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Transport Theory of Charged Particle Swarms in Neutral Gases: Momentum Transfer Theory of Nonconservative Charged Particle Transport in Mixtures of Gases

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A wide variety of phenomena associated with reactive swarms in neutral gases is studied using an extension of Momentum Transfer Theory (MTT). We develop general MTT equations for swarms of particles in mixtures of gases including the effect of non - conservative (reactive) collisions. Hydrodynamics equations for average velocity and mean energy are derived for different degrees of approximation including hydrodynamic limit and small swarm to gas particle mass ratio. Empirical formulae, such as generalized Einstein relations and expression for the energy partitioning are derived, with modifications due to reactive effects made explicit. MTT is used to derive approximate expression for higher order transport coefficients of reacting particle swarms in mixtures of gases. Specific formulae were developed for the criteria for negative differential conductivity in mixtures of gases with elastic collisions only and for a single gas with reactions. We analyzed the application of MTT for the composition dependence of swarm mobilities in gas mixtures at arbitrary field strengths in situations when inelastic and reactive collisions are present. We extended the MTT to study non - conservative charged particle transport in crossed electric and magnetic fields.

I Introduction

Momentum transfer theory is based on considerations of momentum and energy balance, in which the momentum and energy gained by the swarm particle from the field must, for a steady state, be balanced by losses through collisions with neutral molecules. There is nothing approximate about the physics behind these ideas, but approximations enter when we try to calculate the collisional losses. MTT basically consists of applying Taylor expansion to the collision frequencies at the appropriately determined value of the mean energy. Because of its simplicity which however allows reasonable accuracy it has become quite popular in discussing the basic physical explanations of transport phenomena. It however is usually not adequate for very accurate transport calculations and cross section fitting.

MTT was first used to discuss the diffusion of neutral gases and the mobility of ions at very low field strengths where the energy can always be taken to be entirely thermal [1]. MTT has been initially developed for charged particle transport in mixtures of gases having only elastic processes [2]. The results for the corrections of Blanc's law [3, 4] and the relationship between diffusion coefficients and mobility [4] were obtained. Inelastic collisions have been included in the single gas MTT and the corresponding equations for energy, drift velocity and relationship between the mobility and components of the diffusion tensor were developed. MTT has been developed mainly by the efforts of R. E. Robson and his coworkers [5, 6] as an approximate solution to transport equations which gives an opportunity to develop analytic forms of various transport coefficients and their relations. Reactive collisions were included in addition to inelastic and the corresponding effects of attachment, annihilation [5] and ionization [6] on transport coefficients were discussed. MTT has also been applied in crossed electric $\vec{E}$ and magnetic $\vec{B}$ fields for a case of a single gas [7] but the reactive collisions were not considered.

Recently, we have generalized the MTT for a case of *reactive* particle swarms in *mixtures* of gases in the presence of crossed *electric* and *magnetic* fields [8, 9]. This paper should be viewed as a review of the complete theoretical procedure in its most general form and an illustration of the applicability of MTT. Our studies of the development of Negative Differential Conductivity (NDC) were already given its final presentation [9]. Some investigations, such as validity of Blanc's law at high E/n_0 [8], the higher order transport coefficients [10], electron transport in $\vec{E}\times\vec{B}$ fields [11] and time dependent MTT [8] were presented in preliminary form.

In this paper we shall discuss briefly all these applications only as an illustration of the usefulness of the procedure and the analytic forms that are derived. Detailed explanation of the particular physical phenomena will be left for the more focused technical papers.

In the development of the MTT we follow the procedure given by Robson [5] but we generalize it to different degree depending on particular applications. Even when identical analytical forms are obtained the development of the theory in the more general forms is necessary since the meaning of the terms entering the formula may be different and should be defined accurately.

II Theoretical evaluation

II.1 Time development of number density: Hydrodynamic regime

For the purposes of this paper a swarm is defined as an ensemble of *independent* charged particles moving in a neutral background gas. The motion of the particles is determined by the forces exerted by external electric and magnetic fields and collisions with the gas molecules, which may lead to reactions. The mutual interactions between the charges and the influence of the swarm on the neutral gas distribution can be neglected. Throughout this paper the electric $\vec{E}$ and magnetic $\vec{B}$ fields are assumed uniform in space. The phase - space distribution function $f(\vec{r},\vec{v},t)$ contains all the information about swarm behavior but is not directly measured. However, it is convenient to think that the charge density, defined by $n(\vec{r},t)=\int d^3\vec{r} f(\vec{r},\vec{v},t)$ is the measured quantity.

The hydrodynamic description is a phenomenological description of the time development of the number density $n(\vec{r},t)$. The description is applicable in a stationary state when the memory of the initial state $f(\vec{r},\vec{v},0)$ has been lost and the distribution function has become a functional of $n(\vec{r},t)$ as far as its space - time dependence is concerned. The starting point of the hydrodynamic description is the continuity equation for the number density,

$$\frac{\partial}{\partial t}n(\vec{r},t)+\frac{\partial}{\partial \vec{r}}\cdot[n(\vec{r},t)\vec{u}(\vec{r},t)]=\left[\frac{\partial n(\vec{r},t)}{\partial t}\right]_{\mathrm{coll}}. \tag{1}$$

It describes the change in $n(\vec{r},t)$ due to a convective particle current $n(\vec{r},t)\vec{u}(\vec{r},t)$ and a production term $[\partial n/\partial t]_{\mathrm{coll}}$. One now assumes that both of these quantities can be expressed as power series in the gradient operator $\partial/\partial\vec{r}$ with *constant* coefficients, and obtains the transport equation [12]:

$$\left[\frac{\partial}{\partial t} + \sum_{k=0}^{\infty} \hat{\omega}^{(k)} \odot (-\frac{\partial}{\partial \vec{r}})^k\right] n(\vec{r},t) = 0. \tag{2}$$

This equation is often called the Generalized Diffusion Equation (GDE). The constants $\hat{\omega}^{(k)}$ are tensorial transport coefficients of the order k, and $\odot$ indicates a k-fold scalar product. The $\hat{\omega}^{(k)}$ can be taken to be symmetrical under permutation of space indices. By truncating Eq. (2) at $k = 3$, we obtain the familiar diffusion equation

$$\left[\frac{\partial}{\partial t} + \vec{W}^{\mathrm{mix}} \cdot \frac{\partial}{\partial \vec{r}} - \hat{D}^{\mathrm{mix}} : \left\{\frac{\partial}{\partial \vec{r}}, \frac{\partial}{\partial \vec{r}}\right\} + \hat{S}^{\mathrm{mix}} \odot \left(\frac{\partial}{\partial \vec{r}}\right)^3\right] n(\vec{r},t) = -\varrho^* n(\vec{r},t), \tag{3}$$

where we identify $\varrho^* = -\omega^{(0)}$ as the reaction rate, $\vec{W}^{\mathrm{mix}} = (\omega_i^{(1)})_{i=1}^3$ as the drift velocity, $\hat{D}^{\mathrm{mix}} = (\omega_{ij}^{(2)})_{i,j=1,1}^{3,3}$ as the diffusion tensor and $\hat{S}^{\mathrm{mix}} = (\omega_{ijk}^{(3)})_{i,j,k=1,1,1}^{3,3,3}$ as the third-order transport coefficients or skewness. Equation (3) is a universal diffusion equation applicable to *all* experiments that allow the hydrodynamic description. The quantities $\vec{W}$, $\hat{D}$, $\hat{S}$ and ϱ^* are universal properties of the particle swarm - neutral gas combination, independent of the experimental arrangement which is used to measure them.

II.2 Formulation of the moment equations

Consider a swarm of particles of charge e and mass m moving with velocity $\vec{v}$ through neutral gas mixtures under the influence of an applied electric $\vec{E}$ and magnetic $\vec{B}$ fields. The term "swarm" has a connotation of electron and ion swarms, but the analysis can be applied to fast neutral particles or positrons. Suppose that there are several (l) different species of neutral gases present. Let m_α and $\vec{v}_\alpha$ be mass and velocity of molecules of the αth neutral gas, respectively. Let $n(\vec{r},t)$ be the number density of swarm particles and let $n_\alpha(\vec{r},t)$ be the number density of the αth neutral gas. We introduce a standard notation: $n_0 = \sum_\alpha n_\alpha$ (number density of the gas mixture), $\mu_\alpha = mm_\alpha/(m+m_\alpha)$ (reduced mass), $M_\alpha = m_\alpha/(m+m_\alpha)$, $M_\alpha^0 = m/(m+m_\alpha)$, $\vec{v}_{r\alpha} = \vec{v} - \vec{v}_\alpha$ (relative velocity) and $\varepsilon_\alpha = \frac{1}{2}\mu_\alpha v_{r\alpha}^2$ (energy measured in the center-of-mass reference frame).

Let $f^{\mathrm{mix}}(\vec{r},\vec{v},t)$ and $f_\alpha^{\mathrm{mix}}(\vec{r},\vec{v},t)$ be the swarm and αth neutral gas one-particle velocity distribution function in the multicomponent mixture, respectively. If only one of the neutral species (say α) is present, $f^\alpha(\vec{r},\vec{v},t)$ and $f_\alpha(\vec{r},\vec{v},t)$ denote the corresponding velocity distribution functions. By convention, all velocity distribution functions are normalized to number densities.

Averaging operators used in the development of equations are defined as:

$$\langle\Phi(\vec{v},\vec{v}_\alpha)\rangle^{\mathrm{mix}} = \frac{1}{n(\vec{r},t)}\int d^3\vec{v} f^{\mathrm{mix}}(\vec{r},\vec{v},t)\Phi(\vec{v},\vec{v}_\alpha), \tag{4}$$

$$\langle\Phi(\vec{v},\vec{v}_\alpha)\rangle_\alpha^{\mathrm{mix}} = \frac{1}{n_\alpha(\vec{r},t)}\int d^3\vec{v} f_\alpha^{\mathrm{mix}}(\vec{r},\vec{v}_\alpha,t)\Phi(\vec{v},\vec{v}_\alpha), \tag{5}$$

$$\langle\langle\Phi(\vec{v},\vec{v}_\alpha)\rangle\rangle_\alpha^{\mathrm{mix}} = \frac{1}{n(\vec{r},t)n_\alpha(\vec{r},t)}\int\int d^3\vec{v}d^3\vec{v}_\alpha f^{\mathrm{mix}}(\vec{r},\vec{v},t) f_\alpha^{\mathrm{mix}}(\vec{r},\vec{v}_\alpha,t)\Phi(\vec{v},\vec{v}_\alpha), \tag{6}$$

$$\langle\Phi(\vec{v},\vec{v}_\alpha)\rangle^{\alpha} = \frac{1}{n(\vec{r},t)}\int d^3\vec{v} f^{\alpha}(\vec{r},\vec{v},t)\Phi(\vec{v},\vec{v}_\alpha), \tag{7}$$

$$\langle\Phi(\vec{v},\vec{v}_\alpha)\rangle_{\alpha} = \frac{1}{n_0(\vec{r},t)}\int d^3\vec{v}_\alpha f_{\alpha}(\vec{r},\vec{v}_\alpha,t)\Phi(\vec{v},\vec{v}_\alpha), \tag{8}$$

$$\langle\langle \Phi(\vec{v},\vec{v}_\alpha)\rangle\rangle_\alpha = \frac{1}{n(\vec{r},t)n_0(\vec{r},t)}\int\int d^3\vec{v}d^3\vec{v}_\alpha f^\alpha(\vec{r},\vec{v},t)f_\alpha(\vec{r},\vec{v}_\alpha,t)\Phi(\vec{v},\vec{v}_\alpha), \tag{9}$$

where $\Phi(\vec{v},\vec{v}_\alpha)$ is any function of $\vec{v}$ and $\vec{v}_\alpha$. For the sake of brevity, we write $\varepsilon^0_\alpha = \langle\langle\varepsilon_\alpha\rangle\rangle^{\rm mix}_\alpha$, $\varepsilon'_\alpha = \langle\langle\varepsilon_\alpha\rangle\rangle_\alpha$, $\delta\vec{v}^{\rm mix} = \vec{v} - \langle\vec{v}\rangle^{\rm mix}$, $\delta\vec{v}^\alpha = \vec{v} - \langle\vec{v}\rangle^\alpha$, $\varepsilon^0 = \{\varepsilon^0_\alpha \mid \alpha \in I_l\}$ and $\varepsilon' = \{\varepsilon'_\alpha \mid \alpha \in I_l\}$, where $I_l = 1,2,...,l$ denotes an appropriate finite set of indices.

The Boltzmann equation for a swarm of particles moving through a gaseous multicomponent medium is [12]:

$$\left[\frac{\partial}{\partial t} + \vec{v}\cdot\frac{\partial}{\partial\vec{r}} + \frac{e}{m}\left[\vec{E}(\vec{r},t) + \vec{v}\times\vec{B}(\vec{r},t)\right]\cdot\frac{\partial}{\partial\vec{v}}\right] f^{\rm mix}(\vec{r},\vec{v},t) = \sum_\alpha J^{\rm mix}_\alpha(f^{\rm mix}, f^{\rm mix}_\alpha), \tag{10}$$

where $J^{\rm mix}_\alpha$ is the collision operator which represents the rate of change of the distribution function $f^{\rm mix}$ due to collisions between the swarm particles and molecules of the neutral gas α. In the collision terms of Boltzmann equation, only binary collisions are considered.

The chain of moment equations is derived by multiplying both sides of Eq. (10) by various powers of swarm particle velocity $\vec{v}$ and integrating over velocity space. The first moment equation is obtained by multiplying Boltzmann equation by unity and integrating, giving the continuity equation:

$$\frac{\partial}{\partial t}n(\vec{r},t) + \frac{\partial}{\partial\vec{r}}\cdot\left[n(\vec{r},t)\langle\vec{v}\rangle^{\rm mix}\right] = \sum_{\alpha\in I_l}\int d^3\vec{v}J^{\rm mix}_\alpha(f^{\rm mix}, f^{\rm mix}_\alpha). \tag{11}$$

The second moment equation is obtained by multiplying the Boltzmann equation by $m\vec{v}$ and integrating, giving the following momentum balance equation:

$$\begin{aligned}&\frac{\partial}{\partial t}\left[mn(\vec{r},t)\langle\vec{v}\rangle^{\rm mix}\right] + \frac{\partial}{\partial\vec{r}}\left[mn(\vec{r},t)\langle\{\vec{v},\vec{v}\}\rangle^{\rm mix}\right] - n(\vec{r},t)e\left[\vec{E}(\vec{r},t) + \langle\vec{v}\rangle^{\rm mix}\times\vec{B}(\vec{r},t)\right]\\ &= \sum_{\alpha\in I_l}\int d^3\vec{v}m\vec{v}J^{\rm mix}_\alpha(f^{\rm mix}, f^{\rm mix}_\alpha).\end{aligned} \tag{12}$$

The third moment equation is obtained by multiplying Boltzmann equation by $\frac{1}{2}mv^2$ and integrating, giving the following energy balance equation:

$$\begin{aligned}&\frac{\partial}{\partial t}\left[\frac{1}{2}mn(\vec{r},t)\left\langle v^2\right\rangle^{\rm mix}\right] + \frac{\partial}{\partial\vec{r}}\cdot\left[\frac{1}{2}mn(\vec{r},t)\left\langle v^2\vec{v}\right\rangle^{\rm mix}\right] - n(\vec{r},t)e\vec{E}(\vec{r},t)\left\langle\vec{v}\right\rangle^{\rm mix}\\ &= \sum_{\alpha\in I_l}\int d^3\vec{v}\frac{1}{2}mv^2J^{\rm mix}_\alpha(f^{\rm mix}, f^{\rm mix}_\alpha).\end{aligned} \tag{13}$$

The terms on the right-hand side of the moment equations (11), (12) and (13) are equal to the average rate at which swarm particles lose their number density, momentum or energy per unit volume through collisions with gas molecules. In these expressions, tensor $\{\cdot,\cdot\}$ is the dyadic product of two vectors.

II.2.A Collisional processes

To give a definite expression to the right-hand side terms of the moment equation we make several assumptions about collisional processes between the swarm particles and gas molecules. The processes which we investigate are limited to elastic, inelastic and reactive (which include attachment and ionization) collisions of individual swarm particles with normal gas molecules. We characterize all these possible processes by respective collision frequencies. Frequency $\nu_\alpha(\vec{r},v_{r\alpha},t)$ of collisions between the swarm particles and molecules of species α is related to the cross section $\sigma_\alpha(v_{r\alpha})$ characterizing the process by $\nu_\alpha(\vec{r},v_{r\alpha},t) = n_\alpha(\vec{r},t)v_{r\alpha}\sigma_\alpha(v_{r\alpha})$, where $\vec{v}_{r\alpha}$ is the relative velocity and $n_\alpha(\vec{r},t)$ is the number density of the αth neutral gas. We neglect the collisions of swarm particles with other swarm particles, ions and excited molecules.

We take into account the momentum and energy loss of the swarm particles in elastic collisions arising from the finite mas of gas molecules. In the calculation of this momentum and energy loss we assume that the actual thermal energy of the molecules (each neutral gas has the same nonzero temperature T^{mix}) is not negligible compared to the mean energy of the swarm particles. The momentum transfer collision frequency for collisions between the swarm particles and molecules of species α is denoted by $\nu_\alpha^{(m)}(\vec{r}, v_{r\alpha}, t)$.

Let $I_\alpha^{(\mathrm{in})}$ be a set of indices which enumerates possible inelastic collisions of a swarm particle with molecules of the gas α, while $\nu_{s\alpha}(\vec{r}, v_{r\alpha}, t)$, $s \in I_\alpha^{(\mathrm{in})}$ is the corresponding collision frequency. The threshold for these inelastic collisions is denoted by $\Delta E_\alpha^s, s \in I_\alpha^{(\mathrm{in})}, \alpha \in I_l$. Momentum exchange in inelastic processes is not ignored. The total momentum transfer collision frequency is given by

$$\nu_{0\alpha}^{(m)}(\vec{r}, v_{r\alpha}, t) = \nu_\alpha^{(m)}(\vec{r}, v_{r\alpha}, t) + \sum_{s \in I_\alpha^{(\mathrm{in})}} \nu_{s\alpha}^{(m)}(\vec{r}, v_{r\alpha}, t), \alpha \in I_l, \tag{14}$$

where $\nu_{s\alpha}^{(m)}(\vec{r}, v_{r\alpha}, t), s \in I_\alpha^{(\mathrm{in})}$ denotes momentum transfer collision frequency of collision inducing inelastic process $s \in I_\alpha^{(\mathrm{in})}$.

Reactive collisions for electron swarms include creation (ionization by electron impact) or loss (electron attachment to electronegative gas molecules or positron annihilation) of a swarm particle. Let $\nu_{s\alpha}^{(A)}(\vec{r}, v_{r\alpha}, t)$ be the rate at which swarm particles are lost in collisions through attachment channel $s \in I_\alpha^{(A)}$. In the case of electron ionization the incident electron collides with a molecule and two electrons emerge after the collision, one being the scattered incident electron and the other being the ejected electron; it is a three body problem. In approximation of a mass ratio of $m/m_\alpha \ll 1$, we can ignore the motion of the molecule, so that the available kinetic energy and the momentum after the ionization are divided between the two electrons. We consider only single ionization with ionization energy $\varepsilon_\alpha^{(I)}$, but the resulting ion can be left in any one of its internal excited states characterized by excitation energy $\Delta\varepsilon_{s\alpha}^{(I)}, s \in I_\alpha^{(I)}$. Let $\nu_{s\alpha}^{(I)}(\vec{r}, v_{r\alpha}, t)$ be the ionization frequency for the sth ionization channel, $s \in I_\alpha^{(I)}$. The total rates for attachment (A) and ionization (I) are defined by formulas:

$$\nu_\alpha^{(A)}(\vec{r}, v_{r\alpha}, t) = \sum_{s \in I_\alpha^{(A)}} \nu_{s\alpha}^{(A)}(\vec{r}, v_{r\alpha}, t), \quad \nu_\alpha^{(I)}(\vec{r}, v_{r\alpha}, t) = \sum_{s \in I_\alpha^{(I)}} \nu_{s\alpha}^{(I)}(\vec{r}, v_{r\alpha}, t), \quad \alpha \in I_l. \tag{15}$$

II.2.B Moment equations

We extend the moment equations derived by Robson [5] and Robson and Ness [6] to include gas mixtures by taking appropriately weighted sums of collision terms in momentum equations. Using arguments similar to Robson's we find:
(a) Equation of continuity:

$$\begin{aligned} &\frac{\partial}{\partial t} n(\vec{r}, t) + \frac{\partial}{\partial \vec{r}} \cdot \left[n(\vec{r}, t) \langle \vec{v} \rangle^{\mathrm{mix}} \right] \\ &= -n(\vec{r}, t) \sum_{\alpha \in I_l} \langle\langle \nu_\alpha^{(A)}(\vec{r}, v_{r\alpha}, t) \rangle\rangle_\alpha^{\mathrm{mix}} + n(\vec{r}, t) \sum_{\alpha \in I_l} \langle\langle \nu_\alpha^{(I)}(\vec{r}, v_{r\alpha}, t) \rangle\rangle_\alpha^{\mathrm{mix}}; \end{aligned} \tag{16}$$

(b) Momentum balance equation:

$$\begin{aligned} &\frac{\partial}{\partial t} \left[m n(\vec{r}, t) \langle \vec{v} \rangle^{\mathrm{mix}} \right] + \frac{\partial}{\partial \vec{r}} \left[m n(\vec{r}, t) \langle \{\vec{v}, \vec{v}\} \rangle^{\mathrm{mix}} \right] - n(\vec{r}, t) e \left[\vec{E}(\vec{r}, t) + \langle \vec{v} \rangle^{\mathrm{mix}} \times \vec{B}(\vec{r}, t) \right] \\ &= -n(\vec{r}, t) \sum_{\alpha \in I_l} \mu_\alpha \langle\langle \vec{v}_{r\alpha} \nu_{0\alpha}^{(m)}(\vec{r}, v_{r\alpha}, t) \rangle\rangle_\alpha^{\mathrm{mix}} - n(\vec{r}, t) m \sum_{\alpha \in I_l} \langle\langle \vec{v} \nu_\alpha^{(A)}(\vec{r}, v_{r\alpha}, t) \rangle\rangle_\alpha^{\mathrm{mix}}; \end{aligned} \tag{17}$$

(c) Energy balance equation:

$$\begin{aligned}
&\frac{\partial}{\partial t}\left[\frac{1}{2}mn(\vec{r},t)\left\langle v^2\right\rangle^{\mathrm{mix}}\right]+\frac{\partial}{\partial\vec{r}}\cdot\left[\frac{1}{2}mn(\vec{r},t)\left\langle v^2\vec{v}\right\rangle^{\mathrm{mix}}\right]-n(\vec{r},t)e\vec{E}(\vec{r},t)\left\langle\vec{v}\right\rangle^{\mathrm{mix}}\\
&=-n(\vec{r},t)\sum_{\alpha\in I_l}\frac{\mu_\alpha}{m+m_\alpha}\left\langle\left\langle\left[mv^2-m_\alpha v_\alpha^2-(m-m_\alpha)\vec{v}\cdot\vec{v}_\alpha\right]\nu_{0\alpha}^{(m)}(\vec{r},v_{r\alpha},t)\right\rangle\right\rangle_\alpha^{\mathrm{mix}}\\
&-n(\vec{r},t)\sum_{\alpha\in I_l}\sum_{s\in I_\alpha^{(\mathrm{in})}}\langle\langle\nu_{s\alpha}(\vec{r},v_{r\alpha},t)\rangle\rangle_\alpha^{\mathrm{mix}}\,\Delta E_\alpha^s-n(\vec{r},t)\frac{1}{2}m\sum_{\alpha\in I_l}\left\langle\left\langle v^2\nu_\alpha^{(A)}(\vec{r},v_{r\alpha},t)\right\rangle\right\rangle_\alpha^{\mathrm{mix}}\\
&-n(\vec{r},t)\sum_{\alpha\in I_l}\left\langle\left\langle\nu_\alpha^{(I)}(\vec{r},v_{r\alpha},t)\right\rangle\right\rangle_\alpha^{\mathrm{mix}}\varepsilon_\alpha^{(I)}-n(\vec{r},t)\sum_{\alpha\in I_l}\sum_{s\in I_\alpha^{(I)}}\left\langle\left\langle\nu_{s\alpha}^{(I)}(\vec{r},v_{r\alpha},t)\right\rangle\right\rangle_\alpha^{\mathrm{mix}}\Delta\varepsilon_{s\alpha}^{(I)}.
\end{aligned}\tag{18}$$

Note that the magnetic field does not *explicitly* modify the energy balance.

II.3 Momentum transfer approximation

Now let us replace the variables $v_{r\alpha}\to\varepsilon_\alpha=\frac{1}{2}\mu_\alpha v_{r\alpha}^2$, $\alpha\in I_l$ in expressions for collisional frequencies, $\nu_\alpha=\nu_\alpha(\vec{r},v_{r\alpha},t)\to\tilde{\nu}_\alpha=\tilde{\nu}_\alpha(\vec{r},\varepsilon_\alpha,t)$, $\alpha\in I_l$. When we expand $\tilde{\nu}_\alpha(\vec{r},\varepsilon_\alpha,t)$ in Taylor series in the vicinity of ε_α^0 we obtain

$$\tilde{\nu}_\alpha(\vec{r},\varepsilon_\alpha^0,t)=\tilde{\nu}_\alpha(\vec{r},\varepsilon_\alpha^0,t)+\left(\frac{d\tilde{\nu}_\alpha(\vec{r},\varepsilon_\alpha,t)}{d\varepsilon_\alpha}\right)_{\varepsilon_\alpha=\varepsilon_\alpha^0}(\varepsilon_\alpha-\varepsilon_\alpha^0)+o(\varepsilon_\alpha-\varepsilon_\alpha^0)\tag{19}$$

where the remainder consists of terms of higher - order derivatives of $\tilde{\nu}_\alpha(\vec{r},\varepsilon_\alpha,t)$. We assume that Taylor expansion (19) converges rapidly in the neighborhood of ε_α^0. The extended momentum - transfer approximation consists of retention of several terms in Taylor expansion (19). Momentum and energy loss rates for attachment are calculated using the first two terms on the right - hand side of Eq. (19) and the assumption that the distribution function of swarm particles is shifted Maxwellian. We also assume that the background gas is in the thermal equilibrium (characterized by Maxwellian distribution and temperature T^{mix}). Using momentum - transfer approximation [5, 6, 9] we find from Eqs. (1), (17) and (18):

(a) Equation of continuity:

$$\frac{\partial}{\partial t}n(\vec{r},t)+\frac{\partial}{\partial\vec{r}}\cdot\left[n(\vec{r},t)\langle\vec{v}\rangle^{\mathrm{mix}}\right]=-n(\vec{r},t)\tilde{\nu}^{(A)}(\vec{r},\varepsilon^0,t)+n(\vec{r},t)\tilde{\nu}^{(I)}(\vec{r},\varepsilon^0,t);\tag{20}$$

(b) Momentum balance equation:

$$\begin{aligned}
&\frac{\partial}{\partial t}\left[mn(\vec{r},t)\langle\vec{v}\rangle^{\mathrm{mix}}\right]+\frac{\partial}{\partial\vec{r}}\left[mn(\vec{r},t)\langle\{\vec{v},\vec{v}\}\rangle^{\mathrm{mix}}\right]-n(\vec{r},t)e\left[\vec{E}(\vec{r},t)+\langle\vec{v}\rangle^{\mathrm{mix}}\times\vec{B}(\vec{r},t)\right]\\
&=-n(\vec{r},t)m\left\langle\vec{v}\right\rangle^{\mathrm{mix}}\tilde{\nu}_0^{(m)}(\vec{r},\varepsilon^0,t)-n(\vec{r},t)m\left\langle\vec{v}\right\rangle^{\mathrm{mix}}\left[\tilde{\nu}^{(A)}(\vec{r},\varepsilon^0,t)+\xi^{\mathrm{mix}}\tilde{\nu}_1^{(A)}(\vec{r},\varepsilon^0,t)\right];
\end{aligned}\tag{21}$$

(c) Energy balance equation:

$$\begin{aligned}
&\frac{\partial}{\partial t}\left[\frac{1}{2}mn(\vec{r},t)\left\langle v^2\right\rangle^{\mathrm{mix}}\right]+\frac{\partial}{\partial\vec{r}}\cdot\left[\frac{1}{2}mn(\vec{r},t)\left\langle v^2\vec{v}\right\rangle^{\mathrm{mix}}\right]-n(\vec{r},t)e\vec{E}(\vec{r},t)\left\langle\vec{v}\right\rangle^{\mathrm{mix}}\\
&=-n(\vec{r},t)\left[m\left\langle v^2\right\rangle^{\mathrm{mix}}-3kT^{\mathrm{mix}}\right]\tilde{\nu}_{00}^{(m)}(\vec{r},\varepsilon^0,t)-n(\vec{r},t)\sum_{\alpha\in I_l}\sum_{s\in I_\alpha^{(\mathrm{in})}}\tilde{\nu}_{s\alpha}(\vec{r},\varepsilon_\alpha^0,t)\Delta E_\alpha^s\\
&-n(\vec{r},t)\frac{1}{2}m\left\langle v^2\right\rangle^{\mathrm{mix}}\tilde{\nu}^{(A)}(\vec{r},\varepsilon^0,t)-n(\vec{r},t)\frac{1}{2}m\left[\left\langle v^2\right\rangle^{\mathrm{mix}}-(\langle\vec{v}\rangle^{\mathrm{mix}})^2\right]\xi^{\mathrm{mix}}\tilde{\nu}_1^{(A)}(\vec{r},\varepsilon^0,t)\\
&-n(\vec{r},t)\sum_{\alpha\in I_l}\tilde{\nu}_\alpha^{(I)}(\vec{r},\varepsilon_\alpha^0,t)\varepsilon_\alpha^{(I)}-n(\vec{r},t)\sum_{\alpha\in I_l}\sum_{s\in I_\alpha^{(I)}}\tilde{\nu}_{s\alpha}^{(I)}(\vec{r},\varepsilon_\alpha^0,t)\Delta\varepsilon_{s\alpha}^{(I)};
\end{aligned}\tag{22}$$

where we introduced the notation

$$\tilde{\nu}^{(A)}(\vec{r},\varepsilon^0,t) = \sum_{\alpha\in I_l} \tilde{\nu}_\alpha^{(A)}(\vec{r},\varepsilon_\alpha^0,t), \quad \tilde{\nu}^{(I)}(\vec{r},\varepsilon^0,t) = \sum_{\alpha\in I_l} \tilde{\nu}_\alpha^{(I)}(\vec{r},\varepsilon_\alpha^0,t), \tag{23}$$

$$\tilde{\nu}_0^{(m)}(\vec{r},\varepsilon^0,t) = \sum_{\alpha\in I_l} M_\alpha \tilde{\nu}_{0\alpha}^{(m)}(\vec{r},\varepsilon_\alpha^0,t), \quad \tilde{\nu}_{00}^{(m)}(\vec{r},\varepsilon^0,t) = \sum_{\alpha\in I_l} M_\alpha M_\alpha^0 \tilde{\nu}_{0\alpha}^{(m)}(\vec{r},\varepsilon_\alpha^0,t), \tag{24}$$

$$\tilde{\nu}_1^{(A)}(\vec{r},\varepsilon^0,t) = \sum_{\alpha\in I_l} M_\alpha \tilde{\nu}_{\alpha 1}^{(A)}(\vec{r},\varepsilon_\alpha^0,t), \quad \tilde{\nu}_{\alpha 1}^{(A)}(\vec{r},\varepsilon_\alpha^0,t) = \left(\frac{d\tilde{\nu}_\alpha^{(A)}(\vec{r},\varepsilon_\alpha,t)}{d\varepsilon_\alpha}\right)_{\varepsilon_\alpha=\varepsilon_\alpha^0}, \quad \alpha\in I_l, \tag{25}$$

$$\xi^{\mathrm{mix}} = \frac{2}{3}\frac{1}{2}m\left[\left\langle v^2\right\rangle^{\mathrm{mix}} - \left(\langle\vec{v}\rangle^{\mathrm{mix}}\right)^2\right]. \tag{26}$$

If we take the product of $m\langle\vec{v}\rangle^{\mathrm{mix}}$ and $\frac{1}{2}m\langle v^2\rangle^{\mathrm{mix}}$ with equation of continuity (Eq. (20)) and subtract from Eqs. (21) and (22) there result a more useful form of the momentum and energy balance equations [5, 6, 9]:

$$\begin{aligned} &mn(\vec{r},t)\left[\frac{\partial}{\partial t} + \langle\vec{v}\rangle^{\mathrm{mix}}\cdot\frac{\partial}{\partial\vec{r}}\right]\langle\vec{v}\rangle^{\mathrm{mix}} - n(\vec{r},t)e\left[\vec{E}(\vec{r},t) + \langle\vec{v}\rangle^{\mathrm{mix}}\times\vec{B}(\vec{r},t)\right] + \operatorname{div}\hat{P}^{\mathrm{mix}} \\ &= -n(\vec{r},t)m\langle\vec{v}\rangle^{\mathrm{mix}}\tilde{\nu}_0^{(m)}(\vec{r},\varepsilon^0,t) - n(\vec{r},t)m\langle\vec{v}\rangle^{\mathrm{mix}}\left[\xi^{\mathrm{mix}}\tilde{\nu}_1^{(A)}(\vec{r},\varepsilon^0,t) + \tilde{\nu}^{(I)}(\vec{r},\varepsilon^0,t)\right] \end{aligned} \tag{27}$$

$$\begin{aligned} &n(\vec{r},t)\left[\frac{\partial}{\partial t} + \langle\vec{v}\rangle^{\mathrm{mix}}\cdot\frac{\partial}{\partial\vec{r}}\right]\left[\frac{1}{2}m\left\langle v^2\right\rangle^{\mathrm{mix}}\right] + \frac{\partial}{\partial\vec{r}}\cdot\left[\hat{P}^{\mathrm{mix}}\langle\vec{v}\rangle^{\mathrm{mix}} + \frac{3}{2}\vec{Q}^{\mathrm{mix}}\right] \\ &- n(\vec{r},t)e\vec{E}(\vec{r},t)\langle\vec{v}\rangle^{\mathrm{mix}} = -n(\vec{r},t)\left[m\left\langle v^2\right\rangle^{\mathrm{mix}} - 3kT^{\mathrm{mix}}\right]\tilde{\nu}_{00}^{(m)}(\vec{r},\varepsilon^0,t) \\ &- n(\vec{r},t)\sum_{\alpha\in I_l}\sum_{s\in I_\alpha^{(\mathrm{in})}}\tilde{\nu}_{s\alpha}(\vec{r},\varepsilon_\alpha^0,t)\Delta E_\alpha^s - n(\vec{r},t)\frac{1}{2}m\left[\left\langle v^2\right\rangle^{\mathrm{mix}} - (\langle\vec{v}\rangle^{\mathrm{mix}})^2\right]\xi^{\mathrm{mix}}\tilde{\nu}_1^{(A)}(\vec{r},\varepsilon^0,t) \\ &- n(\vec{r},t)\sum_{\alpha\in I_l}\tilde{\nu}_\alpha^{(I)}(\vec{r},\varepsilon_\alpha^0,t)\varepsilon_\alpha^{(I)} - n(\vec{r},t)\sum_{\alpha\in I_l}\sum_{s\in I_\alpha^{(I)}}\tilde{\nu}_{s\alpha}^{(I)}(\vec{r},\varepsilon_\alpha^0,t)\Delta\varepsilon_{s\alpha}^{(I)} \\ &- n(\vec{r},t)\frac{1}{2}m\left\langle v^2\right\rangle^{\mathrm{mix}}\tilde{\nu}^{(I)}(\vec{r},\varepsilon^0,t). \end{aligned} \tag{28}$$

In Eqs. (27) and (28) the pressure tensor $\hat{P}^{\mathrm{mix}}$ and vector heat conductivity $\vec{Q}^{\mathrm{mix}}$ are defined as follows

$$\hat{P}^{\mathrm{mix}} = mn(\vec{r},t)\left\langle\left\{\delta\vec{v}^{\mathrm{mix}},\delta\vec{v}^{\mathrm{mix}}\right\}\right\rangle^{\mathrm{mix}}, \quad \vec{Q}^{\mathrm{mix}} = \frac{1}{3}mn(\vec{r},t)\left\langle\delta\vec{v}^{\mathrm{mix}}\left(\delta\vec{v}^{\mathrm{mix}}\right)^2\right\rangle^{\mathrm{mix}}. \tag{29}$$

If we take the product of $\langle\vec{v}\rangle^{\mathrm{mix}}$ with momentum balance equation (27) and subtract from the energy balance equation (28), it can be written in an equivalent form [5, 6, 9]:

$$\begin{aligned} &n(\vec{r},t)\frac{1}{2}m\left[\frac{\partial}{\partial t} + \langle\vec{v}\rangle^{\mathrm{mix}}\cdot\frac{\partial}{\partial\vec{r}}\right]\left(\left\langle v^2\right\rangle^{\mathrm{mix}} - \left(\langle\vec{v}\rangle^{\mathrm{mix}}\right)^2\right) + \left(\hat{P}^{\mathrm{mix}}\frac{\partial}{\partial\vec{r}}\right)\cdot\langle\vec{v}\rangle^{\mathrm{mix}} + \frac{\partial}{\partial\vec{r}}\cdot\left(\frac{3}{2}\vec{Q}^{\mathrm{mix}}\right) \\ &= -n(\vec{r},t)\left\{\left[m\left\langle v^2\right\rangle^{\mathrm{mix}} - 3kT^{\mathrm{mix}}\right]\tilde{\nu}_{00}^{(m)}(\vec{r},\varepsilon^0,t) - m\left(\langle\vec{v}\rangle^{\mathrm{mix}}\right)^2\tilde{\nu}_0^{(m)}(\vec{r},\varepsilon^0,t)\right\} \\ &- n(\vec{r},t)\sum_{\alpha\in I_l}\sum_{s\in I_\alpha^{(\mathrm{in})}}\tilde{\nu}_{s\alpha}(\vec{r},\varepsilon_\alpha^0,t)\Delta E_\alpha^s - n(\vec{r},t)\sum_{\alpha\in I_l}\tilde{\nu}_\alpha^{(I)}(\vec{r},\varepsilon_\alpha^0,t)\varepsilon_\alpha^{(I)} \\ &- n(\vec{r},t)\sum_{\alpha\in I_l}\sum_{s\in I_\alpha^{(I)}}\tilde{\nu}_{s\alpha}^{(I)}(\vec{r},\varepsilon_\alpha^0,t)\Delta\varepsilon_{s\alpha}^{(I)} - n(\vec{r},t)\frac{1}{2}m\left\langle v^2\right\rangle^{\mathrm{mix}}\left[\xi^{\mathrm{mix}}\tilde{\nu}_1^{(A)}(\vec{r},\varepsilon^0,t) + \tilde{\nu}^{(I)}(\vec{r},\varepsilon^0,t)\right] \\ &+ n(\vec{r},t)\frac{1}{2}m\left(\langle\vec{v}\rangle^{\mathrm{mix}}\right)^2\left[\xi^{\mathrm{mix}}\tilde{\nu}_1^{(A)}(\vec{r},\varepsilon^0,t) + 2\tilde{\nu}^{(I)}(\vec{r},\varepsilon^0,t)\right]. \end{aligned} \tag{30}$$

It is to be emphasized that these equations have not been approximated apart from the MTA representation of the collision terms. In what follows, we specialize to a variety of circumstances by making assumptions about spatial and/or time derivatives, as appropriate to the physical situation.

II.4 Hydrodynamic limit and transport coefficients

We assume that the stage of evolution of the swarm is the hydrodynamic regime. The basic hydrodynamic assumption is that the number density satisfies equation (2) and the space - time dependence of the phase - space distribution has the form

$$f(\vec{r},\vec{v},t) = \sum_{k=0}^{\infty} \hat{f}^{(k)}(\vec{v}) \odot \left(-\frac{\partial}{\partial \vec{r}}\right)^k n(\vec{r},t). \tag{31}$$

The functions $\hat{f}^{(k)}(\vec{v})$ are tensors of rank k and $\odot$ indicates k-fold scalar product. The space - time dependence of $f(\vec{r},\vec{v},t)$ is thus functionally determined by $n(\vec{r},t)$. The HDR is that stage of evolution of the swarm when temporal and/or spatial properties are controlled entirely by the number density $n(\vec{r},t)$ and for any *intensive* property ψ [5]:

$$\frac{\partial}{\partial t}\psi = 0, \quad \frac{\partial}{\partial \vec{r}}\psi = 0, \quad \frac{d}{dt}\psi = 0. \tag{32}$$

Considered in this limit, the momentum and energy balance equations (Eqs. (27),(28)) become

$$-e\left[\vec{E} + \langle\vec{v}\rangle^{\mathrm{mix}} \times \vec{B}\right] + k\hat{T}^{\mathrm{mix}}\vec{G}(\vec{r},t) = -m\,\langle\vec{v}\rangle^{\mathrm{mix}}\,\bar{\nu}^{\mathrm{mix}}_{(m)}, \tag{33}$$

$$\begin{aligned}
&\langle\vec{v}\rangle^{\mathrm{mix}}\, k\hat{T}^{\mathrm{mix}}\vec{G}(\vec{r},t) + \frac{3}{2}\vec{q}^{\mathrm{mix}}\vec{G}(\vec{r},t) - e\vec{E}\,\langle\vec{v}\rangle^{\mathrm{mix}}\\
&= -\left[m\left\langle v^2\right\rangle^{\mathrm{mix}} - 3kT^{\mathrm{mix}}\right]\tilde{\nu}^{(m)}_{00} - \sum_{\alpha\in I_l}\sum_{s\in I^{(\mathrm{in})}_\alpha}\tilde{\nu}_{s\alpha}\Delta E^s_\alpha - \frac{1}{2}m\left[\left\langle v^2\right\rangle^{\mathrm{mix}} + (\langle\vec{v}\rangle^{\mathrm{mix}})^2\right]\xi^{\mathrm{mix}}\tilde{\nu}^{(A)}_1\\
&- \sum_{\alpha\in I_l}\tilde{\nu}^{(I)}_\alpha\varepsilon^{(I)}_\alpha - \sum_{\alpha\in I_l}\sum_{s\in I^{(I)}_\alpha}\tilde{\nu}^{(I)}_{s\alpha}\Delta\varepsilon^{(I)}_{s\alpha} - \frac{1}{2}m\left\langle v^2\right\rangle^{\mathrm{mix}}\tilde{\nu}^{(I)}
\end{aligned} \tag{34}$$

where $\hat{T}^{\mathrm{mix}}$ is the temperature tensor defined by

$$k\hat{T}^{\mathrm{mix}} = \frac{1}{n(\vec{r},t)}\hat{P}^{\mathrm{mix}}, \tag{35}$$

while

$$\vec{q}^{\mathrm{mix}} = \frac{1}{n(\vec{r},t)}\vec{Q}^{\mathrm{mix}} \quad \vec{G}(\vec{r},t) = \frac{1}{n(\vec{r},t)}\frac{\partial}{\partial\vec{r}}n(\vec{r},t) \tag{36}$$

and

$$\bar{\nu}^{\mathrm{mix}}_{(m)} = \tilde{\nu}^{(m)}_0 + \xi^{\mathrm{mix}}\tilde{\nu}^{(A)}_1 + \tilde{\nu}^{(I)}. \tag{37}$$

Equation (30) in HDL reduced to

$$\begin{aligned}
\frac{3}{2}\vec{q}^{\mathrm{mix}}\vec{G}(\vec{r},t) &= -\left[m\left\langle v^2\right\rangle^{\mathrm{mix}} - 3kT^{\mathrm{mix}}\right]\tilde{\nu}^{(m)}_{00} + m\left(\langle\vec{v}\rangle^{\mathrm{mix}}\right)^2\tilde{\nu}^{(m)}_0 - \sum_{\alpha\in I_l}\sum_{s\in I^{(\mathrm{in})}_\alpha}\tilde{\nu}_{s\alpha}\Delta E^s_\alpha\\
&- \sum_{\alpha\in I_l}\tilde{\nu}^{(I)}_\alpha\varepsilon^{(I)}_\alpha - \sum_{\alpha\in I_l}\sum_{s\in I^{(I)}_\alpha}\tilde{\nu}^{(I)}_{s\alpha}\Delta\varepsilon^{(I)}_{s\alpha} - \frac{1}{2}m\left\langle v^2\right\rangle^{\mathrm{mix}}\left[\xi^{\mathrm{mix}}\tilde{\nu}^{(A)}_1 + \tilde{\nu}^{(I)}\right]\\
&+ \frac{1}{2}m\left(\langle\vec{v}\rangle^{\mathrm{mix}}\right)^2\left[\xi^{\mathrm{mix}}\tilde{\nu}^{(A)}_1 + 2\tilde{\nu}^{(I)}\right].
\end{aligned} \tag{38}$$

We can rewrite the Eq. (38) in terms of the average energies in the center of mass frame

$$\langle\langle\varepsilon_\alpha\rangle\rangle_\alpha^{\text{mix}} = M_\alpha \frac{1}{2} m \left\langle v^2 \right\rangle^{\text{mix}} + M_\alpha^0 \frac{3}{2} k T^{\text{mix}}, \quad \alpha \in I_l. \tag{39}$$

When we substitute Eq. (39) in Eq. (38), we obtain the set of equations for *energy partitioning* in reactive gas mixture [5, 6, 9]:

$$\begin{aligned}\langle\langle\varepsilon_\alpha\rangle\rangle_\alpha^{\text{mix}} &= \frac{1}{2} m \left(\langle\vec{v}\rangle^{\text{mix}}\right)^2 M_\alpha r^{\text{mix}} + \frac{3}{2} k T^{\text{mix}} M_\alpha \left[\frac{M_\alpha^0}{M_\alpha} + s^{\text{mix}}\right] - M_\alpha \bar{\Omega}^{\text{mix}} \\ &- M_\alpha \frac{3}{2} \vec{q}^{\text{mix}} \vec{G}(\vec{r}, t) \frac{1}{\bar{\nu}_{(e)}^{\text{mix}}}, \quad \alpha \in I_l.\end{aligned} \tag{40}$$

We use the notations:

$$\bar{\nu}_{(e)}^{\text{mix}} = 2\tilde{\nu}_{00}^{(m)} + \xi^{\text{mix}} \tilde{\nu}_1^{(A)} + \tilde{\nu}^{(I)}. \tag{41}$$

$$r^{\text{mix}} = \frac{2\tilde{\nu}_0^{(m)} + \xi^{\text{mix}} \tilde{\nu}_1^{(A)} + 2\tilde{\nu}^{(I)}}{\bar{\nu}_{(e)}^{\text{mix}}}, \quad s^{\text{mix}} = \frac{2\tilde{\nu}_{00}^{(m)}}{\bar{\nu}_{(e)}^{\text{mix}}}, \tag{42}$$

$$\begin{aligned}\bar{\Omega}^{\text{mix}} &= \frac{\bar{\Lambda}^{\text{mix}}}{\bar{\nu}_{(e)}^{\text{mix}}}, \quad \bar{\Lambda}^{\text{mix}} = \sum_{\alpha \in I_l} \bar{\Lambda}_\alpha^{\text{mix}}, \\ \bar{\Lambda}_\alpha^{\text{mix}} &= \sum_{s \in I_\alpha^{(\text{in})}} \tilde{\nu}_{s\alpha} \Delta E_\alpha^s + \tilde{\nu}_\alpha^{(I)} \varepsilon_\alpha^{(I)} + \sum_{s \in I_\alpha^{(I)}} \tilde{\nu}_{s\alpha}^{(I)} \Delta\varepsilon_{s\alpha}^{(I)}, \quad \alpha \in I_l.\end{aligned} \tag{43}$$

II.4.A Transport coefficients

An analytic treatment of swarms in the presence of crossed electric and magnetic fields presented here is given for the coordinate system $(\vec{e}_x, \vec{e}_y, \vec{e}_z)$ defined by

$$\vec{e}_x || \vec{E}_{||} := \vec{E}\{\vec{b}, \vec{b}\}; \quad \vec{e}_y || \vec{E}_\perp := \vec{E}(\hat{I} - \{\vec{b}, \vec{b}\}); \quad \vec{e}_z || \vec{H} := \vec{b} \times \vec{E}; \tag{44}$$

where $\hat{I}$ is the unit tensor and $\vec{b} = \vec{B}/|\vec{B}|$. Following a similar procedure to Robson's [5], equations (33) and (40) can be written as

$$\langle\vec{v}\rangle^{\text{mix}} = \hat{K}(\varepsilon^0, (\langle\vec{v}\rangle^{\text{mix}})^2, \vec{B}) \left[\vec{E} - \frac{k}{e} \hat{T}^{\text{mix}} \vec{G}\right], \tag{45}$$

$$\langle\langle\varepsilon_\alpha\rangle\rangle_\alpha^{\text{mix}} = \tilde{L}^\alpha(\varepsilon^0, (\langle\vec{v}\rangle^{\text{mix}})^2, \vec{B}) - M_\alpha \frac{3}{2} \vec{q}^{\text{mix}} \vec{G} \frac{1}{\bar{\nu}_{(e)}^{\text{mix}}}, \quad \alpha \in I_l. \tag{46}$$

The tensor $\hat{K}$ (mobility tensor) has the following structure

$$\hat{K} = \frac{1}{\left(\frac{m}{e}\bar{\nu}_{(m)}^{\text{mix}}\right)^2 + B^2} \begin{bmatrix} \frac{\left(\frac{m}{e}\bar{\nu}_{(m)}^{\text{mix}}\right)^2 + B^2}{\frac{m}{e}\bar{\nu}_{(m)}^{\text{mix}}} & 0 & 0 \\ 0 & \frac{m}{e}\bar{\nu}_{(m)}^{\text{mix}} & +B \\ 0 & -B & \frac{m}{e}\bar{\nu}_{(m)}^{\text{mix}} \end{bmatrix} \tag{47}$$

for the previously described system of coordinates and

$$\begin{aligned}\tilde{L}^\alpha(\varepsilon^0, (\langle\vec{v}\rangle^{\text{mix}})^2, \vec{B}) =& \frac{1}{2} m (\langle\vec{v}\rangle^{\text{mix}})^2 M_\alpha r^{\text{mix}} \\ &+ \frac{3}{2} k T^{\text{mix}} M_\alpha \left[\frac{M_\alpha^0}{M_\alpha} + s^{\text{mix}}\right] - M_\alpha \bar{\Omega}^{\text{mix}}, \quad \alpha \in I_l.\end{aligned} \tag{48}$$

If the heat flux on the right - hand side of Eq. (46) is neglected, then both $\langle\vec{v}\rangle^{\text{mix}}$ and $\langle\langle\varepsilon_\alpha\rangle\rangle_\alpha^{\text{mix}}$, $\alpha \in I_l$ depend upon $\vec{E}$ and $\vec{G}$ through the combination $\vec{E} - \frac{k}{e}\hat{T}^{\text{mix}}\vec{G}$. If the functions $\vec{\omega}^{\text{mix}} = \vec{\omega}^{\text{mix}}(\vec{E}, \vec{B})$ and $\varepsilon_\alpha^{\text{mix}} = \varepsilon_\alpha^{\text{mix}}(\vec{E}, \vec{B})$, $\alpha \in I_l$ satisfy the following system of equations:

$$\vec{\omega}^{\text{mix}}(\vec{E}, \vec{B}) = \hat{K}\left(\left(\varepsilon_\alpha^{\text{mix}}\right)_{\alpha\in I_l}, \left(\omega^{\text{mix}}\right)^2, \vec{B}\right)\vec{E}, \tag{49}$$

$$\varepsilon_\alpha^{\text{mix}}(\vec{E}, \vec{B}) = \tilde{L}^\alpha\left(\left(\varepsilon_\alpha^{\text{mix}}\right)_{\alpha\in I_l}, \left(\omega^{\text{mix}}\right)^2, \vec{B}\right), \quad \alpha \in I_l, \tag{50}$$

we can write

$$\langle\vec{v}\rangle^{\text{mix}} = \vec{\omega}^{\text{mix}}\left[\vec{E} - \frac{k}{e}\hat{T}^{\text{mix}}\vec{G}, \vec{B}\right], \tag{51}$$

$$\langle\langle\varepsilon_\alpha\rangle\rangle_\alpha^{\text{mix}} = \varepsilon_\alpha^{\text{mix}}\left[\vec{E} - \frac{k}{e}\hat{T}^{\text{mix}}\vec{G}, \vec{B}\right], \quad \alpha \in I_l. \tag{52}$$

Function $\vec{\omega}^{\text{mix}}$ and $\varepsilon_\alpha^{\text{mix}}, \alpha \in I_l$ are, respectively, average velocity and energy in spatially uniform conditions and are found by solving the system of nonlinear equations (Eqs. (49) and (50)) for the given values of the electric and magnetic fields. The reaction rate $\tilde{\nu}^* = \tilde{\nu}^{(A)} - \tilde{\nu}^{(I)}$ is also a function of $\vec{E} - (k/e)\hat{T}^{\text{mix}}\vec{G}$ and $\vec{B}$:

$$\tilde{\nu}^* = \tilde{\nu}^*\left[\varepsilon^0\left(\vec{E} - \frac{k}{e}\hat{T}^{\text{mix}}\vec{G}, \vec{B}\right)\right] = \alpha^*\left(\vec{E} - \frac{k}{e}\hat{T}^{\text{mix}}\vec{G}, \vec{B}\right). \tag{53}$$

The calculation of transport coefficients of swarm particles in gas mixtures proceeds in a similar manner as in Robson [5]. In the hydrodynamics regime $|\vec{G}|$ is small. We expand functions $\vec{\omega}^{\text{mix}}$ (Eq. (51)) and α^* (Eq. (53)) to the *first* and *second* order in $\vec{G}$, respectively. Substituting these expansions into the equation of continuity ((20)) leads after some algebra to the diffusion equation ((2)), where

$$\vec{W}^{\text{mix}}(\vec{E}, \vec{B}) = \vec{\omega}^{\text{mix}}(\vec{E}, \vec{B}) - \frac{k}{e}\hat{T}^{\text{mix}}\frac{\partial\alpha^*(\vec{E}, \vec{B})}{\partial\vec{E}}, \tag{54}$$

is the drift velocity and,

$$D_{ij}^{\text{mix}} = \frac{k}{e}\sum_{k=1}^{3} t_{ik}^{\text{mix}}\left[\frac{\partial}{\partial E_k}W_j^{\text{mix}} + \frac{1}{2}\frac{k}{e}\sum_{l=1}^{3} t_{jl}^{\text{mix}}\frac{\partial}{\partial E_l}\frac{\partial\alpha^*}{\partial E_k}\right], \ i, j = 1, 2, 3 \tag{55}$$

is the diffusion tensor. Net average reaction rate is evaluated from

$$\varrho^*(\vec{E}, \vec{B}) = \alpha^*(\vec{E}, \vec{B}) = \tilde{\nu}^*[\varepsilon^0(\vec{E}, \vec{B})] = \tilde{\nu}^{(A)}[\varepsilon^0(\vec{E}, \vec{B})] - \tilde{\nu}^{(I)}[\varepsilon^0(\vec{E}, \vec{B})]. \tag{56}$$

Evaluation of Eqs. (54), (55) and (56) are carried out in Appendix (A). Temperature tensor $\hat{T}^{\text{mix}} = [t_{ij}^{\text{mix}}]_{i,j=1,1}^{3,3}$ must be, strictly speaking, evaluated from the higher order moment equations [13]. Equation (55) shows that the generalized Einstein relation is satisfied for reacting swarm particles if the reaction rate $\tilde{\nu}^*$ is independent of energy.

The procedure for finding transport coefficients for reacting swarm particles is as follows [5, 6]:

(a) Find $l+3$ functions ω_q^{mix}, $q = x, y, z$, $\varepsilon_\alpha^{\text{mix}}$, $\alpha \in I_l$ by solving the system of nonlinear equations (Eqs. (49) and (50)) for the given values of the electric and magnetic fields.

(b) Find the reaction rate α^* from Eq. (56).

(c) Find the temperature tensor $\hat{T}^{\text{mix}}$ from the higher-order moment equations.

(d) Find the drift velocity $\vec{W}^{\text{mix}}$ and diffusion tensor $\hat{D}^{\text{mix}}$ from Eqs. (54) and (55).

II.5 Higher order transport coefficients

Momentum-transfer theory will also be used to derive approximate expression for higher order transport coefficients of reacting particle swarms in mixtures of gases. Higher order transport coefficients can be useful in obtaining the cross section data and removing the discrepancies between the existing sets [14]. Our basic aim is to derive *relationships* between experimentally measurable quantities. In particular, the aim is to obtain semi-quantitative relations between skewness and lower-order transport coefficients.

We expand functions $\vec{\omega}^{\text{mix}}$ (51) and α^* (53) to the *second* and *third* order in $\vec{G}$, respectively. Substituting these expansions into the equation of continuity (20) leads to the diffusion equation (3), where

$$S_{ijk}^{\text{mix}} = \frac{1}{2}\frac{k^2}{e^2}\sum_{l=1}^{3} t_{il}^{\text{mix}} \sum_{m=1}^{3} t_{jm}^{\text{mix}} \left[\frac{\partial}{\partial E_m}\frac{\partial}{\partial E_l} W_k^{\text{mix}} + \frac{1}{6}\frac{k}{e}\sum_{n=1}^{3} t_{kn}^{\text{mix}} \frac{\partial}{\partial E_n}\frac{\partial}{\partial E_m}\frac{\partial \alpha^*}{\partial E_l}\right], \quad i,j,k = 1,2,3 \tag{57}$$

is the skewness. Evaluation of the Eq. (57) is carried out in Appendix (A). Equation (57) establishes the relationship between skewness $\hat{S}^{\text{mix}}$, drift velocity $\vec{W}^{\text{mix}}$ and reaction rate α^*. This equation has the same role that GER play for diffusion. From measurements of drift velocity $\vec{W}^{\text{mix}}$ and reaction rates α^* as a function of the reduced electric field E/n_0, it is possible to predict diffusion coefficients and skewness, as long as the temperature tensor $\hat{T}^{\text{mix}}$ can be estimated.

Following a similar procedure to Appendix (A), a relation for transport coefficients of the order $k > 3$ can be obtained. This consideration lead to the conclusion that k-th order transport coefficients $(k \geq 2)$ depend upon $(k-1)$-th derivate of drift velocity $\vec{W}^{\text{mix}}$ and k-th derivate of reaction rate α^* with respect to the electric field.

In the derivation of the Eqs. (51) and (52)), it was noted that we can neglected the heat flux in the energy balance equation. Since this assumption is decreasingly valid as the swarm particles have larger mass, one may expect that Eqs. (55) and (57) is not correct in cases where mass of swarm particle is comparable with the mass of gas atoms. Using MTT, Robson [15] have established that Eq. (55) is inadequate for the description of ion transport. He showed that an additional factor involving heat flux must appear in the GER.

Here we assume that $\vec{B} = 0$ and no reactive collisions. Symmetry considerations lead to the conclusion that the 27 - component tensor $\hat{S}^{\text{mix}}$ has only *three* independent components [16]. This can also be deduced directly from Eq. (55). If $\vec{E}$ is aligned with z-axis of a system of coordinate temperature tensor $\hat{T}^{\text{mix}} \to [t_{ij}^{\text{mix}}]_{i,j=1,1}^{3,3}$ has the diagonal structure:

$$t_{ij}^{\text{mix}} = [T_{\perp}^{\text{mix}}(\delta_{i1}+\delta_{i2}) + T_{||}^{\text{mix}}\delta_{i3}]\delta_{ij}, \quad i,j = 1,2,3. \tag{58}$$

If we write $\vec{W}^{\text{mix}} = K^{\text{mix}}\vec{E}$, where K^{mix} is the mobility coefficient, then

$$\begin{aligned} \frac{\partial}{\partial E_k} W_j^{\text{mix}} &= \delta_{kj} K^{\text{mix}} + (K^{\text{mix}})' \frac{E_k E_j}{E} \\ \frac{\partial}{\partial E_l}\frac{\partial}{\partial E_k} W_j^{\text{mix}} &= \delta_{kj}(K^{\text{mix}})'\frac{E_l}{E} + (K^{\text{mix}})''\frac{E_l E_k E_j}{E^2} + (K^{\text{mix}})'\frac{(\delta_{kl}E_j + \delta_{lj}E_k)E^2 - E_k E_j E_l}{E^3}. \end{aligned} \tag{59}$$

Then, assuming the diagonal form for the temperature tensor (58), we find that Eq. (57) and (59) yields the skewness tensor $\hat{S}^{\text{mix}} \to [S_{ijk}^{\text{mix}}]_{i,j,k=1,1,1}^{3,3,3}$:

$$[S_{ijk}^{\text{mix}}]_{i,j=1,1}^{3,3} = \begin{bmatrix} 0 & 0 & \frac{1}{2}\frac{k^2}{e^2}T_{\perp}T_{||}K' \\ 0 & 0 & 0 \\ \frac{1}{2}\frac{k^2}{e^2}T_{\perp}T_{||}K' & 0 & 0 \end{bmatrix} \quad k = 1; \tag{60}$$

$$[S_{ijk}^{\text{mix}}]_{i,j=1,1}^{3,3} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & \frac{1}{2}\frac{k^2}{e^2}T_{\perp}T_{||}K' \\ 0 & \frac{1}{2}\frac{k^2}{e^2}T_{\perp}T_{||}K' & 0 \end{bmatrix} \quad k = 2; \tag{61}$$

$$[S^{\rm mix}_{ijk}]^{3,3}_{i,j=1,1} = \begin{bmatrix} \frac{1}{2}\frac{k^2}{e^2}T_\perp^2 K' & 0 & 0 \\ 0 & \frac{1}{2}\frac{k^2}{e^2}T_\perp^2 K' & 0 \\ 0 & 0 & \frac{1}{2}\frac{k^2}{e^2}T_\parallel^2[2K' + EK''] \end{bmatrix} \quad k = 3. \tag{62}$$

Equations (60),(61) and (62) show that $\hat{S}^{\rm mix}$ has only *three* independent components depend on transverse $T_\perp^{\rm mix}$ and/or longitudinal $T_\parallel^{\rm mix}$ components of temperature tensor and the first $(K^{\rm mix})'$ and/or the second $(K^{\rm mix})''$ derivate of the mobility.

II.5.A Calculation for rare gases

In this section we will present calculation of the third-order transport coefficients for the swarm of electrons in the rare gases He, Ne and Ar. We deal with E/n_0's for which the mean electron energies are well below the first inelastic threshold. In that case, distribution of electron velocities is nearly isotropic. Hence,

$$t_{ij}^{\rm mix} = T\delta_{ij} \approx \frac{2}{3}\frac{1}{k}\varepsilon^{\rm mix}\delta_{ij}, \;\; i,j = 1,2,3, \tag{63}$$

where $\varepsilon^{\rm mix} \approx (1/2)m\langle v^2\rangle^{\rm mix}$. Using the above assumptions, we can simplify Eq. (57),

$$S_{ijk}^{\rm mix} = \frac{1}{2}\frac{k^2}{e^2}T^2\frac{\partial}{\partial E_j}\frac{\partial}{\partial E_i}W_k^{\rm mix} + \frac{1}{12}\frac{k^3}{e^3}T^3\frac{\partial}{\partial E_k}\frac{\partial}{\partial E_j}\frac{\partial\alpha^*}{\partial E_i}, \;\; i,j,k = 1,2,3. \tag{64}$$

If electric field $\vec{E}$ is aligned with the z-axis of a system of coordinate ($E = E_z$, $W_z^{\rm mix} = -W^{\rm mix}$), then it is clear that longitudinal component of skewness is given by

$$S_L^{\rm mix} \equiv S_{zzz}^{\rm mix} = -\frac{1}{2}\frac{k^2}{e^2}T^2\frac{\partial^2 W^{\rm mix}}{\partial E^2} + \frac{1}{12}\frac{k^3}{e^3}T^3\frac{\partial^3\alpha^*}{\partial E^3}. \tag{65}$$

If we assume no reactive collisions or that reaction rate α^* is independent of energy, the second term on the right-hand side of Eq. (65) is equal to zero. Finally, combining Eqs. (65) and (63) yields

$$S_L^{\rm mix} = -\frac{2}{9}\frac{\varepsilon^2}{e^2}\frac{\partial^2 W^{\rm mix}}{\partial E^2}. \tag{66}$$

In the first, most straightforward application of the results in this section we take a light swarm particles in a cold gas, neglect inelastic and reactive processes and assume constant elastic cross section, $\sigma^{(el)} = \sigma^0/(4\pi) = \text{const}$ (hard-sphere model). Following a procedure for finding transport coefficients (Sec. (II.4.A)), drift velocity can be written in an analytical form

$$W = e^{\frac{1}{2}}\left(\frac{m+m_0}{mm_0^2}\right)^{\frac{1}{4}}\frac{\left(\frac{E}{n_0}\right)^{\frac{1}{2}}}{(\sigma^0)^{\frac{1}{2}}}. \tag{67}$$

Differentiation of Eq. (67) w.r.t. electric field E and Eq. (66) gives

$$n_0^2 S_L = \frac{1}{72}e^{\frac{1}{2}}m_0\left(\frac{m+m_0}{mm_0^2}\right)^{\frac{5}{4}}\frac{\left(\frac{E}{n_0}\right)^{\frac{1}{2}}}{(\sigma^0)^{\frac{5}{2}}}. \tag{68}$$

This formula is explicitly given in Huxley and Crompton [17].

Results for skewness S_L versus E/n_0 for He, Ne and Ar, calculated according to Eq. (66) are compared with Boltzmann solutions [14] in Fig. ((1)). Calculations have been performed with a standard MTT solutions and with the cross sections for rare gases from the recommendations of the JILA Data Center [18]. We were not able to obtain identical sets of cross sections. The higher order

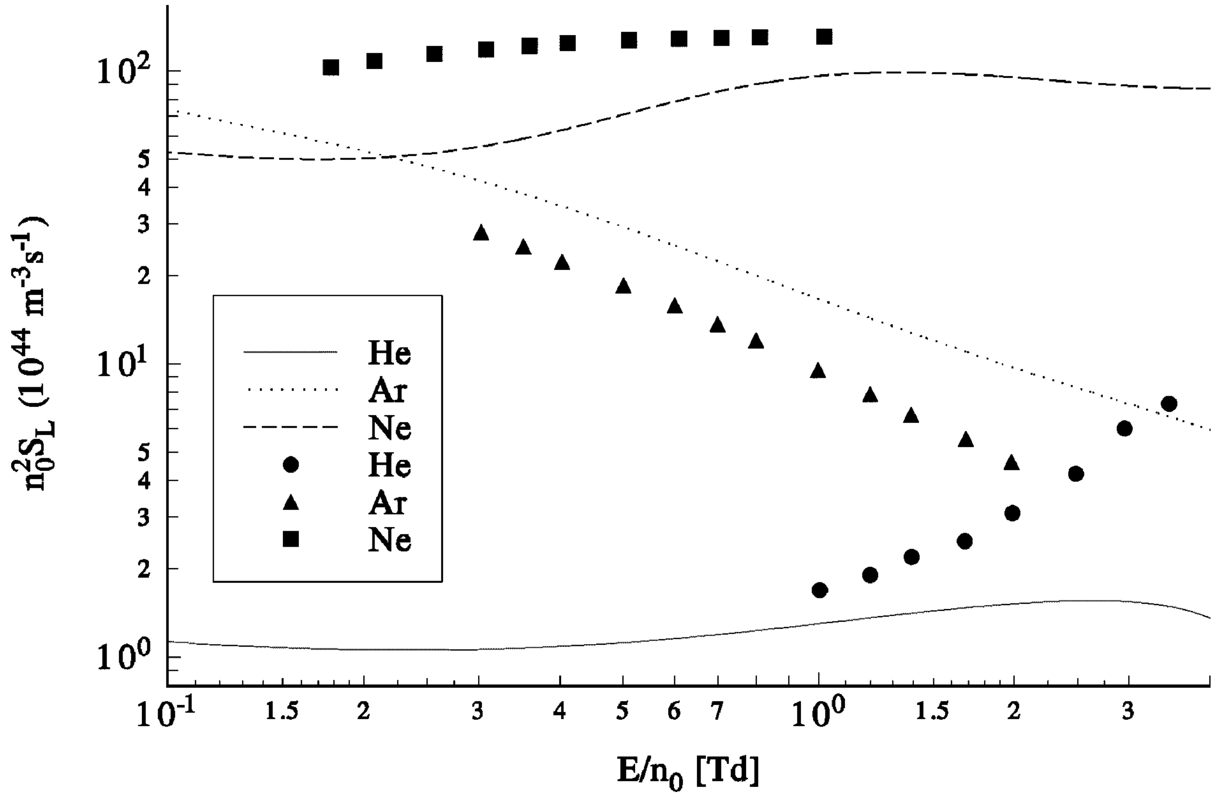

FIG. 1. Points – Boltzmann solutions; Lines – MTT solutions, Eq. ((66)).

coefficients are very sensitive to the details in the shape of the cross section. It is obvious that cross sections with a more pronounced structure will give a better agreement, as in the case of argon, while in the case of helium the results may be affected by smoothing procedure required to obtain higher order derivatives of the drift velocity (mobility). The numerical agreement would be improved by using experimental data or drift velocity (mobility) obtained from Boltzmann equation, which are more accurate than the MTT - based drift velocity.

We may conclude that MTT has been developed for higher order transport coefficients for the most general case of transport in reactive gases. Good qualitative agreement with the only available data is achieved. Quantitative agreement is satisfactory but not as good as for the lower order transport coefficients. The sensitivity of $\hat{S}^{\mathrm{mix}}$ on the details of the cross section energy dependence gives a possibility to use it as an additional source of information for cross section determination based on transport data.

III Negative Differential Conductivity

Negative Differential Conductivity (NDC) may be, rather narrowly, defined as a decrease of the drift velocity with an increasing normalized driving field E/n_0. NDC is a kinetic phenomenon which is both of fundamental interest [19, 20, 21] and of interest for a number of applications such as determination of accurate electron scattering cross sections [22] and diffuse discharge switches [23]. A specific form of time dependent NDC was found to exist in rf discharges [24, 25] which may affect power deposition efficiency.

Several explanations of NDC were available in the literature prior to 1984. Those were mostly based on the condition that the Ramsauer - Townsend minimum is present [19] and even that the conditions for the NDC and for the breakdown of the two term theory coincide [26]. Earlier theories have included most of the correct ideas [19, 20] in explanation of the NDC but were limited by the accepted assumptions and thus could not cover all its basic aspects. Petrović *et al.* [27] (PCH) have made a series of model calculations that led them do develop a set of conditions for the shapes of the cross sections that favour NDC. Robson [28] has put the NDC criterion on a much firmer ground by applying MTT.

The conditions for NDC summarized by PCH, based on their model calculation and theory by Robson [28], are the following:

(a) Inelastic processes are necessary;
(b) Increasing momentum transfer cross section favours NDC;
(c) Decreasing inelastic cross section favours NDC;
(d) Occurrence of NDC depends on magnitudes of the factors (b) and (c);
(e) Superelastic processes will have a tendency to reduce the NDC.

The above conditions represent what can be regarded as the standard NDC, i.e. the NDC that is induced by the shape of the cross sections under non-reactive swarm conditions. It turned out that the condition 1. was valid only for pure gases [28]. For mixtures it is possible to use two different atomic gases in the mixture under conditions when only elastic collisions are important and still observe NDC [9, 29, 30, 31]. The NDC criterion may be obtained in an analytic form from Eqs. (49) and (50) applied to a mixture of gases having elastic collisions only [9]:

$$1-\left[1-\frac{\frac{3}{2}kT^{\mathrm{mix}}}{\varepsilon^{\mathrm{mix}}}\right]\frac{d\ln}{d\ln\varepsilon^{\mathrm{mix}}}\left[\frac{\sum_{\alpha\in I_l}\tilde{\nu}_{0\alpha}^{(m)}}{\sum_{\alpha\in I_l}\frac{m}{m_\alpha}\tilde{\nu}_{0\alpha}^{(m)}}\right]<0. \tag{69}$$

For binary gas mixtures, the criterion (69) becomes [9]:

$$1-\frac{1}{2}\left[\frac{eE}{m}\right]^2\frac{m_1}{(\tilde{\nu}_{01}^{(m)}+\tilde{\nu}_{02}^{(m)})^2}\frac{d}{d\varepsilon}\left[\frac{\tilde{\nu}_{01}^{(m)}+\tilde{\nu}_{02}^{(m)}}{\tilde{\nu}_{01}^{(m)}+\frac{m_1}{m_2}\tilde{\nu}_{02}^{(m)}}\right]<0. \tag{70}$$

From Eqs. (69) and (70) one can find the conditions for the left-hand side of the equation to be negative. The derivate with respect to the energy has the collision frequencies in the numerator and the mass normalized collision frequencies in the denominator. The different energy dependencies of collision frequencies are required with quite different masses as well, which would reduce or amplify the difference in the denominator. The NDC will not occur for low mean energies close to the thermal equilibrium because of the first bracket in the second term of Eq. (69).

For binary mixtures when $m_1 \ll m_2$ and $\tilde{\nu}_{01}^{(m)} \approx$ const a rapid increase of $\tilde{\nu}_{02}^{(m)}$ with E/n_0 will tend to induce a negative slope of drift velocity provided that n_{01}/n_{02} is sufficiently small. In case that mean energy dependencies of collisional frequencies are similar or masses of atoms of the two constituent gases are similar no NDC will develop. What basically occurs in this case is that energy losses in collisions of electrons with lighter atomic gas play the role of the energy controlling inelastic process [9, 30, 31].

In case of a pure gas with inelastic and reactive collisions the criterion for the development of NDC may be written in the form [9]:

$$\frac{d}{d\varepsilon}\left[\varepsilon\tilde{s}_e^{(*)}\right]+\frac{d\bar{\Omega}_e^{(*)}}{d\varepsilon}<0, \tag{71}$$

where

$$\tilde{s}_e^{(*)}=1+\frac{\tilde{\nu}^{(I)}+\frac{2}{3}\varepsilon\tilde{\nu}_1^{(A)}}{\tilde{\nu}_e},\qquad \bar{\Omega}_e^{(*)}=\frac{\bar{\Lambda}}{\tilde{\nu}_e}. \tag{72}$$

If we neglect the reactive collisions, the equation (71) becomes identical to the criterion developed by Robson [28].

It is possible to predict easily the effect of superelastic collisions on NDC [24]. In all criteria superelastic collision rates appear with symmetry to the inelastic losses i.e. only the sign is changed.

That means that exactly the opposite conditions, i.e. an increase with the mean energy, will be valid for superelastic collisions. While it is difficult to find exact conditions when the shape of cross sections will be such that superelastic process will contribute more to the NDC than the negative effect of inelastic collisions, it may not be impossible.

The basic difference between Robson's criterion and that of Vrhovac and Petrović [9] is in the first term on the l.h.s. (See Eq. (71)). That term can be written explicitly as:

$$\frac{d}{d\varepsilon}(\varepsilon \tilde{s}_e^{(*)}) = 1 + \frac{\tilde{\nu}^{(I)} + \frac{2}{3}\varepsilon\tilde{\nu}_1^{(A)}}{\tilde{\nu}_e} + \varepsilon\frac{d}{d\varepsilon}\left[\frac{\tilde{\nu}^{(I)} + \frac{2}{3}\varepsilon\tilde{\nu}_1^{(A)}}{\tilde{\nu}_e}\right]. \tag{73}$$

One should note that attachment and ionization are not equivalent in this criterion, i.e. attachment collision rate is present through it's first derivative $\tilde{\nu}_1^{(A)}$. NDC will be supported by decreasing ionization collision frequency, a condition that is not likely to occur for realistic conditions. However, for the attachment, collision frequency may have a shape that is required to induce the NDC, may even have it for two different E/n_0 below and above the maximum. The requirement is that the rate of change of the slope changes sign to negative values which is commonly realized. One should be warned that this effect promoting the NDC is purely due to the reactive nature of collisions and should be added to the standard cause of the NDC due to the shape of cross sections. If we make a set of cross sections that gives the mean velocity of electrons just on the verge of NDC then NDC may be induced by changing the nature of one of the inelastic processes into reactive - ionization or by adding attachment.

When reactive collisions are present the drift velocity is different from the mean velocity of electrons [32] and thus the condition for the NDC for drift velocity changes to [9]:

$$\frac{dW}{dE} = \frac{d\omega}{dE} - \frac{1}{e}\frac{2}{3}\left[\frac{d\varepsilon}{dE}\frac{d\alpha^*}{dE} + \varepsilon\frac{d^2\alpha^*}{dE^2}\right] < 0, \tag{74}$$

which contains the first term due to the NDC in the mean velocity and the second two terms due to correction for reactive collisions which will affect greatly the occurrence and the position of the NDC. If we neglect ionization and all inelastic collisions from Eq. (71) we obtain:

$$1 + \frac{\frac{2}{3}\varepsilon\tilde{\nu}_1^{(A)}}{\tilde{\nu}_e} + \varepsilon\frac{d}{d\varepsilon}\left[\frac{\frac{2}{3}\varepsilon\tilde{\nu}_1^{(A)}}{\tilde{\nu}_e}\right] < 0, \tag{75}$$

which indicates that in the presence of elastic collisions only, even in a single gas, attachment may induce the NDC.

The NDC can thus be induced by the basic shape of the cross sections (collisional rates) as a function of the energy, by reactive corrections and by reactive collision term as well. It was also discovered that electron-electron collisions may induce the NDC [33] though the effect may be reduced due to the thermalizing effect of electron - electron collisions. We may expect additional causes of NDC due to different kinetic processes and this effect may prove to be one of the most interesting targets for fundamental studies of gas discharge kinetics, with some possible applications [34].

IV Composition dependence of drift velocities in gas mixtures in the presence of inelastic and reactive collisions

Experimental data on ion drift velocities in gas mixtures especially at high values of E/n_0 are usually not available for all gases and E/n_0 values. Also there are no analytical procedures to calculate approximately the drift velocities at high values of E/n_0. The values of drift velocities in gas mixtures at very low E/n_0 may be obtained by using Blanc's law:

$$\frac{1}{W^{\text{mix}}} = \sum_{\alpha \in I_l} x_\alpha \frac{1}{W_\alpha} \tag{76}$$

where W^{mix} is the drift velocity in gas mixture, x_α the concentration of the α-th gas in which the drift velocity is W_α. Equation (76) has been tested experimentally [35, 36] for situation when it is not exactly applicable i.e. for E/n_0 not very close to zero. Since it is practically impossible to achieve the conditions for which Blanc's law is exactly satisfied we are interested in producing a correction factor δ_B which could give more reliable results:

$$\frac{1}{W^{\text{mix}}} = \sum_{\alpha \in I_l} x_\alpha \frac{1}{W_\alpha} + \delta_B. \tag{77}$$

A number of attempts to predict theoretically the deviation δ_B at higher E/n_0 values were published. Mason and Hahn [2] derived an equation to calculate mobilities in gas mixtures at arbitrary reduced field strengths. There was an error in the expression for the energy partitioning. Correcting the error Whealton *et al.* [4] obtained an equation which is successful in explaining *qualitatively* the deviations δ_B for K^+ ions [36] in mixtures of $He + Ne$, $Ne + Ar$ and $H_2 + N_2$. The theoretical treatment applies only to situations when all the collisions between ions and gas atoms are *elastic* and the collision frequencies are slowly varying functions of E/n_0. The quantitative agreement between the equations and the experimental values is however not satisfactory.

In this section we analyze the application of Blanc's law in situations when *inelastic* and *reactive* collisions are present. A momentum - transfer theory [5, 9] is used to obtain an expression for the deviation δ_B.

Under spatially uniform, steady state conditions the momentum (33) and energy balance (34) equations may be written in the following forms:

$$e\vec{E} = m\langle\vec{v}\rangle^{\text{mix}} \left[\sum_{\alpha \in I_l} M_\alpha n_\alpha \tilde{\eta}^{(m)}_{\alpha 0}(\varepsilon^0_\alpha) + \sum_{\alpha \in I_l} n_\alpha \tilde{\eta}^{(I)}_\alpha(\varepsilon^0_\alpha) + \xi^{\text{mix}} \sum_{\alpha \in I_l} M_\alpha n_\alpha \tilde{\eta}^{(A)}_{\alpha 1}(\varepsilon^0_\alpha) \right] \tag{78}$$

$$\begin{aligned} e\vec{E}\langle\vec{v}\rangle^{\text{mix}} &= \left[m\langle v^2\rangle^{\text{mix}} - 3kT^{\text{mix}} \right] \sum_{\alpha \in I_l} M_\alpha M^0_\alpha n_\alpha \tilde{\eta}^{(m)}_{\alpha 0}(\varepsilon^0_\alpha) + \sum_{\alpha \in I_l} n_\alpha \bar{\Delta}_\alpha(\varepsilon^0_\alpha) \\ &+ \frac{1}{2} m\langle v^2\rangle^{\text{mix}} \sum_{\alpha \in I_l} n_\alpha \tilde{\eta}^{(I)}_\alpha(\varepsilon^0_\alpha) + \frac{1}{2}\left[\langle v^2\rangle^{\text{mix}} + \left(\langle\vec{v}\rangle^{\text{mix}} \right)^2 \right] \xi^{mix} \sum_{\alpha \in I_l} M_\alpha n_\alpha \tilde{\eta}^{(A)}_{\alpha 1}(\varepsilon^0_\alpha). \end{aligned} \tag{79}$$

where we use the notation

$$\bar{\Delta}_\alpha(\varepsilon^0_\alpha) = \sum_{s \in I^{(\text{in})}_\alpha} \Delta E^s_\alpha \tilde{\nu}_{s\alpha}(\varepsilon^0_\alpha) + \varepsilon^{(I)}_\alpha \tilde{\nu}^{(I)}_\alpha(\varepsilon^0_\alpha) + \sum_{s \in I^{(I)}_\alpha} \Delta\varepsilon^{(I)}_{s\alpha} \tilde{\nu}^{(I)}_{s\alpha}(\varepsilon^0_\alpha), \quad \alpha \in I_l. \tag{80}$$

All collision frequencies $\tilde{\eta}$ are normalized to unit number density. In pure gases consisting of one of the mixture components (α) we can simplify the equations (78) and (79):

$$e\vec{E} = m\langle\vec{v}\rangle^\alpha \left[M_\alpha n_0 \tilde{\eta}^{(m)}_{\alpha 0}(\varepsilon'_\alpha) + n_0 \tilde{\eta}^{(I)}_\alpha(\varepsilon'_\alpha) + \xi^\alpha M_\alpha n_0 \tilde{\eta}^{(A)}_{\alpha 1}(\varepsilon'_\alpha) \right] \tag{81}$$

$$\begin{aligned} e\vec{E}\langle\vec{v}\rangle^\alpha &= \left[m\langle v^2\rangle^\alpha - 3kT^\alpha \right] M_\alpha M^0_\alpha n_0 \tilde{\eta}^{(m)}_{\alpha 0}(\varepsilon'_\alpha) + n_0 \bar{\Delta}_\alpha(\varepsilon'_\alpha) \\ &+ \frac{1}{2} m\langle v^2\rangle^\alpha n_0 \tilde{\eta}^{(I)}_\alpha(\varepsilon'_\alpha) + \frac{1}{2} m \left[\langle v^2\rangle^\alpha + (\langle\vec{v}\rangle^\alpha)^2 \right] \xi^\alpha M_\alpha n_0 \tilde{\eta}^{(A)}_{\alpha 1}(\varepsilon'_\alpha). \end{aligned} \tag{82}$$

Elimination of electric field E between Eqs. (78), (79) and (81) gives

$$\frac{1}{\langle\vec{v}\rangle_z^{\mathrm{mix}}} = \sum_{\alpha\in I_l} x_\alpha \frac{1}{\langle\vec{v}\rangle_z^\alpha} + \delta_B \tag{83}$$

where

$$\begin{aligned}\delta_B = \sum_{\alpha\in I_l} &\Big[M_\alpha\left(\tilde{\eta}^{(m)}_{\alpha 0}(\varepsilon^0_\alpha) - \tilde{\eta}^{(m)}_{\alpha 0}(\varepsilon'_\alpha)\right) + \left(\tilde{\eta}^{(I)}_{\alpha}(\varepsilon^0_\alpha) - \tilde{\eta}^{(I)}_{\alpha}(\varepsilon'_\alpha)\right)\\ &+\xi^\alpha M_\alpha\left(\tilde{\eta}^{(A)}_{\alpha 1}(\varepsilon^0_\alpha) - \tilde{\eta}^{(A)}_{\alpha 1}(\varepsilon'_\alpha)\right)\Big]\left[M_\alpha\tilde{\eta}^{(m)}_{\alpha 0}(\varepsilon'_\alpha) + \tilde{\eta}^{(I)}_{\alpha}(\varepsilon'_\alpha) + \xi^\alpha M_\alpha\tilde{\eta}^{(A)}_{\alpha 1}(\varepsilon'_\alpha)\right]^{-1} x_\alpha\frac{1}{\langle\vec{v}\rangle_z^\alpha}.\end{aligned} \tag{84}$$

Equation (83) is of the same form as Eq. (77). Note that, in the presence of reactive collisions mean velocities $\langle\vec{v}\rangle^{mix}$ and $\langle\vec{v}\rangle^\alpha$ are not equal to the corresponding drift velocities.

Here we assume no attaching collisions. When we expand collision frequencies in Taylor series in the vicinity of the mean energy ε'_α we obtain

$$\tilde{\eta}^{(m)}_{\alpha 0}(\varepsilon^0_\alpha) - \tilde{\eta}^{(m)}_{\alpha 0}(\varepsilon'_\alpha) \approx \frac{d\tilde{\eta}^{(m)}_{\alpha 0}}{d\varepsilon'_\alpha}\delta\varepsilon'_\alpha, \qquad \tilde{\eta}^{(I)}_{\alpha}(\varepsilon^0_\alpha) - \tilde{\eta}^{(I)}_{\alpha}(\varepsilon'_\alpha) \approx \frac{d\tilde{\eta}^{(I)}_{\alpha}}{d\varepsilon'_\alpha}\delta\varepsilon'_\alpha. \tag{85}$$

We assume that the Taylor expansions (85) converge rapidly in the neighborhood of mean energies ε'_α. Substituting these expansions into Eq. (84) leads after some algebra to the following expression

$$\delta_B = \sum_{\alpha\in I_l} x_\alpha \frac{\dfrac{1}{E_z} - \dfrac{1}{\langle\vec{v}\rangle_z^\alpha}\dfrac{d\langle\vec{v}\rangle_z^\alpha}{dE_z}}{\langle\vec{v}\rangle_z^\alpha}\,\frac{\delta\varepsilon'_\alpha}{\dfrac{d\varepsilon'_\alpha}{dE_z}}. \tag{86}$$

Equation (86) can be recommended as an expression for finding deviation δ_B from Blanc's law in the presence of inelastic and reactive (ionization) collisions. If no reactive collisions are present we can express the deviation from Blanc's law entirely in terms of the properties of the swarm particles in pure components:

$$\begin{aligned}\delta_B = \sum_{\alpha\in I_l} &\frac{1 + \frac{m_\alpha}{2e}\frac{\langle\vec{v}\rangle_z^\alpha}{E_z/n_0}\frac{d\Xi^{(\mathrm{in})}_\alpha}{d\varepsilon'_\alpha}}{\frac{m_\alpha}{2e}\frac{\langle\vec{v}\rangle_z^\alpha}{E_z/n_0}\Xi^{(\mathrm{in})}_\alpha(\varepsilon'_\alpha) + m_\alpha\left(\langle\vec{v}\rangle_z^\alpha\right)^2\frac{d\ln\langle\vec{v}\rangle_z^\alpha}{d\ln E}\left(1 - \frac{d\ln\langle\vec{v}\rangle_z^\alpha}{d\ln E}\right)^{-1}}\\ &\frac{1}{2}\mu_\alpha\left\{\frac{1 - \sum_\alpha x_\alpha\frac{1}{\langle\vec{v}\rangle_z^\alpha}\sum_\alpha x_\alpha\frac{\Xi^{(\mathrm{in})}_\alpha(\varepsilon'_\alpha)}{e(E_z/n_0)}}{\sum_\alpha x_\alpha\frac{1}{\langle\vec{v}\rangle_z^\alpha}\sum_\alpha x_\alpha M^0_\alpha\frac{1}{\langle\vec{v}\rangle_z^\alpha}} - \frac{1}{M^0_\alpha}\left[\left(\langle\vec{v}\rangle_z^\alpha\right)^2 + \frac{\Xi^{(\mathrm{in})}_\alpha(\varepsilon'_\alpha)}{e(E_z/n_0)}\langle\vec{v}\rangle_z^\alpha\right]\right\} x_\alpha\frac{1}{\langle\vec{v}\rangle_z^\alpha}.\end{aligned} \tag{87}$$

The quantity $\Xi^{(\mathrm{in})}_\alpha$ is

$$\Xi^{(\mathrm{in})}_\alpha = \sum_{s\in I^{(\mathrm{in})}_\alpha}\Delta E^s_\alpha\tilde{\eta}_{s\alpha}(\varepsilon'_\alpha). \tag{88}$$

Here the collision frequency for inelastic proces (s) is $\tilde{\eta}_{s\alpha}$. The threshold for these inelastic collisions is denoted by ΔE^s_α. The calculation of δ_B proceeds in a similar manner as in [4]. When inelastic collisions are not present, Eq. ((87)) is identical to the deviation from Blanc's law proposed by Whealton *et al.* [4].

The extension of Blanc's law correction was done on the basic of a belief that the application of the corrected form of Blanc's law may become an option at high E/n_0 where electron energy distribution function for different gases may be of similar shape and elastic collisions may play an increasing role. At the same time inelastic collisions would be dominated by ionization and shapes of cross sections for numerous different gases are likely to be similar. These conditions are however useful for gas discharge modeling which requires a broad range of data for gas mixtures currently lacking in the literature.

V Concluding remarks

The main results reviewed in this paper are:

(a) Extension of momentum - transfer theory to study non - conservative swarm particle transport in gas *mixtures*.

(b) Development of MTT for *reactive* swarm particles in crossed electric and *magnetic* fields. We have obtained approximate formulas for relationships between the transport properties of swarm in $\vec{E}\times\vec{B}$ fields. Non-particle-conserving collisions, such as ionisation and attachment, have been included.

(c) Unified treatment of negative differential conductivity in mixtures of gases with elastic collisions only and for a single gas with reaction.

(d) Derivation of formulas, such as the (modified) generalized Einstein relations and the expressions for the energy partitioning (Wannier energy relations).

(e) Derivation of approximate expression for higher order transport coefficients.

(f) Extension of Blanc's law correction to include inelastic collisions and reactions.

The MTT is useful within the physics of charged particle (and neutral) swarms [37] as it provides a set of useful analytic relations and very accurate conditions for the occurrence of some phenomena (such as NDC). At the same time it may be used as a source of quantitative data provided that high accuracy is not required. This may be so in gas discharge modelling, especially for DC and RF plasmas [24, 38, 39, 40] used in plasma processing. In fact, our main motivation for developing MTT was to develop a basis for fluid like modelling of plasmas that would be more efficient than the simulation schemes [41] yet still flexible enough to take account of non - local electron kinetics in a natural way. Such equations have been developed for beam - like electron kinetics in high E/n_0 swarms [42, 43]. In addition space - time dependent formulation of MTT is required which is already available in a preliminary form [8].

A Appendix

When we expand functions $\vec{\omega}^{\mathrm{mix}}$ (17) and α^* (53) in Taylor series in the vicinity of $\vec{E}$ to the second and third order in $\vec{G}$, respectively, we obtain the following equations:

$$\begin{aligned}\langle\vec{v}\rangle_q^{\mathrm{mix}} &= \omega_q^{\mathrm{mix}}\left(\vec{E}-\frac{k}{e}\hat{T}^{\mathrm{mix}}\vec{G}\right) = \omega_q^{\mathrm{mix}}(\vec{E}) - \frac{k}{e}\sum_{i=1}^{3}(\hat{T}^{\mathrm{mix}}\vec{G})_i\frac{\partial\omega_q^{\mathrm{mix}}}{\partial E_i}\\ &+\frac{1}{2}\frac{k^2}{e^2}\sum_{i=1}^{3}\sum_{j=1}^{3}(\hat{T}^{\mathrm{mix}}\vec{G})_i(\hat{T}^{\mathrm{mix}}\vec{G})_j\frac{\partial^2\omega_q^{\mathrm{mix}}}{\partial E_i\partial E_j},\quad q=1,2,3;\end{aligned} \tag{89}$$

$$\begin{aligned}\tilde{\nu}^* &= \alpha^*\left(\vec{E}-\frac{k}{e}\hat{T}^{\mathrm{mix}}\vec{G}\right) = \alpha^*(\vec{E}) - \frac{k}{e}\sum_{i=1}^{3}(\hat{T}^{\mathrm{mix}}\vec{G})_i\frac{\partial\alpha^*}{\partial E_i}\\ &+\frac{1}{2}\frac{k^2}{e^2}\sum_{i=1}^{3}\sum_{j=1}^{3}(\hat{T}^{\mathrm{mix}}\vec{G})_i(\hat{T}^{\mathrm{mix}}\vec{G})_j\frac{\partial^2\alpha^*}{\partial E_i\partial E_j}\\ &-\frac{1}{6}\frac{k^3}{e^3}\sum_{i=1}^{3}\sum_{j=1}^{3}\sum_{k=1}^{3}(\hat{T}^{\mathrm{mix}}\vec{G})_i(\hat{T}^{\mathrm{mix}}\vec{G})_j(\hat{T}^{\mathrm{mix}}\vec{G})_k\frac{\partial^3\alpha^*}{\partial E_i\partial E_j\partial E_k}.\end{aligned} \tag{90}$$

Substituting these expansions into equation of continuity (1), we obtain

$$\frac{\partial}{\partial t}n(\vec{r},t)+k_1-k_2+k_3=-n(\vec{r},t)\alpha^*(\vec{E}), \tag{91}$$

where

$$k_1=\vec{\omega}^{\mathrm{mix}}\cdot\frac{\partial n}{\partial\vec{r}}-\frac{k}{e}\sum_{i=1}^{3}\frac{\partial\alpha^*}{\partial E_i}\left(\hat{T}^{\mathrm{mix}}\frac{\partial n}{\partial\vec{r}}\right)_i, \tag{92}$$

$$k_2 = -\frac{k}{e}\frac{\partial}{\partial \vec{r}}\sum_{i=1}^{3}\frac{\partial \vec{\omega}^{\mathrm{mix}}}{\partial E_i}\left(\hat{T}^{\mathrm{mix}}\frac{\partial n}{\partial \vec{r}}\right)_i - \frac{1}{n}\frac{1}{2}\frac{k^2}{e^2}\sum_{i=1}^{3}\sum_{j=1}^{3}\frac{\partial^2 \alpha^*}{\partial E_i \partial E_j}\left(\hat{T}^{\mathrm{mix}}\frac{\partial n}{\partial \vec{r}}\right)_i\left(\hat{T}^{\mathrm{mix}}\frac{\partial n}{\partial \vec{r}}\right)_j, \tag{93}$$

$$\begin{aligned} k_3 = {} & \frac{\partial}{\partial \vec{r}}\left[\frac{1}{n}\frac{1}{2}\frac{k^2}{e^2}\sum_{i=1}^{3}\sum_{j=1}^{3}\frac{\partial^2 \vec{\omega}^{\mathrm{mix}}}{\partial E_i \partial E_j}\left(\hat{T}^{\mathrm{mix}}\frac{\partial n}{\partial \vec{r}}\right)_i\left(\hat{T}^{\mathrm{mix}}\frac{\partial n}{\partial \vec{r}}\right)_j\right] \\ & - \frac{1}{n^2}\frac{1}{6}\frac{k^3}{e^3}\sum_{i=1}^{3}\sum_{j=1}^{3}\sum_{k=1}^{3}\frac{\partial^3}{\partial E_i \partial E_j \partial E_k}\left(\hat{T}^{\mathrm{mix}}\frac{\partial n}{\partial \vec{r}}\right)_i\left(\hat{T}^{\mathrm{mix}}\frac{\partial n}{\partial \vec{r}}\right)_j\left(\hat{T}^{\mathrm{mix}}\frac{\partial n}{\partial \vec{r}}\right)_k. \end{aligned} \tag{94}$$

Equation (92) can be arranged to give

$$k_1 = \vec{W}^{\mathrm{mix}} \cdot \frac{\partial n}{\partial \vec{r}}, \tag{95}$$

where vector $\vec{W}^{\mathrm{mix}}$ (drift velocity) is given by Eq. (54).

We now suppose that temperature tensor $\hat{T}^{\mathrm{mix}}$ is a spatially homogeneous quantity. We can also assume that

$$\frac{1}{n}\frac{\partial n}{\partial x_i}\frac{\partial n}{\partial x_j} \approx \frac{\partial}{\partial x_i}\frac{\partial}{\partial x_j}n, \quad i,j = 1,2,3. \tag{96}$$

Using above assumption and Eq. (54), we can rearrange Eq. (93):

$$k_2 = \sum_{i=1}^{3}\sum_{j=1}^{3} D_{ij}^{\mathrm{mix}}\frac{\partial}{\partial x_i}\frac{\partial}{\partial x_j}n. \tag{97}$$

Tensor D_{ij}^{mix} (diffusion tensor) is given by Eq. (55). Following approximation is also an acceptable substitution:

$$\frac{1}{n^2}\frac{\partial n}{\partial x_i}\frac{\partial n}{\partial x_j}\frac{\partial n}{\partial x_k} \approx \frac{\partial}{\partial x_i}\frac{\partial}{\partial x_j}\frac{\partial}{\partial x_k}n, \quad i,j,k = 1,2,3. \tag{98}$$

With Eq. (98), equation (94) becomes

$$\begin{aligned} k_3 = {} & \frac{1}{2}\frac{k^2}{e^2}\sum_{i=1}^{3}\sum_{j=1}^{3}\sum_{k=1}^{3}\left\{\sum_{l=1}^{3} t_{il}^{\mathrm{mix}}\sum_{m=1}^{3} t_{jm}^{\mathrm{mix}}\left[\frac{\partial}{\partial E_m}\left(\frac{\partial \omega_k^{\mathrm{mix}}}{\partial E_l}\right)\right.\right. \\ & \left.\left. -\frac{1}{3}\frac{k}{e}\sum_{n=1}^{3} t_{kn}^{\mathrm{mix}}\frac{\partial}{\partial E_n}\frac{\partial}{\partial E_m}\left(\frac{\partial \alpha^*}{\partial E_l}\right)\right]\right\}\frac{\partial}{\partial x_i}\frac{\partial}{\partial x_j}\frac{\partial}{\partial x_k}n. \end{aligned} \tag{99}$$

Using Eq. (54), we can rearrange Eq. (99),

$$k_3 = \sum_{i=1}^{3}\sum_{j=1}^{3}\sum_{k=1}^{3} S_{ijk}^{\mathrm{mix}}\frac{\partial}{\partial x_i}\frac{\partial}{\partial x_j}\frac{\partial}{\partial x_k}n, \tag{100}$$

where tensor S_{ijk}^{mix} (skewness) is given by Eq. (57).

It is obvious from Eqs. (3) and (91) that net average reaction rate ϱ^* is given by Eq. (56).

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