we consider the following expansion for the variables in terms of the electron-gas Knudsen number,

$$\begin{aligned} \mathbf{u}_{\epsilon} &= \mathbf{u}_{\epsilon}^0 + \text{Kn}_{\epsilon\text{g}} \mathbf{u}_{\epsilon}^1 + \dots, \\ \mathbf{q}_{\epsilon} &= \mathbf{q}_{\epsilon}^0 + \text{Kn}_{\epsilon\text{g}} \mathbf{q}_{\epsilon}^1 + \dots. \end{aligned}$$

We consider the following ordering in Knudsen number, characteristic of weakly-ionized plasmas, that scales with the electron-to-atom mass ratio $\varepsilon = m_{\epsilon}/m_{\text{h}}$ as,

$$\text{Kn}_{\epsilon\text{g}} = \mathcal{O}(\varepsilon^{1/2}) < \text{Kn}_{\epsilon\epsilon} \ll \text{Kn}_{\epsilon\text{g}}^{\text{inel}}. \quad (12)$$

By following the procedure described in Ref. [1], we find that the moment equations can be regularized. Whereas the density, energy, and fourth moment are described by a non-linear mixed hyperbolic-parabolic equations, the electron particle and heat-fluxes are described as transport fluxes that are function of the gradients of the density, pressure, and fourth moment and the electric field. In this manner, as considered in the two-term approximation, the anisotropic part of the distribution function is assumed to be small as compared to the isotropic part.

3. System of regularized moment equations with non-Maxwellian EEDFs

We summarize the set of equations and transport model that was proposed in Ref. [1]. The general expression of the collision frequencies and the transport coefficients for arbitrary collision cross sections are given in Appendix A.

Conservation equations

$$\frac{\partial n_{\epsilon}}{\partial t} + \nabla \cdot (n_{\epsilon} \mathbf{u}_{\epsilon}) = \dot{n}_{\epsilon}, \quad (13)$$

$$\frac{3}{2} \frac{\partial p_{\epsilon}}{\partial t} + \nabla \cdot \left(\frac{5}{2} p_{\epsilon} \mathbf{u}_{\epsilon} + \mathbf{q}_{\epsilon} \right) = -en_{\epsilon} \mathbf{u}_{\epsilon} \cdot \mathbf{E} - \dot{Q}_{elast} - \dot{Q}_{(excit,iz)}, \quad (14)$$

$$\frac{\partial p_{\epsilon}^{(4)}}{\partial t} + \nabla \cdot \left(\frac{7}{3} p_{\epsilon}^{(4)} \mathbf{u}_{\epsilon} + 14 \frac{p_{\epsilon}}{\rho_{\epsilon}} \mathbf{q}_{\epsilon} \right) = -4 \frac{e}{m_{\epsilon}} \mathbf{q}_{\epsilon} \cdot \mathbf{E} - 10 \frac{e}{m_{\epsilon}} p_{\epsilon} \mathbf{u}_{\epsilon} \cdot \mathbf{E} + \dot{Q}_{ee}^{(4)} - \dot{Q}_{elast}^{(4)} - \dot{Q}_{(excit,iz)}^{(4)} \quad (15)$$

Transport fluxes

$$\mathbf{u}_{\epsilon} = -D_{\epsilon} \left(\frac{1}{n_{\epsilon}} \nabla n_{\epsilon} + (1 + \chi_{\epsilon}) \nabla \ln T_{\epsilon} + \alpha_{\epsilon} \nabla \Delta_{\epsilon} \right) - \mu_{\epsilon} \mathbf{E}, \quad (16)$$

$$\mathbf{q}_{\epsilon} = \Lambda_{\epsilon} n_{\epsilon} \mathbf{u}_{\epsilon} - \kappa_{\epsilon} \nabla T_{\epsilon} - \vartheta_{\epsilon} \nabla n_{\epsilon} - \varkappa_{\epsilon} \nabla \Delta_{\epsilon}. \quad (17)$$

Collision exchange terms

$$\dot{n}_{\epsilon} = n_{\epsilon} n_{\mathbf{g}} K_{iz}^{(0)} \quad (18)$$

$$\dot{Q}_{elast} = \sum_{\alpha}^{i,g} \left\{ \frac{m_{\epsilon}}{m_{\alpha}} n_{\epsilon} \nu_{\epsilon\alpha}^{(T,2)} e (T_{\epsilon} - T_{\alpha}) + \frac{m_{\epsilon}}{m_{\alpha}} n_{\epsilon} \nu_{\epsilon\alpha}^{(\Delta,2)} \Delta_{\epsilon} e T_{\alpha} \right\} \quad (19)$$

$$\dot{Q}_{(excit,iz)} = \sum_k^{(excit,iz)} n_{\epsilon} \nu_{\epsilon\mathbf{g}}^{(0,k)} e \phi_k^* \quad (20)$$

$$\dot{Q}_{ee}^{(4)} = -\frac{p_{\epsilon}^2}{\rho_{\epsilon}} \nu_{ee}^{\Delta} \Delta_{\epsilon} - 4 \nu_{ee}^q q_{\epsilon k} u_{\epsilon k} \quad (21)$$

$$\dot{Q}_{elast}^{(4)} = \sum_{\alpha}^{i,g} \left\{ \frac{m_{\epsilon}}{m_{\alpha}} \nu_{\epsilon\alpha}^{(T,4)} \frac{p_{\epsilon}^2}{\rho_{\epsilon}} \left(1 - \frac{T_{\alpha}}{T_{\epsilon}} \right) + \nu_{\epsilon\alpha}^{(\Delta,4)} \Delta_{\epsilon} \frac{p_{\epsilon}^2}{\rho_{\epsilon}} \frac{T_{\alpha}}{T_{\epsilon}} \right\} \quad (22)$$

$$\dot{Q}_{(excit,iz)}^{(4)} = 2 \left(\frac{p_{\epsilon}^2}{\rho_{\epsilon}} \right) n_{\mathbf{g}} \sum_k^{(excit,iz)} \left(2K_k^{(1)} \left(\frac{\phi_k^*}{T_e} \right) - K_k^{(0)} \left(\frac{\phi_k^*}{T_e} \right)^2 \right) \quad (23)$$

4. Discussion of the transport equations

The transport phenomena appearing in the fluxes of Eqs. (16)-(17) are summarized in Table 1, with the analytical expressions of the transport coefficients in Appendix A.2, for general cross sections interaction. We can identify some of the classical transport effects in multicomponent plasmas: Fick's, Ohm's, and Fourier's law, and Peltier, Soret and Dufour effects. In addition, two novel transport effects appear that are consequence of the spatial gradients in the kurtosis of the EEDF. The general equations can be reduced to less complicated expressions if the cross section varies in a simple way with velocity. For this reason, we consider two simplified cases: constant electron-gas collision frequency, i.e., cross-section has a dependence as $\sigma_{\epsilon\mathbf{g}} \propto v_{\epsilon}^{-1}$ (Maxwell's

Table 1. Summary of the transport effects that are captured by the regularized 9-M model. The analytical expressions for a general interaction is in Appendix A.2.

	Transport effect	Associated transport coefficient	Thermodynamic force
Particle flux	Fick's law	D_{ϵ}	∇n_{ϵ}
	Thermophoresis or Soret effect	χ_{ϵ}	∇T_{ϵ}
	Ohm's law	μ_{ϵ}	\mathbf{E}
	Novel non-local effect	α_{ϵ}	$\nabla \Delta_{\epsilon}$
Heat flux	Peltier effect	Λ_{ϵ}	$n_{\epsilon} \mathbf{u}_{\epsilon}$
	Fourier's law	κ_{ϵ}	∇T_{ϵ}
	Dufour effect	ϑ_{ϵ}	∇n_{ϵ}
	Novel non-local effect	\varkappa_{ϵ}	$\nabla \Delta_{\epsilon}$

molecules or Langevin interaction), and constant cross section (hard sphere interaction). Both cases can be interpreted in terms of interparticle forces. The constant collision frequency corresponds to a polarizing force between the electron and the neutral particle. Alternatively, the constant cross section model is preferred as it can approximate the elastic collision between an electron and a gas atom for limited energy ranges, in most common gases.

4.1. Constant electron-neutral collision frequency

We consider the first the case where the electron-gas collision frequency is constant $\bar{\nu}_{\epsilon g}$, which corresponds to the Langevin interaction or the so-called Maxwell's molecules. The momentum transfer cross section is $Q_{\epsilon g}^{(1)} = \bar{\nu}_{\epsilon g} / (n_g |\mathbf{v}_{\epsilon} - \mathbf{v}_g|)$.

As a result, the collisional frequencies read:

$$\begin{aligned} \nu_{\epsilon g}^{(u,1)} = \nu_{\epsilon g}^{(q,3)} = \bar{\nu}_{\epsilon g}, \quad \nu_{\epsilon g}^{(q,1)} = \nu_{\epsilon g}^{(\Delta,2)} = 0, \\ \nu_{\epsilon g}^{(T,2)} = 3\bar{\nu}_{\epsilon g}, \quad \nu_{\epsilon g}^{(u,3)} = \frac{5}{2}\bar{\nu}_{\epsilon g}, \quad \nu_{\epsilon g}^{(T,4)} = 30(1 + \Delta_{\epsilon})\bar{\nu}_{\epsilon g}, \quad \nu_{\epsilon g}^{(\Delta,4)} = 30\bar{\nu}_{\epsilon g}. \end{aligned} \quad (24)$$

The transport coefficients read

$$\begin{aligned} D_{\epsilon} = \frac{eT_{\epsilon}}{m_{\epsilon}\bar{\nu}_{\epsilon g}}, \quad \mu_{\epsilon} = \frac{e}{m_{\epsilon}\bar{\nu}_{\epsilon g}}, \quad \chi_{\epsilon} = \alpha_{\epsilon} = \Lambda_{\epsilon} = 0, \\ \kappa_{\epsilon} = \frac{5}{2} \frac{p_{\epsilon} e}{m_{\epsilon}\bar{\nu}_{\epsilon g}} \frac{1 + 2\Delta_{\epsilon}}{1 + \frac{\nu_{\epsilon\epsilon}^a}{\bar{\nu}_{\epsilon g}}}, \quad \vartheta_{\epsilon} = \frac{5}{2} \frac{(eT_{\epsilon})^2}{m_{\epsilon}\bar{\nu}_{\epsilon g}} \frac{\Delta_{\epsilon}}{1 + \frac{\nu_{\epsilon\epsilon}^a}{\bar{\nu}_{\epsilon g}}}, \quad \varkappa_{\epsilon} = \frac{5}{2} \frac{n_{\epsilon} (eT_{\epsilon})^2}{m_{\epsilon}\bar{\nu}_{\epsilon g}} \frac{1}{1 + \frac{\nu_{\epsilon\epsilon}^a}{\bar{\nu}_{\epsilon g}}}. \end{aligned} \quad (25)$$

Note that the thermophoresis, the novel non-local effect in the flux of particles, and the Peltier effects are lost. This can be explained due to the fact that as the collision frequency does not depend on the energy of the electrons, the cross-effects that couple the high-energy and lower-energy electrons disappear.

4.1.1. Weakly-ionized plasma We consider a case that where the electron-gas elastic frequency is much larger than the electron-electron collision, i.e., $\bar{\nu}_{\epsilon g} \gg \nu_{\epsilon\epsilon}$. The set of

moment equations for constant electron-neutral collision frequency reads

$$\frac{\partial n_\epsilon}{\partial t} - \nabla \cdot \left(\frac{e}{m_\epsilon \bar{\nu}_{\epsilon g}} n_\epsilon \mathbf{E} + \frac{1}{m_\epsilon \bar{\nu}_{\epsilon g}} \nabla p_\epsilon \right) = \dot{n}_\epsilon, \quad (26)$$

$$\begin{aligned} \frac{3}{2} \frac{\partial p_\epsilon}{\partial t} - \nabla \cdot \left(\frac{5}{2} \frac{e}{m_\epsilon \bar{\nu}_{\epsilon g}} p_\epsilon \mathbf{E} + \frac{1}{3 \bar{\nu}_{\epsilon g}} \nabla p_\epsilon^{(4)} \right) = \\ \frac{e^2 n_\epsilon}{m_\epsilon \bar{\nu}_{\epsilon g}} E^2 + \frac{e}{m_\epsilon \bar{\nu}_{\epsilon g}} \nabla p_\epsilon \cdot \mathbf{E} + 3 \frac{m_\epsilon}{m_g} n_\epsilon \bar{\nu}_{\epsilon g} e (T_g - T_\epsilon) - \dot{Q}_{(excit,iz)}, \end{aligned} \quad (27)$$

$$\begin{aligned} \frac{\partial p_\epsilon^{(4)}}{\partial t} - \nabla \cdot \left(\frac{7}{3} \frac{e}{m_\epsilon \bar{\nu}_{\epsilon g}} p_\epsilon^{(4)} \mathbf{E} + \frac{14}{3} \frac{e T_\epsilon}{m_\epsilon \bar{\nu}_{\epsilon g}} \nabla p_\epsilon^{(4)} + \left(\frac{7}{3} p_\epsilon^{(4)} - 35 \frac{p_\epsilon^2}{\rho_\epsilon} \right) \frac{1}{m_\epsilon n_\epsilon \bar{\nu}_{\epsilon g}} \nabla p_\epsilon \right) = \\ 10 \frac{e^2 p_\epsilon}{m_\epsilon^2 \bar{\nu}_{\epsilon g}} E^2 + \frac{4}{3} \frac{e}{m_\epsilon \bar{\nu}_{\epsilon g}} \nabla p_\epsilon^{(4)} \cdot \mathbf{E} + 30 \frac{m_\epsilon}{m_g} \bar{\nu}_{\epsilon g} \frac{p_\epsilon^2}{\rho_\epsilon} \left(\frac{T_g}{T_\epsilon} - (1 + \Delta_\epsilon) \right) - \dot{Q}_{(excit,iz)}^{(4)}. \end{aligned} \quad (28)$$

The system of transport equations is simpler than the original system that considers a collision frequency that depends on the collision relative velocity. It is interesting to note that the scalar (density, energy, fourth moment) moment equations present a similar structure: an advection term due to the electric field that is proportional to the mobility, a diffusion term, and a dissipation term. The diffusion term couples the equation with the subsequent moment, except the fourth moment equation that due to the truncation is coupled to the energy. The dissipation terms are similar in both the energy and fourth moment equations: with a term that is proportional to the square of the electric field, a dissipation term proportional to the scalar product of the gradient of the moment and the electric field, and the collisional exchange with the neutral gas. Note that the average energy exchange due to elastic collisions reads the form that is proportional to $3\bar{\nu}_{\epsilon g}$, as in the case of Maxwellian EEDF, c.f., e.g., Ref. [21].

Concerning the transport coefficients in the fluxes of particles and energy of Eqs. (25), one can find that, although we obtained the equations with a methodology that is completely different to the two-term Boltzmann equation, we find the same mobility and diffusion coefficient as in the two-term Boltzmann approximation. In a DC case, the two-term Boltzmann theory shows that the mobility and the diffusion coefficients depend on the isotropic distribution function and the momentum transfer frequency, as follows:

$$\mu_\epsilon = \frac{4\pi e}{3m_\epsilon} \int_0^\infty \frac{d}{dv_\epsilon} \left(\frac{v_\epsilon^3}{\nu_m(v_\epsilon)} \right) \frac{f_\epsilon^0}{n_\epsilon} dv_\epsilon \quad \text{and} \quad D_\epsilon = \frac{4\pi}{3} \int_0^\infty \frac{v_\epsilon^4}{\nu_m(v_\epsilon)} \frac{f_\epsilon^0}{n_\epsilon} dv_\epsilon, \quad (29)$$

with the momentum transfer collision frequency $\nu_m(v_\epsilon) = n_g Q_{\epsilon g}^{(1)} v_\epsilon$. In the case of constant collision frequency, i.e., $\nu_m(v_\epsilon) \equiv \bar{\nu}_{\epsilon g}$, and injecting the definitions of Eqs. (10)-(11), we obtain,

$$\begin{aligned} \mu_\epsilon &= \frac{e}{m_\epsilon \bar{\nu}_{\epsilon g}} \int_0^\infty 4\pi v_\epsilon^2 \frac{f_\epsilon^0}{n_\epsilon} dv_\epsilon = \frac{e}{m_\epsilon \bar{\nu}_{\epsilon g}} \\ D_\epsilon &= \frac{2}{3} \frac{1}{m_\epsilon \bar{\nu}_{\epsilon g}} \int_0^\infty 2\pi m_\epsilon v_\epsilon^4 \frac{f_\epsilon^0}{n_\epsilon} dv_\epsilon = \frac{2}{3} \frac{\bar{\epsilon}}{m_\epsilon \bar{\nu}_{\epsilon g}} \equiv \frac{e T_\epsilon}{m_\epsilon \bar{\nu}_{\epsilon g}}. \end{aligned}$$

Here, we have used the relations $\int_0^\infty 4\pi v_\epsilon^2 f_\epsilon^0 dv_\epsilon = n_\epsilon$ and $\int_0^\infty 2\pi m_\epsilon v_\epsilon^4 f_\epsilon^0 dv_\epsilon = 3/2 n_\epsilon e T_\epsilon$, as $\int_0^\infty v_\epsilon^2 f_\epsilon^{Max} \chi_{iso}^{(9M)} dv_\epsilon = \int_0^\infty v_\epsilon^4 f_\epsilon^{Max} \chi_{iso}^{(9M)} dv_\epsilon = 0$ where f_ϵ^{Max} is the Maxwellian distribution (cf. Balescu [20] Eq. (4.3.9)).

Similarly, one can retrieve the following relations for a Maxwellian EEDF ($\Delta_\epsilon = 0$):

$$\frac{D_\epsilon}{\mu_\epsilon} = \frac{D_{\epsilon_\epsilon}}{\mu_{\epsilon_\epsilon}} = T_\epsilon = \frac{2}{3} \bar{\epsilon} \quad \text{and} \quad \frac{D_\epsilon}{D_{\epsilon_\epsilon}} = \frac{\mu_\epsilon}{\mu_{\epsilon_\epsilon}} = \frac{5}{3}. \quad (30)$$

Here, we have defined D_{ϵ_ϵ} and μ_{ϵ_ϵ} as the energy diffusion and mobility, as defined in [14]. The relations of Eq. (30) are the Einstein relations of Maxwellian electrons, as described in classical books, e.g., [33]. These relations are consistent with the two-term Boltzmann equation. Note that, as the new transport effect in the particle and heat flux is proportional to $\nabla \Delta_\epsilon$, it also vanishes with a Maxwellian EEDF. In summary, the regularized moment model shows to be consistent with the classical theory for Maxwellian electrons and constant electron-gas collision frequency.

4.2. Constant electron-neutral cross section

We now consider the case where the electron-gas collision frequency is a hard sphere interaction, i.e., the momentum-transfer cross section is constant $Q_{\epsilon g}^{(1)} = \pi r_g^2$ where r_g is the radius of the neutral particle (we assume it to be constant and much larger than the electron). The collision frequency can be written as a function of the mean electron peculiar speed, $\nu_{\epsilon g}^{HS} = n_g Q_{\epsilon g}^{(1)} \bar{v}_{\epsilon th}$ with $\bar{v}_{\epsilon th} = \sqrt{8eT_\epsilon/\pi m_\epsilon}$. As a result, the collisional frequencies read,

$$\nu_{\epsilon g}^{(u,1)} = \frac{4}{3} \nu_{\epsilon g}^{HS}, \quad \nu_{\epsilon g}^{(q,1)} = \frac{4}{15} \nu_{\epsilon g}^{HS}, \quad \nu_{\epsilon g}^{(T,2)} = \left(4 + \frac{3}{2} \Delta_\epsilon\right) \nu_{\epsilon g}^{HS}, \quad \nu_{\epsilon g}^{(\Delta,2)} = 2 \nu_{\epsilon g}^{HS}, \quad (31)$$

$$\nu_{\epsilon g}^{(u,3)} = 4 \nu_{\epsilon g}^{HS}, \quad \nu_{\epsilon g}^{(q,3)} = \frac{12}{5} \nu_{\epsilon g}^{HS}, \quad \nu_{\epsilon g}^{(T,4)} = (48 + 90 \Delta_\epsilon) \nu_{\epsilon g}^{HS}, \quad \nu_{\epsilon g}^{(\Delta,4)} = 72 \nu_{\epsilon g}^{HS}. \quad (32)$$

The transport coefficients read,

$$D_\epsilon = \frac{13}{16} \frac{eT_\epsilon}{m_\epsilon \nu_{\epsilon g}^{HS}} \frac{1 - \frac{5}{13} \Delta_\epsilon + \frac{15}{26} \frac{\nu_{\epsilon \epsilon}^q}{\nu_{\epsilon g}^{HS}}}{1 + \frac{5}{8} \frac{\nu_{\epsilon \epsilon}^q}{\nu_{\epsilon g}^{HS}}}, \quad \chi_\epsilon = -\frac{5}{13} \frac{1 + \Delta_\epsilon}{1 - \frac{5}{13} \Delta_\epsilon + \frac{15}{26} \frac{\nu_{\epsilon \epsilon}^q}{\nu_{\epsilon g}^{HS}}},$$

$$\alpha_\epsilon = -\frac{5}{13} \frac{1}{1 - \frac{5}{13} \Delta_\epsilon + \frac{15}{26} \frac{\nu_{\epsilon \epsilon}^q}{\nu_{\epsilon g}^{HS}}}, \quad \mu_\epsilon = \frac{13}{16} \frac{e}{m_\epsilon \nu_{\epsilon g}^{HS}} \frac{1 + \frac{15}{26} \frac{\nu_{\epsilon \epsilon}^q}{\nu_{\epsilon g}^{HS}}}{1 + \frac{5}{8} \frac{\nu_{\epsilon \epsilon}^q}{\nu_{\epsilon g}^{HS}}},$$

$$\Lambda_\epsilon = -\frac{5}{13} eT_\epsilon \frac{1}{1 + \frac{15}{26} \frac{\nu_{\epsilon \epsilon}^q}{\nu_{\epsilon g}^{HS}}}, \quad \kappa_\epsilon = \frac{75}{52} \frac{p_\epsilon e}{m_\epsilon \nu_{\epsilon g}^{HS}} \frac{1 + 2\Delta_\epsilon}{1 + \frac{15}{26} \frac{\nu_{\epsilon \epsilon}^q}{\nu_{\epsilon g}^{HS}}}, \quad (33)$$

$$\vartheta_\epsilon = \frac{75}{52} \frac{(eT_\epsilon)^2}{m_\epsilon \nu_{\epsilon g}^{HS}} \frac{\Delta_\epsilon}{1 + \frac{15}{26} \frac{\nu_{\epsilon \epsilon}^q}{\nu_{\epsilon g}^{HS}}}, \quad \text{and} \quad \varkappa_\epsilon = \frac{75}{52} \frac{n_\epsilon (eT_\epsilon)^2}{m_\epsilon \nu_{\epsilon g}^{HS}} \frac{1}{1 + \frac{15}{26} \frac{\nu_{\epsilon \epsilon}^q}{\nu_{\epsilon g}^{HS}}}. \quad (34)$$

In the case of hard-sphere collisions, all the transport processes that were summarized in Table 1 are present. In this case, the transport coefficients depend on the temperature of electrons (as $\bar{v}_{\epsilon th}$ is a function of the temperature) and also depend on the EEDF (through the variable Δ_ϵ).

4.2.1. *Weakly-ionized plasma* As done above, we consider a case where the electron-gas elastic frequency is much larger than the electron-electron collision, i.e., $\nu_{\text{eg}}^{HS} \gg \nu_{\text{ee}}$. The set of moment equations for constant electron-neutral collision frequency reads

$$\frac{\partial n_{\text{e}}}{\partial t} - \nabla \cdot \left(\frac{13}{16} \frac{e}{m_{\text{e}} \nu_{\text{eg}}^{HS}} n_{\text{e}} \mathbf{E} + \frac{9}{8} \frac{1}{m_{\text{e}} \nu_{\text{eg}}^{HS}} \nabla p_{\text{e}} - \frac{1}{24} \frac{1}{e T_{\text{e}} \nu_{\text{eg}}^{HS}} \nabla p_{\text{e}}^{(4)} \right) = \dot{n}_{\text{e}}, \quad (35)$$

$$\begin{aligned} \frac{3}{2} \frac{\partial p_{\text{e}}}{\partial t} - \nabla \cdot \left(\frac{55}{32} \frac{e}{m_{\text{e}} \nu_{\text{eg}}^{HS}} p_{\text{e}} \mathbf{E} + \frac{15}{16} \frac{e T_{\text{e}}}{m_{\text{e}} \nu_{\text{eg}}^{HS}} \nabla p_{\text{e}} + \frac{5}{48 \nu_{\text{eg}}^{HS}} \nabla p_{\text{e}}^{(4)} \right) = \\ \frac{13}{16} \frac{e^2 n_{\text{e}}}{m_{\text{e}} \nu_{\text{eg}}^{HS}} E^2 + \left(\frac{9}{8} \frac{e}{m_{\text{e}} \nu_{\text{eg}}^{HS}} \nabla p_{\text{e}} - \frac{1}{24} \frac{1}{T_{\text{e}} \nu_{\text{eg}}^{HS}} \nabla p_{\text{e}}^{(4)} \right) \cdot \mathbf{E} \\ + 4 \frac{m_{\text{e}}}{m_{\text{g}}} n_{\text{e}} \nu_{\text{eg}}^{HS} e \left\{ \left(1 - \frac{1}{8} \Delta_{\text{e}} \right) T_{\text{g}} - \left(1 + \frac{3}{8} \Delta_{\text{e}} \right) T_{\text{e}} \right\} - \dot{Q}_{(\text{excit}, \text{iz})}, \end{aligned} \quad (36)$$

$$\begin{aligned} \frac{\partial p_{\text{e}}^{(4)}}{\partial t} - \nabla \cdot \left\{ \frac{e}{m_{\text{e}} \nu_{\text{eg}}^{HS}} \left(\frac{91}{48} p_{\text{e}}^{(4)} - \frac{35}{8} \frac{p_{\text{e}}^2}{\rho_{\text{e}}} \right) \mathbf{E} \right. \\ \left. + \frac{n_{\text{e}}}{e T_{\text{e}} \nu_{\text{eg}}^{HS}} \left(\frac{7}{72} p_{\text{e}}^{(4)} + \frac{35}{8} \frac{p_{\text{e}}^2}{\rho_{\text{e}}} \right) \nabla p_{\text{e}}^{(4)} + \frac{1}{m_{\text{e}} n_{\text{e}} \nu_{\text{eg}}^{HS}} \left(\frac{21}{8} p_{\text{e}}^{(4)} - \frac{105}{8} \frac{p_{\text{e}}^2}{\rho_{\text{e}}} \right) \nabla p_{\text{e}} \right\} = \\ \frac{55}{8} \frac{e^2 p_{\text{e}}}{m_{\text{e}}^2 \nu_{\text{eg}}^{HS}} E^2 + \frac{e}{m_{\text{e}} \nu_{\text{eg}}^{HS}} \left(\frac{5}{12} \nabla p_{\text{e}}^{(4)} + \frac{15}{4} \frac{p_{\text{e}}}{\rho_{\text{e}}} \nabla p_{\text{e}} \right) \cdot \mathbf{E} \\ + \frac{m_{\text{e}}}{m_{\text{g}}} \nu_{\text{eg}}^{HS} \frac{p_{\text{e}}^2}{\rho_{\text{e}}} \left((48 + 18 \Delta_{\text{e}}) \frac{T_{\text{g}}}{T_{\text{e}}} - (48 + 90 \Delta_{\text{e}}) \right) - \dot{Q}_{(\text{excit}, \text{iz})}^{(4)}. \end{aligned} \quad (37)$$

The system of transport equations is more complex than the case with constant collision frequency. We note that the diffusion terms couple the moments at different order. Similarly, in the energy dissipation due to the electric field, an additional term appears that is proportional to the gradient of the fourth moment. Finally, the effect of a non-Maxwellian EEDF is present in the electron-neutral collisions.

Concerning the transport coefficients in the fluxes of particles of Eqs. (26), one can retrieve the Einstein relation for a Maxwellian EEDF ($\Delta_{\text{e}} = 0$):

$$\frac{D_{\text{e}}}{\mu_{\text{e}}} = T_{\text{e}}. \quad (38)$$

Note that this relation is also consistent with the coefficients as obtained with the two-term Boltzmann approach, i.e., Eq. (29). In this case, the Einstein relation is not found for the energy diffusion due to the effect of the Soret effect, that in this case is not zero. Consequently, the Peltier effect is modified, as compared to the case with constant collision frequency.

5. Order of magnitude of the transport fluxes in an ICP discharge

We study the order of magnitude of the transport fluxes of Eqs. (16)-(17), and the transport effects of Table 1. We use the Langmuir probe measurements in the ICP discharge described in Refs. [34, 35]. In Fig. 1, we present the measurements at $p_{\text{g}} = 5$

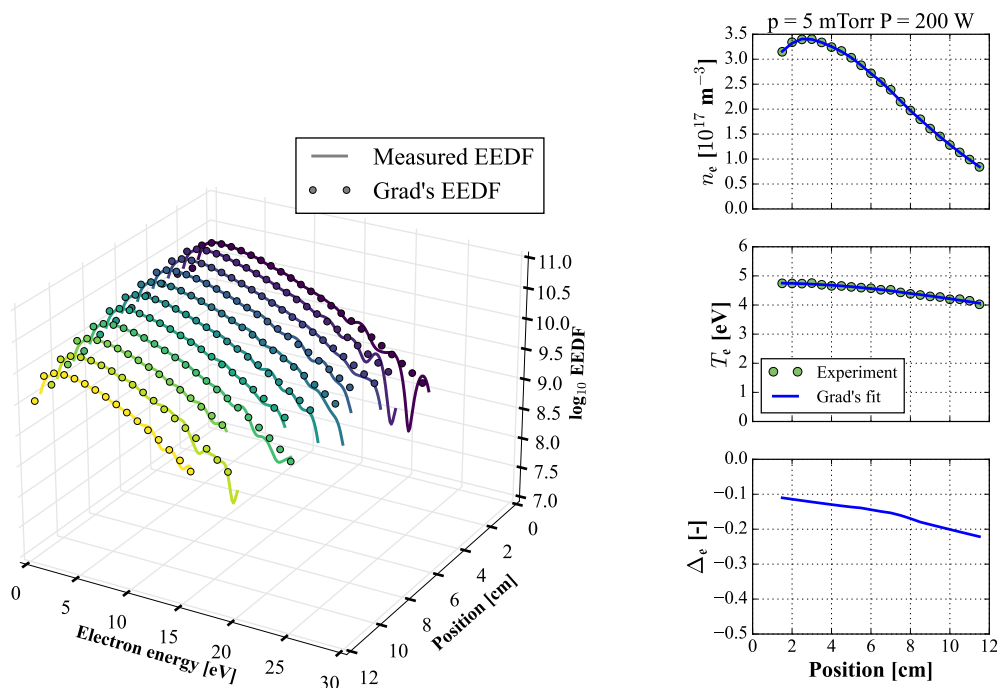


Figure 1. Left: Lagmuir probe measurements of the EEDF along the axis of an argon ICP discharge at 5 mTorr and 200 W, the antenna is placed at $x = 0$. Right: Electron density, temperature and deviation of the contracted fourth moment along the axis as fitted by a 9M Grad's distribution function.

mTorr and RF power 200 W. In the same figure, we present a fit of the EEDF that uses the Grad's 9M non-equilibrium distribution function. As the EEDF is not Maxwellian, the contracted fourth moment of the fitted Grad's distribution is $\Delta_e < 0$, which means that the tail is depopulated (as compared to a Maxwellian at the same temperature). The axial distribution of density, temperature and normalized contracted fourth moment is presented on the left panel of Fig. 1, where $x = 0$ is the powered antenna and $x = 13$ cm is the position of the exhaust grid. The density is localized close to the antenna, whereas the temperature and the fourth moment slightly decrease along the axis. As the RF field tends to thermalize the electron distribution function, the distribution function is less Maxwellian for increasing distances from the antenna. Similarly, one can note that the local field approximation would fail in representing these low-pressure conditions.

We compute the transport coefficients with the cross section from [36] in (Eqs. (A.10)-(A.16)) with the measured electron density and temperature and by considering a homogeneous argon gas background at 300 K. Similarly, the thermodynamic forces are computed by considering the gradients of the measured quantities. The velocity is assumed to be of the order of the local Bohm's velocity in order to estimate the Peltier effect. In Fig. 2, we show the contribution of the different terms of the transport velocity (left) and heat-flux (right). The transport velocity is dominated by Fick's diffusion as the density gradients are large under these pressure conditions. Note that this diffusion velocity is compensated by the electric field, which is

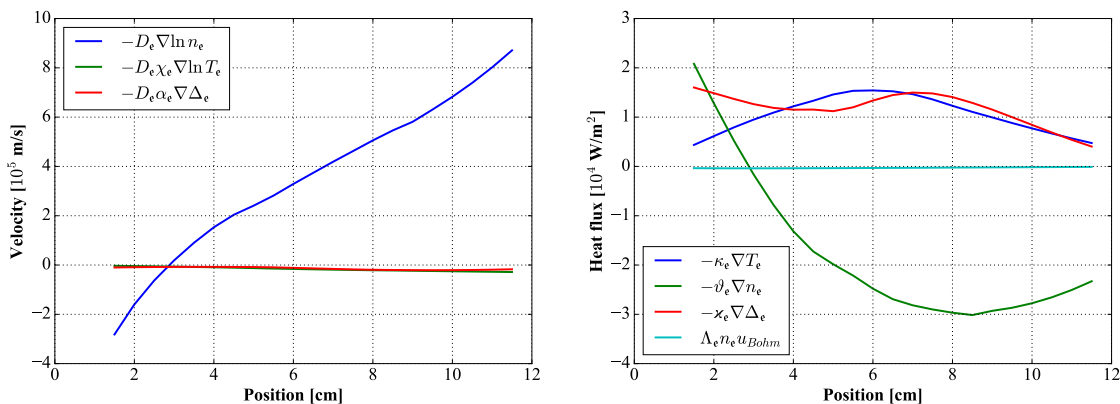


Figure 2. Order of magnitude of the different transport effects (velocity on the left and heat flux on the right) along the axis of an argon ICP discharge at 5 mTorr and 200 W.

not measured and hence not presented in the figure. The thermodiffusion and transport of particles due to EEDF spatial gradients are much smaller than the Fick's diffusion as the temperature and EEDF kurtosis gradients are small.

Alternatively, the heat-flux contributions of Fig. 2 show that the Dufour effect (heat flux produced by density gradients), Fourier's law (heat flux produced by temperature gradients) and the heat-flux produced by fourth-moment gradients are of the same order of magnitude. Note that Dufour's effect has a different sign than Fourier's law and that this effect does not appear for Maxwellian EEDFs. We highlight the magnitude of the novel transport effect that transports energy from regions with a more thermalized (Maxwellian) distribution function to regions with more non-equilibrium distribution function (with depopulated tails).

As discussed in Section 4, the particle and heat fluxes depend on the collision cross section and the distribution function. In particular, simplified local-field models that assume the Einstein relation for the energy coefficients, as shown in Eq. (30), are widely used in the literature although these relations only hold for Maxwellian EEDFs and constant collision frequency. However, as explained above these models provide a wrong representation of the heat-flux as $\vartheta_e = 0$ and hence Dufour's effect is not captured by the model. Similarly, the novel non-local transport effect $\varkappa_e \nabla \Delta_e$ is not captured with this assumption of Eq. (30). Similarly, models that assume a Maxwellian EEDF have the same problem in the representation of the heat-flux as both Dufour's effect nor $\varkappa_e \nabla \Delta_e$ are null. In addition, we would like to highlight that models that consider a constant momentum transfer cross section like BGK models (Bhatnagar-Gross-Krook) do not capture Soret and Peltier effects as well as the flux of particles due to gradients of the kurtosis. Finally, under these low-pressure conditions, the local field approximation is not justified. In Eq. (15), the local-field approximation assumes that the left-hand-side of the equation is negligible and the electric field terms balance the collisional terms. However, we can see that the gradients in the fluxes are not negligible and therefore

the EEDF is not local but a consequence of the both transport and the local collisional processes.

6. Conclusions

We have presented a discussion on the transport effects of the regularized 9M equations for non-equilibrium electrons in a partially-ionized plasma. This set of equations takes into account the effect of electrons out of equilibrium with a non-Maxwellian EEDF. The model shows that, besides the classical Fick's, Fourier's, and Ohm's laws, and Soret, Dufour and Peltier effects, additional transport effects result from spatial gradients in the kurtosis of the EEDF. This effects can be regarded as non-local transport phenomena, as they account for the spatial variations of the shape of the EEDF. In addition, the novel transport model takes into account the effect of the EEDF in the transport coefficients, in particular in the diffusion coefficient, heat conductivity, the thermodiffusion, and the Dufour's coefficient.

We have studied the transport model in two limiting cases: a constant collision frequency and a constant cross section. The constant collision frequency model is not able to capture Soret, Peltier and the transport of particles due to EEDF gradients. The diffusion and mobility coefficients are found to be consistent with the two-term Boltzmann theory. As a result, the model finds the classical Einstein relations for Maxwellian electrons. Alternatively, the constant cross section model captures all the mentioned transport processes as well as the Einstein relation for particle diffusion in Maxwellian electrons.

In this paper, by using the measured EEDFs in an ICP argon discharge, we have estimated the order of magnitude of the transport effects. The results show that, under these conditions, the transport of particles is dominated by Fick's diffusion. On the other hand, the spatial gradients in the kurtosis of the EEDF plays an important role in the heat-flux with a contribution that is of the same order as the Fourier's and Dufour's contributions. This transport processes are beyond the local-field approximation or the expansion in density gradients, both models widely used in the modeling of electrons in low-temperature plasmas.

The model is based on the regularized Grad's method and has the following assumptions and conditions of validity:

- The anisotropic part of the distribution function is assumed to be much smaller than the isotropic one. This assumption holds for a plasma where the collision between the electrons and the heavy species are dominant, i.e., $\text{Kn}_{\text{eg}} \ll 1$.
- The energy tensor is assumed to be isotropic (i.e., no tensorial polynomials are considered in the expansion). This assumption holds for electric fields that are not too large ($(eEt_c/m_e)^2 \ll v_{th}^2$), where t_c is the time between collisions.
- Grad's distribution is known to admit non-physical negative values, since the polynomial expansion may change its sign at high energies. The equations lose

their validity if the negative part of the tail is close to the energy of the threshold in inelastic collisional processes.

- The model considers the main collision mechanisms in an atomic gas in a weakly-ionized plasma, where the ionization dominates as compared to the recombination.

Note that the first two assumptions are the same of the two-term Boltzmann approach. These assumptions hold, for instance, in low-pressure ICP discharges of noble gases plasmas. Possible developments of the model to extend the region of validity are explained below.

Extensions of the model, using the same methodology, are possible. In particular, the model can be extended to study higher pressure conditions and more complex plasma mixtures. Under these conditions, superelastic collisions [37] and recombination and attachment [38] can modify the EEDF and need to be taken into account in the kinetic equation. Similarly, in more complex mixtures with molecular gases need more detailed models [39] for the concentration of excited species that influence the EEDF [40]. The Hermite basis is able to approximate any EEDF (with certain regularity conditions [41]), however, it converges better (with less number of moments) for distributions closer to Maxwellian. The number of Hermite polynomials needed to represent the EEDF in complex molecular mixtures will depend largely on the conditions. In particular, the ionization degree of the plasma (as the electron-electron collisions thermalize the distribution function), the effect of the electric field (the effect of the tails of the EEDF in the transport in partially-ionized plasmas depends on the E/N and the mean energies), and the spatial gradients (diffusion in the velocity space is usually coupled to the diffusion in the physical space).

Appendix A. Collisional frequencies and transport coefficients

Appendix A.1. Collision frequencies

The collisional frequencies, which are proportional to the heavy particle density, read

$$\nu_{\epsilon\alpha}^{(u,1)} = \frac{16}{3}n_\alpha\Omega_{\epsilon\alpha}^{(1,1)}, \quad \nu_{\epsilon\alpha}^{(g,1)} = \frac{16}{3}n_\alpha \left[\frac{2}{5}\Omega_{\epsilon\alpha}^{(1,2)} - \Omega_{\epsilon\alpha}^{(1,1)} \right], \quad (\text{A.1})$$

$$\nu_{\epsilon\alpha}^{(T,2)} = n_\alpha \left[(16 + 30\Delta_\epsilon)\Omega_{\epsilon\alpha}^{(1,1)} - 40\Delta_\epsilon\Omega_{\epsilon\alpha}^{(1,2)} + 8\Delta_\epsilon\Omega_{\epsilon\alpha}^{(1,3)} \right], \quad (\text{A.2})$$

$$\nu_{\epsilon\alpha}^{(u,3)} = \frac{16}{3}n_\alpha\Omega_{\epsilon\alpha}^{(1,2)}, \quad \nu_{\epsilon\alpha}^{(g,3)} = \frac{16}{3}n_\alpha \left[\frac{2}{5}\Omega_{\epsilon\alpha}^{(1,3)} - \Omega_{\epsilon\alpha}^{(1,2)} \right], \quad (\text{A.3})$$

$$\nu_{\epsilon\alpha}^{(T,4)} = n_\alpha \left[(64 + 120\Delta_\epsilon)\Omega_{\epsilon\alpha}^{(1,2)} - 160\Delta_\epsilon\Omega_{\epsilon\alpha}^{(1,3)} + 32\Delta_\epsilon\Omega_{\epsilon\alpha}^{(1,4)} \right] \quad (\text{A.4})$$

$$\nu_{\epsilon\alpha}^{(\Delta,2)} = 16n_\alpha \left[\Omega_{\epsilon\alpha}^{(1,2)} - \frac{5}{2}\Omega_{\epsilon\alpha}^{(1,1)} \right], \quad \nu_{\epsilon\alpha}^{(\Delta,4)} = 64n_\alpha \left(\Omega_{\epsilon\alpha}^{(1,3)} - \frac{5}{2}\Omega_{\epsilon\alpha}^{(1,2)} \right) \quad (\text{A.5})$$

where $\alpha \in \{\mathbf{i}, \mathbf{g}\}$ and

$$\Omega_{\epsilon\alpha}^{(l,r)}(T_\epsilon) = \frac{1}{2} \left(\frac{1}{\pi\beta_\epsilon} \right)^{1/2} \int_0^\infty \xi^{2r+3} e^{-\xi^2} Q_{\epsilon\alpha}^{(l)} d\xi \quad \text{with} \quad \xi = \sqrt{\beta_\epsilon}g \quad \text{and} \quad \beta_\epsilon = \frac{m_\epsilon}{2eT_\epsilon}. \quad (\text{A.6})$$

The electron–electron collision frequencies read,

$$\nu_{ee}^q = \frac{16}{15} n_e \Omega_{ee}^{(2,2)}, \quad \text{and} \quad \nu_{ee}^\Delta = 8 n_e \Omega_{ee}^{(2,2)}, \quad (\text{A.7})$$

where

$$\Omega_{ee}^{(2,2)}(T_e) = \frac{1}{2} \left(\frac{2}{\pi \beta_e} \right)^{1/2} \int_0^\infty \xi^7 e^{-\xi^2} Q_{ee}^{(2)} d\xi \quad \text{with} \quad \xi = \sqrt{\beta_e/2} g. \quad (\text{A.8})$$

Note that only the linear terms are considered in the computation of the electron–electron collisions. As the Coulomb viscosity cross section $Q_{ee}^{(2)} \propto v_e^{-4}$, decreases very fast with the energy, the second-order term $\chi_e \chi_{e_1}$ can be neglected as it only becomes important at high energies. Nevertheless, this small correction can be added in future extensions of the model.

The inelastic collision rates read,

$$K_{inel}^{(r)} = 4\pi \left(\frac{m_e}{2eT_e} \right)^r \int_0^\infty v_e^{2r+3} Q_{inel}^{(T)} f_e^{(0)} \left(1 + \chi_{isot}^{(9M)} \right) dv_e. \quad (\text{A.9})$$

Appendix A.2. Transport coefficients

The transport coefficients are written as a function of the frequencies and the normalized contracted fourth moment as follows

$$D_e = \frac{eT_e}{m_e} \frac{\nu_{eh}^{(q,3)} + \nu_{ee}^q - \frac{5}{2}(1 + \Delta_e)\nu_{eh}^{(q,1)}}{(\nu_{eh}^{(q,3)} + \nu_{ee}^q)\nu_{eh}^{(u,1)} - \nu_{eh}^{(q,1)}\nu_{eh}^{(u,3)}}, \quad (\text{A.10})$$

$$\chi_e = -\frac{5}{2} \frac{(1 + \Delta_e)\nu_{eh}^{(q,1)}}{\nu_{eh}^{(q,3)} + \nu_{ee}^q - \frac{5}{2}(1 + \Delta_e)\nu_{eh}^{(q,1)}}, \quad \alpha_e = \frac{\chi_e}{1 + \Delta_e}, \quad (\text{A.11})$$

$$\mu_e = \frac{e}{m_e} \frac{\nu_{eh}^{(q,3)} + \nu_{ee}^q - \frac{5}{2}\nu_{eh}^{(q,1)}}{(\nu_{eh}^{(q,3)} + \nu_{ee}^q)\nu_{eh}^{(u,1)} - \nu_{eh}^{(q,1)}\nu_{eh}^{(u,3)}} \quad (\text{A.12})$$

$$\Lambda_e = -eT_e \frac{\nu_{eh}^{(u,3)} - \frac{5}{2}\nu_{eh}^{(u,1)}}{\nu_{eh}^{(q,3)} + \nu_{ee}^q - \frac{5}{2}\nu_{eh}^{(q,1)}}, \quad (\text{A.13})$$

$$\kappa_e = \frac{5 p_e e}{2 m_e} \frac{1 + 2\Delta_e}{\nu_{eh}^{(q,3)} + \nu_{ee}^q - \frac{5}{2}\nu_{eh}^{(q,1)}} \quad (\text{A.14})$$

$$\vartheta_e = \frac{5 (eT_e)^2}{2 m_e} \frac{\Delta_e}{\nu_{eh}^{(q,3)} + \nu_{ee}^q - \frac{5}{2}\nu_{eh}^{(q,1)}}, \quad \text{and} \quad (\text{A.15})$$

$$\varkappa_e = \frac{5 n_e (eT_e)^2}{2 m_e} \frac{1}{\nu_{eh}^{(q,3)} + \nu_{ee}^q - \frac{5}{2}\nu_{eh}^{(q,1)}} \quad (\text{A.16})$$

where the electron heavy collision frequency is computed as the sum of the contributions of ions and gas, i.e., $\nu_{eh} = \nu_{ei} + \nu_{eg}$.

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