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Validity of the Onsager relations in relativistic binary mixtures

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In this work we study the properties of a relativistic mixture of two nonreacting dilute species in thermal local equilibrium. Following the conventional ideas in kinetic theory, we use the concept of chaotic velocity. In particular, we address the nature of the density, or pressure gradient term that arises in the solution of the linearized Boltzmann equation in this context. Such an effect, also present for the single component problem, has, so far, not been analyzed from the point of view of the Onsager resciprocity relations. To address this matter, we propose two alternatives for the Onsagerian matrix which comply with the corresponding reciprocity relations. The implications of both representations are briefly analyzed.

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I. INTRODUCTION

Relativistic kinetic theory has become a rather fashionable subject in recent years. Not only is this due to many astrophysical and cosmological phenomena which occur in dilute gases at high temperatures, but also because, for a while, it was believed it would find an important application in the study of the quark-gluon plasma which is formed in relativistic heavy ion collisions (RHIC). Although this last feature is questionable [1–3], mostly because a relativistic quantum hydrodynamical theory is required, interest in it still remains due to its applications to classical systems.

It is our view that, in spite of the existence of the wealth of approaches to this classical problem, which goes back to Israel *et al*. [4–7], there are two aspects that have been so far ignored in previous works. First, the formulation of the theory using the rather useful concept of chaotic (thermal) velocities of the molecules composing the gas. Second, the study of the so-called cross effects in irreversible thermodynamics. Also, and even more important, the possibility of selecting appropriate representations of fluxes and their conjugate forces in which one can provide an airtight proof on the validity on Onsager's reciprocity relations (ORR) has, to our knowledge, never been given. It is important to emphasize at this point that the validity of the ORR is one of the fundamental postulates of linear irreversible thermodynamics (LIT).

The introduction of the concept of thermal velocity has been successfully accomplished for a single-component dilute gas and its advantages clearly underlined in the calculation of their transport properties [8,9]. Perhaps it is worth stressing that in this formulation the relativistic generalization of the classical expression for the heat flux obtained emphasizes the nature of heat, namely, the transport of the kinetic or thermal energy of the molecules. Further, one can obtain in a rigorous way the expression for the relativistic stress tensor as proposed phenomenologically by Eckart [10].

In this paper we study the second feature as mentioned above, the cross effects and the validity of the ORR in a binary, nonreactive, dilute mixture of gases. The most surprising result is that there are two representations in which the ORR hold true depending on how fluxes and forces are selected. One of the representations follows the idea formulated by previous authors of coupling in one single force both the temperature and pressure gradients, this force being the direct drive for the heat flux. The ORR are verified in that context. The second one is based on the novel idea that due to the noninvariance of the volume elements of the gas under Lorentz transformations, a "volume flux" results whose conjugate force is the pressure gradient. Resemblance to this idea arose in at least one phenomenological derivation of Burnett's constitutive equations, but it has no connection with our result [11]. Further, two cross effects are present in this approach which are completely absent in the nonrelativistic case.

To accomplish this task, we divide the article as follows. Section II is devoted to the basic concepts of the relativistic kinetic theory, as well as the derivation of the conservation equations. In Sec. III we use the Chapman-Enskog method to linearize the Boltzmann equation. In Sec. IV we select the appropriate thermodynamic forces following the ideas in Refs. [6,7], and we show that the Onsager reciprocity relations [12–14] in a 2×2 matrix hold. In Sec. V we propose the idea of a purely relativistic flux directly coupled with the pressure gradient, which satisfies the symmetry of a 3×3 "Onsagerian" matrix. Finally, in Sec. VI, we include a discussion and concluding remarks.

II. RELATIVISTIC KINETIC THEORY

As mentioned above, we study a relativistic, dilute mixture of two nonreacting species in thermal local equilibrium. In the framework of kinetic theory, we consider the quantity

$$f_{(1)} d^3x d^3v_{(1)} + f_{(2)} d^3x d^3v_{(2)}, \tag{1}$$

which represents the number of particles of species (1) and (2) in $d^3x d^3v_{(1)}$ and $d^3x d^3v_{(2)}$, where $v^{\alpha}_{(i)}$ denotes molecular velocity. To establish a clear notation, we use parentheses in the subscripts to denote species. For components, Latin subscripts run form 1 to 3 for the spatial ones while Greek subscripts are used for four-vectors and tensors running from 1 to 4 in Minkowski's space-time with a $+++-$ signature, also a comma will be used to denote a covariant derivative.

The invariant Boltzmann equations for the mixture are

$$v_{(i)}^{\alpha} f_{(i),\alpha} = \sum_{i,j=1}^{2} J_{(ij)}, \tag{2}$$

where the collisional term is given by [7]

$$\sum_{i,j=1}^{2} J_{(ij)} = \sum_{i,j=1}^{2} \int (f'_{(i)} f'_{(j)} - f_{(i)} f_{(j)}) F_{(ij)} \sigma_{(ij)} d\Omega_{(ji)} d^3 v^*_{(j)}. \tag{3}$$

Here, $F_{(ij)}$, $\sigma_{(ij)}$, and $d\Omega_{(ji)}$ denote the invariant flux, the invariant differential elastic cross section, and the element of the solid angle that characterize a binary collision between the particles of constituent i with those of constituent j, respectively. The differential $d^3 v^*_{(i)}$ stands for $\frac{d^3 v_{(i)}}{v^4_{(i)}}$, also an invariant. The cross-section $\sigma_{(ij)}$ has special symmetries [15] that guarantee the existence of inverse collisions such that the principle of microscopic reversibility is satisfied. The quantities $f_{(i)}$ and $f'_{(i)}$ denote the distribution functions before and after a collision, respectively.

The collisional invariants in this case are the rest mass of each species $m_{(i)}$ and the four-momentum $m_{(i)} v^{\alpha}_{(i)}$, where the energy is included in the temporal component $m_{(i)} v^4_{(i)}$. In the following sections these quantities will be used to obtain balance equations. It is important to notice at this point that the molecular velocity in the previous equations is measured by an observer in an arbitrary frame, which we call laboratory frame.

A. Particle number conservation

By multiplying the Boltzmann equation (2), by $m_{(i)}$ and integrating over $d^3 v^*_{(i)}$ one finds

$$\left(m_{(i)} \int v^{\alpha}_{(i)} f_{(i)} d^3 v^*_{(i)} \right)_{,\alpha} = 0, \tag{4}$$

where

$$N^{\alpha}_{(i)} = m_{(i)} \int v^{\alpha}_{(i)} f_{(i)} d^3 v^*_{(i)}, \tag{5}$$

is the mass four-flux in an arbitrary frame. The barycentric velocity is thus defined as

$$n U^{\alpha} = \frac{N^{\alpha}_{(1)}}{m_{(1)}} + \frac{N^{\alpha}_{(2)}}{m_{(2)}}, \tag{6}$$

which is consistent with Eckart's definition for the hydrodynamic four-velocity. Here $n = n_{(1)} + n_{(2)}$ is the particle number density and represents an invariant. We also define the relativistic diffusive four-flux in the comoving frame as

$$J^{\alpha}_{(i)} = m_{(i)} \int K^{\alpha}_{(i)} f_{(i)} d^3 K^*_{(i)}, \tag{7}$$

where $K^{\alpha}_{(i)}$ is the four-velocity of the particles of the species i measured in the comoving frame (i.e., $K^{\alpha}_{(i)}$ is the chaotic or thermal velocity) [16–18]. Then $N^{\alpha}_{(i)}$ and $J^{\beta}_{(i)}$ are related by a Lorentz transformation as follows

$$N^{\alpha}_{(i)} = \mathcal{L}^{\alpha}_{\beta} J^{\beta}_{(i)}, \tag{8}$$

where $\mathcal{L}^{\alpha}_{\beta}$ is the transformation from the comoving frame, where $U^m = 0$, to an arbitrary one moving with a four-velocity U^{α}.

With the help of these equations, one can find the complete particle number conservation equation, see Ref. [19]. In this work we only need them at Euler's level because we will use the Chapman and Enskog method up to first order in the gradients. Thus, we have that

$$n_{(i)} U^{\alpha}_{,\alpha} + U^{\alpha} n_{(i),\alpha} = 0, \tag{9}$$

for the particle number conservation.

B. Momentum and energy balance

To obtain the energy-momentum balance for the mixture, Boltzmann's equation is now multiplied by $m_{(i)} v^{\alpha}_{(i)}$ and integrated over $d^3 v^*_{(i)}$, which yields

$$T^{\beta\alpha}_{,\alpha} = \left(T^{\beta\alpha}_{(1)} + T^{\beta\alpha}_{(2)} \right)_{,\alpha} = 0, \tag{10}$$

where

$$T^{\beta\alpha} = \sum_i m_{(i)} \int v^{\beta}_{(i)} v^{\alpha}_{(i)} f_{(i)} d^3 v^*_{(i)}. \tag{11}$$

To establish the form of the tensor $T^{\beta\alpha}$ we recognize that, as defined in Eq. (11), it is referred to an arbitrary reference frame. Thus, we can express it in terms of $\tilde{T}^{\gamma\phi}$, measured in the comoving frame defined above, as

$$T^{\beta\alpha} = \mathcal{L}^{\beta}_{\gamma} \mathcal{L}^{\alpha}_{\phi} \tilde{T}^{\gamma\phi}, \tag{12}$$

where again $\mathcal{L}^{\alpha}_{\phi}$ and $\mathcal{L}^{\beta}_{\gamma}$ are the Lorentz transformations from the comoving frame to an arbitrary one moving with a four-velocity U^{α}. Following Weinberg [20] and using the fact that the stress-energy tensor is symmetric [see Eq. (11)], we assume that in the comoving frame it has the form

$$\tilde{T}^{\beta\alpha} \doteq \begin{pmatrix} p & 0 & 0 & 0 \\ 0 & p & 0 & 0 \\ 0 & 0 & p & 0 \\ 0 & 0 & 0 & ne \end{pmatrix} + \begin{pmatrix} 0 & 0 & 0 & q^1 \\ 0 & 0 & 0 & q^2 \\ 0 & 0 & 0 & q^3 \\ q^1 & q^2 & q^3 & 0 \end{pmatrix} + \begin{pmatrix} \pi^{11} & \pi^{12} & \pi^{13} & 0 \\ \pi^{12} & \pi^{22} & \pi^{23} & 0 \\ \pi^{13} & \pi^{23} & \pi^{33} & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}. \tag{13}$$

In Eq. (13) we have separated the proper equilibrium quantities, namely, the hydrostatic pressure

$$p = \tfrac{1}{3} \tilde{T}^{mm}, \tag{14}$$

and the energy density per particle

$$ne = \tilde{T}^{44}. \tag{15}$$

On the other hand, the nonequilibrium quantities are

$$q^m = c\tilde{T}^{4m} = c\tilde{T}^{m4}, \tag{16}$$

for the heat flux and

$$\Pi^{mn} = \tilde{T}^{mn}, \tag{17}$$

for the Navier tensor. Introducing Eq. (13) in Eq. (12) yields

$$T^{\alpha\beta} = pg^{\alpha\beta} + \frac{1}{c^2}(p+ne)U^\alpha U^\beta + \frac{1}{c^2}\left(U^\alpha \mathcal{L}^\beta_\mu q^\mu + U^\beta \mathcal{L}^\alpha_\mu q^\mu\right) + \mathcal{L}^\beta_\mu \mathcal{L}^\alpha_\nu \Pi^{\mu\nu}, \tag{18}$$

where $g^{\alpha\beta}$ is the metric tensor. In Eq. (18) we identify the first two terms as the relativistic energy-momentum tensor at Euler's level. The third and fourth terms represent the nonequilibrium generalization with the heat and viscous dissipation terms as found from kinetic theory grounds for the single fluid in Ref. [9].

We now calculate the derivative in Eq. (10) using Eq. (18) and its projection with the four-velocity, namely, $U_\mu T^{\mu\nu}_{,\nu}$. Neglecting all the terms which contain corrections whose order is beyond Euler's regime [19] and after laborious calculations one finds

$$\tilde{\rho}\dot{U}^\beta + h^{\beta\nu} p_{,\nu} = 0, \tag{19}$$

and

$$n\dot{e} = -pU^\mu_{,\mu}, \tag{20}$$

where Eqs. (19) and (20) are the momentum and internal energy balance equations, respectively. Here

$$\tilde{\rho} = \sum_i m_{(i)} n_{(i)} G(z_{(i)}) = \tilde{\rho}_{(1)} + \tilde{\rho}_{(2)}, \tag{21}$$

and

$$G(z_{(i)}) = \frac{\mathcal{K}_3\left(\frac{1}{z_{(i)}}\right)}{\mathcal{K}_2\left(\frac{1}{z_{(i)}}\right)}, \tag{22}$$

with $z_{(i)} = \frac{kT}{m_{(i)}c^2}$ being the well-known relativistic parameter. The dot denotes a proper time derivative and is defined as $\dot{(\,)} = U^\mu(\,)_{,\mu}$. Here $\mathcal{K}_n(\frac{1}{z_{(i)}})$ is the n^{th} modified Bessel function of the second kind.

Equation (20) is related to the temperature evolution by assuming that the internal energy density depends only on the temperature $e = C_v T$. The details of the calculations above can be found in Refs. [9,19].

III. LINEARIZATION OF THE BOLTZMANN EQUATION

In this section we proceed to apply the well-known Chapman-Enskog method to linearize the covariant form of Boltzmann's equation. Following the ideas in Ref. [9], we will perform all calculations in the comoving frame such that Eq. (2) now reads

$$K^\alpha_{(i)} f_{(i),\alpha} = \sum_{j=1}^{2} J_{(ij)}, \tag{23}$$

where $K^\alpha_{(i)}$ is the four-velocity measured in such a frame. As usual, we now assume that the distribution functions $f_{(i)}(x^\alpha, K^\alpha_{(i)}, t)$ can be taken as functionals of the locally conserved variables, namely $f_{(i)}(x^\alpha, K^\alpha_{(i)} | n_{(i)}, U^\alpha, T)$, and further, they may be expanded in a power series of an inhomogeneity parameter around the local equilibrium distribution function $f^{(0)}_{(i)}$ defined in an arbitrary frame as [21–23]

$$f^{(0)}_{(i)} = \frac{n_{(i)}}{4\pi c^3 z_{(i)} \mathcal{K}_2\left(\frac{1}{z_{(i)}}\right)} \exp\left(\frac{U^\beta v_{(i)\beta}}{z_{(i)} c^2}\right), \tag{24}$$

which in the comoving frame reduces to

$$f^{(0)}_{(i)} = \frac{n_{(i)}}{4\pi c^3 z_{(i)} \mathcal{K}_2\left(\frac{1}{z_{(i)}}\right)} \exp\left(-\frac{\gamma_{k_{(i)}}}{z_{(i)}}\right), \tag{25}$$

where $\gamma_{k_{(i)}} = (1 - k^2_{(i)}/c^2)^{-1/2}$ is the usual Lorentz factor and $k^2_{(i)}$ is the squared magnitude of the chaotic or thermal three-velocity. Omitting unnecessary arguments, we resort to the linear theory [24] and expand Eq. (23) as

$$f_{(i)} = f^{(0)}_{(i)}(1 + \phi_{(i)}). \tag{26}$$

The substitution of Eq. (26) into Eq. (23) with the help of the functional hypothesis and Eqs. (9), (19), and (20) leads to

$$K^m_{(i)}\left\{-\gamma_{k_{(i)}} \frac{1}{z_{(i)} c^2 \tilde{\rho}} p_{,m} + (\ln n_{(i)})_{,m} + \left[1 + \frac{1}{z_{(i)}}[\gamma_{k_{(i)}} - G(z_{(i)})]\right](\ln T)_{,m}\right\} = [C(\phi_{(i)}) + C(\phi_{(i)} + \phi_{(j)})]. \tag{27}$$

Notice that in Eq. (27) we have omitted the second rank tensorial terms since Curie's principle establishes that in isotropic systems only forces and fluxes of the same tensorial rank couple among themselves. Clearly, there is an equation similar to Eq. (27) for species j. The linearized collision kernel now reads

$$C(\phi_{(i)} + \phi_{(j)}) = \int \cdots \int f^{(0)}_{(i)} f^{(0)}_{(j)} (\phi_{(j)}{}' + \phi_{(i)}{}' - \phi_{(j)} - \phi_{(i)}) \times F_{(ij)} \sigma_{(ij)} d\Omega_{(ji)} d^3 v^*_{(j)}, \tag{28}$$

and

$$C(\phi_{(i)}) = \int \cdots \int f^{(0)}_{(i)} f^{(0)}_{(i)} (\phi_{(i)}{}' + \phi_{(i)}{}' - \phi_{(i)} - \phi_{(i)}) \times F_{(ii)} \sigma_{(ii)} d\Omega_{(ii)} d^3 v^*_{(i)}.$$

The left-hand side of Eq. (27) contains terms involving gradients of the intensive thermodynamical variables $p_{,m}$, $(n_{(i)})_{,m}$, and $T_{,m}$, which we identify with thermodynamic forces. The question that arises is how to select among them a representation in which Onsager's reciprocity relations hopefully turn out to be valid. This will be discussed in the following sections.

IV. SOLUTION WITH TWO THERMODYNAMIC FORCES

Following the statement issued above, we will proceed to discuss the aforementioned representations. For instance, we first rearrange the left-hand side of Eq. (27) to read as

$$K^m_{(i)}\left\{[d_{m(ij)}] + \frac{1}{z_{(i)}}[\gamma_{k_{(i)}} - G(z_{(i)})]\left[\frac{T_{,m}}{T} - \frac{1}{nh_E} p_{,m}\right]\right\} = [C(\phi_{(i)}) + C(\phi_{(i)} + \phi_{(j)})], \tag{29}$$

where

$$d_{m(ij)} = n_{(j)}\left(\frac{m_{(j)}G(z_{(j)}) - m_{(i)}G(z_{(i)})}{\tilde{\rho}}\right)\frac{p_{,m}}{p} + \frac{n}{n_{(i)}}(n_{i0})_{,m}, \tag{30}$$

and using the notation $nh_E = \tilde{\rho}c^2$, and $n_{i0} = \frac{n_{(i)}}{n}$ representing an invariant. This choice implies that we are considering two vector forces in the system, namely

$$d_{m(ij)} \quad \text{and} \quad \frac{T_{,m}}{T} - \frac{1}{nh_E}p_{,m}. \tag{31}$$

The second term may be regarded as related to a generalized Fourier's equation with a thermal force that includes both temperature and pressure gradients [7]. Further it may be shown that in the nonrelativistic limit, the coefficient of such a force in Eq. (29) reduces to

$$\frac{1}{z_{(i)}}[\gamma_{k_{(i)}} - G(z_{(i)})] \rightarrow \frac{m_{(i)}k_{(i)}^2}{2k_BT} - \frac{5}{2}, \tag{32}$$

and the coefficient of the pressure gradient vanishes because $[\gamma_{k_{(i)}} - G(z_{(i)})] \rightarrow 0$. Thus, the inhomogeneous term in Eq. (27) reduces to the well-known expression of the classical linearized Boltzmann equation.

On the other hand, $d_{m(ij)} = -d_{m(ji)} \equiv d_m$ may be considered as a generalization of the standard diffusive force to a relativistic scheme since, indeed, in the nonrelativistic case Eq. (30) reduces to

$$d_{m(ij)} \rightarrow \frac{n_{(j)}}{\rho p}(m_{(j)} - m_{(i)})\nabla p + \frac{n}{n_{(i)}}\nabla n_{i0}, \tag{33}$$

which is in accordance with phenomenological [26] and kinetic [27] classical expressions.

Having selected the above thermodynamic forces, the solution to Eq. (29) reads as [28,29]

$$\phi_{(i)} = -K_{(i)}^m A_{(i)}\left[\frac{T_{,m}}{T} - \frac{1}{nh_E}p_{,m}\right] - \sum_i K_{(i)}^m D_{(i)} d_m. \tag{34}$$

The substitution of Eq. (34) in Eq. (29) leads to two independent equations. The first one is for the scalar $A_{(i)}$ and is related to the temperature and pressure gradients

$$K_{(i)}^m \frac{1}{z_{(i)}}[\gamma_{k_{(i)}} - G(z_{(i)})] = -\sum_j \int \cdots \int f_{(i)}^{(0)} f_{(j)}^{(0)}\left[K_{(j)}^{m}{}' A_{(j)}{}' + K_{(i)}^{m}{}' A_{(i)}{}' - K_{(j)}^m A_{(j)} - K_{(i)}^m A_{(i)}\right] F_{(ij)}\sigma_{(ij)} d\Omega_{(ji)} d^3K_{(j)}^*. \tag{35}$$

The second one is related to the scalar function $D_{(i)}$ related to the diffusive force, namely

$$K_{(i)}^m = -\sum_j \int \cdots \int f_{(i)}^{(0)} f_{(j)}^{(0)}\left[K_{(j)}^{m}{}' D_{(j)}{}' + K_{(i)}^{m}{}' D_{(i)}{}' - K_{(j)}^m D_{(j)} - K_{(i)}^m D_{(i)}\right] F_{(ij)}\sigma_{(ij)} d\Omega_{(ji)} d^3K_{(j)}^*. \tag{36}$$

We will now use the expressions for the mass and energy fluxes arising in this representation to prove the validity of Onsager's reciprocity relations in this scheme. The diffusive mass flux has been defined in Eq. (7), which, with the help of Eqs. (34) and (26), can be written as follows:

$$\frac{J_{(i)}^m}{m_{(i)}} = -\frac{1}{3}\int f_{(i)}^{(0)} K_{(i)}^n K_{n(i)} A_{(i)} d^3K_{(i)}^* \left[\frac{T^{,m}}{T} - \frac{1}{nh_E}p^{,m}\right] - \frac{1}{3}\int f_{(i)}^{(0)} K_{(i)}^n K_{n(i)} D_{(i)} d^3K_{(i)}^* d^m. \tag{37}$$

In Eq. (37) the transport coefficients are identified as

$$\frac{J_{(i)}^m}{m_{(i)}} = -L_{dq}\left[\frac{T^{,m}}{T} - \frac{1}{nh_E}p^{,m}\right] - L_{dd}d^m, \tag{38}$$

where L_{dq} and L_{dd} are the integrals appearing in Eq. (37).

For the energy flux we propose the form which is given in the literature [26]

$$\frac{q_{\text{tot}}^m}{kT} = \frac{1}{kT}\sum_i \left(q_{(i)}^m - h_{(i)}J_{(i)}^m\right), \tag{39}$$

where

$$h_{(i)} = \frac{kT}{z_{(i)}}G(z_{(i)}), \tag{40}$$

is the enthalpy [6]. After Eqs. (16) and (7) are introduced in Eq. (39) one obtains the complete form for the total heat flux q_{tot}^m which reads

$$\begin{aligned}\frac{q_{\text{tot}}^m}{kT} = &-\frac{1}{3}\sum_i \int f_{(i)}^{(0)} \frac{1}{z_{(i)}}[\gamma_{k_{(i)}} - G(z_{(i)})] K_{(i)}^n K_{n(i)} A_{(i)} d^3K_{(i)}^* \left[\frac{T^{,m}}{T} - \frac{1}{nh_E}p^{,m}\right] \\ &- \frac{1}{3}\sum_i \int f_{(i)}^{(0)} \frac{1}{z_{(i)}}[\gamma_{k_{(i)}} - G(z_{(i)})] K_{(i)}^n K_{n(i)} D_{(i)} d^3K_{(i)}^* d^m,\end{aligned} \tag{41}$$

as we can see, this expression has two contributions, the one given by the first term to which we may refer as the modified Fourier's equation and the second term related with the diffusive force d^m. In short, this equation may be rewritten as

$$\frac{q^m_{\rm tot}}{kT} = -L_{qq}\left[\frac{T^{,m}}{T} - \frac{1}{nh_E}p^{,m}\right] - L_{qd}d^m. \tag{42}$$

Equations (38) and (42) are now in a form which, by a similar analysis as the one performed in the classical case (see Ref. [27]), are bound to lead to the required relations of symmetry.

To show this we start by constructing an "Onsagerian" matrix, namely

$$\begin{pmatrix} q^m_{\rm tot} \\ J^m_{(i)} \end{pmatrix} = -\begin{pmatrix} L_{qq} & L_{qd} \\ L_{dq} & L_{dd} \end{pmatrix}\begin{pmatrix} \frac{T^{,m}}{T} - \frac{1}{nh_E}p^{,m} \\ d^m \end{pmatrix}. \tag{43}$$

Then one proceeds by multiplying both sides of Eq. (36) by $K_{(i)m}A_{(i)}$ and integrating over $d^3K^*_{(i)}$ to obtain the form

$$\int \frac{1}{3}(K^n_{(i)}K_{n(i)})A_{(i)}d^3K^*_{(i)} = -\sum_j \int \cdots \int f^{(0)}_{(i)} f^{(0)}_{(j)}\left[K^m_{(j)}{}'D_{(j)}{}' + K^m_{(i)}{}'D_{(i)}{}' - K^m_{(j)}D_{(j)} - K^m_{(i)}D_{(i)}\right]$$
$$\times K_{(i)m}A_i F_{(ij)}\sigma_{(ij)}d\Omega_{(ji)}d^3K^*_{(j)}d^3K^*_{(i)} \equiv \{D,A\}. \tag{44}$$

On the other hand, multiplying Eq. (35) by $K_{(i)m}D_{(i)}$ and integrating over $dK^*_{(i)}$ yields

$$\int \frac{1}{3}(K^n_{(i)}K_{n(i)})\frac{1}{z_{(i)}}\left[\gamma_{k_{(i)}} - G(z_{(i)})\right]D_{(i)}d^3K^*_{(i)} = -\sum_j \int \cdots \int f^{(0)}_{(i)} f^{(0)}_{(j)}\left[K^m_{(j)}{}'A_{(j)}{}' + K^m_{(i)}{}'A_{(i)} - K^m_{(j)}A_{(j)} - K^m_{(i)}A_{(i)}\right]$$
$$\times K_{(i)m}D_{(i)}F_{(ij)}\sigma_{(ij)}d\Omega_{(ji)}d^3K^*_{(j)}d^3K^*_{(i)} \equiv \{A,D\}. \tag{45}$$

Equations (44) and (45) may be symmetrized by taking into account the invariance of $F_{(ij)}\sigma_{(ij)}d\Omega_{(ji)}d^3K^*_{(j)}d^3K^*_{(i)}$ (see Ref. [7]), and using the same symmetry arguments as in the conventional proof of the H-theorem. Such a procedure leads to

$$\{A,D\} = \{D,A\}, \tag{46}$$

and thus

$$L_{dq} = L_{qd}. \tag{47}$$

Emphasis should be made on the fact that we verified the reciprocity of the Onsager relations using the standard kinetic definition for two fluxes, but not for the forces. In this section, we are assuming that the generalization for the Fourier's equation has the form given by Eq. (42). Here $p^{,m}$ is considered as part of this force to obtain integral equations in which the transformation of their kernels fulfill the symmetry requirements. Thus in this representation one cannot speak of the canonical forms of the Dufour-Soret effects that relate the diffusion coefficients to *strictly the thermal conductivity*.

However, in the following section we will overcome this difficulty by introducing a volumetric flow which arises solely from the fact that in the theory of relativity volumes are not invariants. This representation is completely new in the context of relativistic kinetic theory.

V. SOLUTION WITH THREE THERMODYNAMIC FORCES

In this section we explore the possibility of a third thermodynamic flux in the system. The motivation behind such a task is the interest in keeping the temperature and pressure gradients as independent forces which would yield a Fourier-type constitutive equation for the heat flux relating it exclusively to a temperature gradient. This will imply that the heat flux caused by a pressure gradient constitutes a cross effect. This is a purely relativistic effect and we shall see how it relates to the pressure, or density, gradient term that arises in the case of the high temperature one-component gas.

To achieve this new representation we start by rearranging Eq. (27) as follows:

$$K^m_{(i)}\left\{d_m + \frac{1}{z_{(i)}}[\gamma_{k_{(i)}} - G(z_{(i)})]\frac{T_{,m}}{T} - [\gamma_{k_{(i)}} - G(z_{(i)})]\left[\frac{n_{(i)}m_{(i)}}{\tilde\rho}\frac{p_{,m}}{p_{(i)}}\right]\right\} = [C(\phi_{(i)}) + C(\phi_{(i)} + \phi_{(j)})], \tag{48}$$

for species i, recalling that there is a similar equation for species j. Notice that we are considering a new force

$$V_{(i)m} \equiv \frac{n_{(i)}m_{(i)}}{\tilde\rho}\frac{p_{,m}}{p_{(i)}},$$

which satisfies

$$V_{(1)m} = \frac{m_{(1)}}{m_{(2)}}V_{(2)m} \equiv V_m. \tag{49}$$

Equation (48) leads to a solution of the form

$$\phi_{(i)} = -K^m_{(i)}A_{(i)}\frac{T_{,m}}{T} - \sum_j K^m_{(j)}B_{(j)}V_m - \sum_j K^m_{(j)}D_{(j)}d_m. \tag{50}$$

The substitution of Eq. (50) into Eq. (48) yields three independent equations. The first one, which is related with the heat flux, is for the scalar function $A_{(i)}$, namely

$$K^m_{(i)}\frac{1}{z_{(i)}}[\gamma_{k_{(i)}} - G(z_{(i)})] = [C(K^m_{(i)}A_{(i)}) + C(K^m_{(i)}A_{(i)} + K^m_{(j)}A_{(j)})]. \tag{51}$$

The second one is related to the volume flux V^m and is for the scalar function $B_{(i)}$, namely

$$K^m_{(i)}[\gamma_{k_{(i)}} - G(z_{(i)})] = [C(K^m_{(i)}B_{(i)}) + C(K^m_{(i)}B_{(i)} + K^m_{(j)}B_{(j)})]. \tag{52}$$

And finally the third one must be satisfied by the scalar function $D_{(i)}$ and reads

$$K^m_{(i)} = [C(K^m_{(i)}D_{(i)}) + C(K^m_{(i)}D_{(i)} + K^m_{(j)}D_{(j)})]. \tag{53}$$

Equation (52) is now the new ingredient in this representation. To understand its physical meaning we proceed as follows. Consider the motion of an individual particle which collides with another one. After the collision it will travel a length λ, the mean free path, before colliding with a third one. Recall also that the mean free time is much greater than the collision time. One can thus construct a sphere centered in the particle (in general, it can be any other geometric figure) with volume $V = \frac{4}{3}\pi\lambda^3$ that, when the speed of the particle is comparable with the speed of light, by Lorentz's contraction, is deformed into a ellipsoid with volume $\frac{4}{3}\pi\lambda^3\gamma_k$. Therefore, in the relativistic case, an observer sees a change in this volume with a privileged direction $\vec{k}$. This is the process which gives rise to "volume or volumetric flow" and a system with an apparently additional state variable. To explore its significance we establish the transport equation characterizing its flow. In the case of a binary mixture by multiplying Boltzmann's equation by the microscopic change in the volume $a\gamma_{k_{(i)}}$ where a is a constant, and integrating over the velocities $d^3K^*_{(i)}$ yields

$$\left(\int \gamma_{k(i)}K^\alpha_{(i)}f_{(i)}d^3K^*_{(i)}\right)_{,\alpha} = \int \gamma_{k_{(i)}}(J_{(ii)} + J_{(ij)})d^3K^*_{(i)} = \pi_{\text{vol}}, \tag{54}$$

which is a balance equation for the change in the volume in the gas. Notice that in the nonrelativistic limit, the right-hand side vanishes, implying that there is no such change in volume. The physical implications of this flux are further discussed in the final section.

In the case of mixtures, the energy flux corresponding to heat dissipation to be considered in Onsager's formalism is constructed by subtracting the diffusive mass flux times the enthalpy from the heat flux [26,30]. In a similar fashion, we define the total volume (adimensional) flux as

$$J^m_V = \sum_i\left(\int \gamma_{k_{(i)}}K^m_{(i)}f_{(i)}d^3K^*_{(i)} - \frac{h_{E(i)}}{m_{(i)}c^2}\frac{J^m_{(i)}}{m_{(i)}}\right), \tag{55}$$

where $n_{(i)}h_{E(i)} = c^2\tilde{\rho}_{(i)}$. Thus, using Eqs. (50) and (26) we have that

$$\begin{aligned} J^m_V = &-\frac{1}{3}\sum_i\int f^{(0)}_{(i)}[\gamma_{k_{(i)}} - G(z_{(i)})]K^n_{(i)}K_{(i)n}A_{(i)}d^3K^*_{(i)}\frac{T^{,m}}{T}\\ &-\frac{1}{3}\sum_i\int f^{(0)}_{(i)}[\gamma_{k_i} - G(z_{(i)})]K^n_{(i)}K_{(i)n}B_{(i)}d^3K^*_{(i)}V^m\\ &-\frac{1}{3}\sum_i\int f^{(0)}_{(i)}[\gamma_{k_{(i)}} - G(z_{(i)})]K^n_{(i)}K_{(i)n}D_{(i)}d^3K^*_{(i)}d^m, \end{aligned} \tag{56}$$

or

$$J^m_V = -L_{Vq}\frac{T^{,m}}{T} - L_{VV}V^m - L_{Vd}d^m, \tag{57}$$

which introduces two new transport cross-coefficients L_{Vq}, L_{Vd} and one corresponding to the direct effect L_{VV}.

As mentioned before, the dissipative energy flux is given by

$$\begin{aligned} \frac{q^m_{\text{tot}}}{k_BT} = &-\frac{1}{3}\sum_i\int f^{(0)}_{(i)}\frac{1}{z_{(i)}}[\gamma_{k_{(i)}} - G(z_{(i)})]K^n_{(i)}K_{n(i)}A_{(i)}d^3K^*_{(i)}\frac{T^{,m}}{T} - \frac{1}{3}\sum_i\int f^{(0)}_{(i)}\frac{1}{z_{(i)}}[\gamma_{k_{(i)}} - G(z_{(i)})]K^n_{(i)}K_{n(i)}B_{(i)}d^3K^*_{(i)}V^m\\ &-\frac{1}{3}\sum_i\int f^{(0)}_{(i)}\frac{1}{z_{(i)}}[\gamma_{k_{(i)}} - G(z_{(i)})]K^n_{(i)}K_{n(i)}D_{(i)}d^3K^*_{(i)}d^m \end{aligned} \tag{58}$$

or

$$\frac{q^m_{\text{tot}}}{k_BT} = -L_{qq}\frac{T^{,m}}{T} - L_{qV}V^m - L_{qd}d^m, \tag{59}$$

and for the mass flow we have

$$\frac{J^m_{(i)}}{m_{(i)}} = -\frac{1}{3}\int f^{(0)}_{(i)}K^n_{(i)}K_{n(i)}A_{(i)}d^3K^*_{(i)}\frac{T^{,m}}{T} - \frac{1}{3}\int f^{(0)}_{(i)}K^n_{(i)}K_{n(i)}B_{(i)}d^3K^*_{(i)}V^m - \frac{1}{3}\int f^{(0)}_{(i)}K^n_{(i)}K_{n(i)}D_{(i)}d^3K^*_{(i)}d^m, \tag{60}$$

which can also be written as

$$\frac{J^m_{(i)}}{m_{(i)}} = -L_{dq(i)}\frac{T^{,m}}{T} - L_{dV(i)}V^m - L_{dd(i)}d^m. \tag{61}$$

From the previous equations, one can readily identify the Soret and Dufour cross effects. The verification of the Onsager reciprocity relations will support that these are the correct generalizations for such effects.

Equations (57), (59), and (61) will be explored to see whether they comply with the Onsager reciprocity relations. As before, we construct the Onsagerian matrix

$$\begin{pmatrix} q^m_{\text{tot}} \\ J^m_{(i)} \\ J^m_V \end{pmatrix} = -\begin{pmatrix} L_{qq} & L_{qd} & L_{qV} \\ L_{dq(i)} & L_{dd(i)} & L_{dV(i)} \\ L_{Vq} & L_{Vd} & L_{VV} \end{pmatrix} \begin{pmatrix} \frac{T^{,m}}{T} \\ d^m_{(i)} \\ V^m \end{pmatrix}, \tag{62}$$

where we introduced the term V^m as the direct driving force for the volume flux J^m_V. Then, by the same procedure and arguments as those in the previous section, we will verify the symmetries

$$\sum L_{dq(i)} \stackrel{?}{=} L_{qd}, \tag{63}$$

$$L_{Vq} \stackrel{?}{=} L_{qV}, \tag{64}$$

$$L_{Vd} \stackrel{?}{=} \sum L_{dV(i)}. \tag{65}$$

First, for Eqs. (63), (51), and (53) are multiplied by $K^m_{(i)}D_{(i)}$ and $K^m_{(i)}A_{(i)}$, respectively. After integration over $d^3K^*_{(i)}$ one finds

$$\int K^n_{(i)}\frac{1}{z_{(i)}}\big[\gamma_{k_{(i)}} - G(z_{(i)})\big]K_{n(i)}D_{(i)}dK^*_{(i)} = -\sum_j \int\cdots\int f^{(0)}_{(i)} f^{(0)}_{(j)}\big[K^m_{(j)}{}'A_{(j)}{}' + K^m_{(i)}{}'A_{(i)}{}' - K^m_{(j)}A_{(j)} - K^m_{(i)}A_{(i)}\big] \times K^m_{(i)}D_{(i)}F_{(ij)}\sigma_{(ij)}d\Omega_{(ji)}d^3K^*_{(j)}d^3K^*_{(i)} \equiv \{A,D\}, \tag{66}$$

or

$$\int K^n_{(i)}K_{n(i)}A_{(i)}dK^*_{(i)} = -\sum_j \int\cdots\int f^{(0)}_{(i)} f^{(0)}_{(j)}\big[K^m_{(j)}{}'D_{(j)}{}' + K^m_{(i)}{}'D_{(i)}{}' - K^m_{(j)}D_{(j)} - K^m_{(i)}D_{(i)}\big] \times K_{(i)m}A_{(i)}F_{(ij)}\sigma_{(ij)}d\Omega_{(ji)}d^3K^*_{(j)}d^3K^*_{(i)} \equiv \{D,A\}, \tag{67}$$

where, by the symmetry properties of the collisional term, $\{A,D\} = \{D,A\}$, implying that Eq. (63) holds. Second, multiplying Eqs. (51) and (52) by $K^m_{(i)}B_{(i)}$ and $K^m_{(i)}A_{(i)}$, respectively, and integrating over $d^3K^*_{(i)}$ yields

$$\int K^n_{(i)}\frac{1}{z_{(i)}}\big[\gamma_{k_{(i)}} - G(z_{(i)})big\big]K_{n(i)}B_{(i)}dK^*_{(i)} = -\sum_j \int\cdots\int f^{(0)}_{(i)} f^{(0)}_{(j)}\big[K^m_{(j)}{}'A_{(j)}{}' + K^m_{(i)}{}'A_{(i)}{}' - K^m_{(j)}A_{(j)} - K^m_{(i)}A_{(i)}\big] \times K_{m(i)}B_{(i)}F_{(ij)}\sigma_{(ij)}d\Omega_{(ji)}d^3K^*_{(j)}d^3K^*_{(i)}, \equiv \{A,B\}, \tag{68}$$

or

$$\int K^n_{(i)}\big[\gamma_{k_{(i)}} - G(z_{(i)})\big]K_{n(i)}A_{(i)}dK^*_{(i)} = -\sum_j \int\cdots\int f^{(0)}_{(i)} f^{(0)}_{(j)}\big[K^m_{(j)}{}'B_{(j)}{}' + K^m_{(i)}{}'B_{(i)}{}' - K^m_{(j)}B_{(j)} - K^m_{(i)}B_{(i)}\big] \times K_{m(i)}A_{(i)}F_{(ij)}\sigma_{(ij)}d\Omega_{(ji)}d^3K^*_{(j)}d^3K^*_{(i)}, \equiv \{B,A\}, \tag{69}$$

and since $\{A,B\} = \{B,A\}$, Eq. (64) holds. Last, Eqs. (52) and (53) are multiplied by $K^m_{(i)}D_{(i)}$ and $K^m_{(i)}B_{(i)}$, respectively, yielding

$$\int K^n_{(i)}\big[\gamma_{k_{(i)}} - G(z_{(i)})\big]K_{n(i)}D_{(i)}d^3K^*_{(i)} = -\sum_j \int\cdots\int f^{(0)}_{(i)} f^{(0)}_{(j)}\big[K^m_{(j)}{}'B_{(j)}{}' + K^m_{(i)}{}'B_{(i)}{}' - K^m_{(j)}B_{(j)} - K^m_{(i)}B_{(i)}\big] \times K_{m(i)}D_{(i)}d^3K^*_{(i)} \equiv \{B,D\}, \tag{70}$$

$$\int K^n_{(i)}K_{n(i)}B_{(i)}dK^*_{(i)} = -\sum_j \int\cdots\int f^{(0)}_{(i)} f^{(0)}_{(j)}\big[K^m_{(j)}{}'D_{(j)}{}' + K^m_{(i)}{}'D_{(i)}{}' - K^m_{(j)}D_{(j)} - K^m_{(i)}D_{(i)}\big] \times K_{(i)m}B_{(i)}F_{(ij)}\sigma_{(ij)}d\Omega_{(ji)}d^3K^*_{(j)}d^3K^*_{(i)}, \equiv \{D,B\}, \tag{71}$$

where again, $\{B,D\} = \{D,B\}$, justifying Eq. (65).

At this point we have verified that Onsager's symmetries hold in this representation. The authentic Dufour effect corresponds to the transport coefficient L_{qd}, while the Soret effect is related to L_{dq}, whose explicit expressions are depicted in Eqs. (59) and (61). Now, from the Onsagerian matrix we identify two new cross effects represented by L_{dV} and L_{qV}. These effects do not appear in the nonrelativistic theory and their physical meaning will be discussed elsewhere.

The Onsager reciprocity relations are not necessarily fulfilled in other representations. In classical irreversible thermodynamics one often finds that many writers believe that the appropriate thermodynamic forces to describe cross effects in the case of mixtures are the chemical potentials of the species. For the nonrelativistic case, when the mixture is nonisothermal it was clearly shown in Ref. [27] that this is incorrect. In such a representation the ORR do not hold true. The same statement is valid for a nonisothermal binary mixture of inert gases in special relativity, the details of the proof are practically identical as in the nonrelativistic case so we omit it.

VI. DISCUSSION

In this paper we have shown that the introduction of the concept of thermal velocity is equally useful to deal with transport properties of diluted mixtures. In fact, the expression we obtained for the total heat flux $J^m_{\rm tot}$ is consistent with its expression in the phenomenological theory as well as in the nonrelativistic case. Second, we insist that the new result exhibits the existence of two representations in which the ORR are valid. The one discussed in Sec. IV, where the forces are those that have been used by the authors of Refs. [6,7] for the simple component gas, is characterized by the fact that Fourier's like equation has to be modified by the presence of a pressure gradient.

In the second case as discussed in Sec. V, we propose the new idea of the volume flux, which may be introduced without modifying the classical Fourier equation, and also gives rise to the canonical form for the Dufour and Soret effects related with L_{qd} and L_{dq}. This representation provides two cross effects that are only present in the relativistic case, namely L_{qV} and L_{dV}. Indeed, one can immediately see from Eq. (52) that this contribution vanishes in the nonrelativistic limit.

Notice that the volume flow as introduced in Eq. (57) may be regarded as a multiple of the heat flux Eq. (59) in the single-fluid limit. As shown in the Appendix, the constitutive equation for the heat flux and for the volume flux in this limit coincide. Thus, what in the binary mixture is a cross effect turns into a direct effect with a Fourier-type constitutive equation in the single-fluid limit.

The variable associated with the volume transport has a peculiar thermodynamical meaning. This volume flux with its conjugated force is indeed related to the thermodynamic description of the system, and when taken into account, clarifies the nature of the transport phenomena in a relativistic mixture. This coupling of the volume flux with a pressure gradient is indeed confirmed when calculating the entropy production of the mixture, which constitutes a work in progress and will be published elsewhere.

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APPENDIX

In this Appendix we will take the single-fluid limit of the equations for the volume flux and the heat flux. Taking $m_{(i)} = m_{(j)} = m$, $n_{(i)} = n_{(j)} = n$, from Eqs. (16) and (54) we have for the heat flux

$$\frac{q^m_{\rm tot}}{k_B T} = \frac{mc^2}{k_B T}\int \gamma_k K^m f d^3K^*, \tag{A1}$$

and for the volume flux

$$J^m_V = \int \gamma_k K^m f d^3K^*, \tag{A2}$$

thus

$$\frac{q^m_{\rm tot}}{k_B T} = \frac{1}{z} J^m_V. \tag{A3}$$

It remains to verify that the transport coefficients satisfy the same relations, namely, from Eqs. (57) and (59) with the fact that $d^m = 0$, recalling that $J^m_{(i)} = J^m_{(j)} = 0$ we get

$$J^m_V = -L_{Vq}\frac{T^{,m}}{T} - L_{VV}\left[\frac{nm}{\tilde\rho}\frac{p^{,m}}{p}\right], \tag{A4}$$

and

$$\frac{q^m_{\rm tot}}{k_B T} = -L_{qq}\frac{T^{,m}}{T} - L_{qV}\left[\frac{nm}{\tilde\rho}\frac{p^{,m}}{p}\right], \tag{A5}$$

where

$$L_{Vq} = -\frac{1}{3}\int f^{(0)}[\gamma_k - G(z)]K^n K_n A d^3K^*, \tag{A6}$$

$$L_{VV} = -\frac{1}{3}\int f^{(0)}[\gamma_k - G(z)]K^n K_n B d^3K^*, \tag{A7}$$

$$L_{qq} = -\frac{1}{3}\int f^{(0)}\frac{1}{z}[\gamma_k - G(z)]K^n K_n A d^3K^*, \tag{A8}$$

$$L_{qV} = -\frac{1}{3}\int f^{(0)}\frac{1}{z}[\gamma_k - G(z)]K^n K_n B d^3K^*. \tag{A9}$$

Where again we can immediately see that

$$\frac{q^m_{\rm tot}}{k_B T} = \frac{1}{z} J^m_V. \tag{A10}$$

Then, in the single-fluid limit, the volume flux turns out to be a multiple of the heat flux.

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