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Structure of transport linear systems in dilute isotropic gas mixtures

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We consider the linear systems associated with the evaluation of transport coefficients in dilute isotropic gas mixtures and corresponding to the first-order Enskog expansion. We obtain the structure and the mathematical properties of these systems directly from the properties of the integral collision Boltzmann operator and the variational framework used for its approximation. Using these structure properties, all the transport coefficients can be expanded into convergent series. These asymptotic expansions are particularly relevant to computational models in a wide range of practical applications.

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I. INTRODUCTION

In the last decades, the study of transport properties in gas mixtures has witnessed extensive progress thanks, in part, to the wide range of related practical applications [1–3]. In the context of the first-order Enskog expansion where small deviations from the local Maxwellian equilibrium distribution are considered, it is well known that the governing equations of multicomponent flows contain the terms for transport fluxes, written in turn in terms of transport coefficients and macroscopic variable gradients [1–5]. However, these transport coefficients are not given explicitly by the kinetic theory. Upon using matrix approximations for the inverse collision operator, the transport coefficients can be evaluated by solving large linear systems. Although direct inversion of these transport linear systems is feasible, it is prohibitively expensive for most practical applications involving multidimensional multicomponent flows [6].

With an eye toward the development of efficient numerical models, there is thus a strong motivation for deriving cost effective approximations for transport coefficients in a manner consistent with the underlying kinetic theory rather than by simply using empirical mixture-averaged expressions which are often less accurate. Recently, a mathematical and numerical theory of iterative algorithms for multicomponent transport has become available [3]. The basic idea of this theory is to write the transport coefficients as convergent series which yield by truncation a sequence of approximations for the transport coefficients of increasing accuracy. In practice, only a few iterations yield approximate transport coefficients which are at least an order of magnitude more accurate than their empirical counterparts.

The purpose of the present paper is now to extract the fundamental structure of the transport linear systems from the kinetic theory and to generalize the results of [3]. More specifically, we derive a set of basic assumptions for the integral collision operator and the variational space used for its approximation. These assumptions are valid in either a classical, semiclassical, or quantum mechanical isotropic kinetic theory. With these assumptions, we define a sparse transport matrix, denoted by $d_b(G)$, formed by the diagonal of the blocks of the original transport linear system matrix G. The sparse transport matrix $d_b(G)$ is then shown to satisfy some structure properties which yield convergent asymptotic expansions for the transport coefficients.

In order to keep the present discussion somewhat general, we refer to the kinetic equation for the species distribution functions simply as the generalized Boltzmann equation without any specific reference to its version (classical, semiclassical or quantum mechanical) [5]. The theory presented in this paper is, however, restricted in four ways. First, as stated earlier, we only consider small deviations from a local Maxwellian equilibrium distribution. Second, we assume the gas mixture to be sufficiently dilute so that only binary collisions are relevant. Third, transport properties are investigated in the isotropic case, thus excluding the presence of an external electric or magnetic field. Finally, with a slight exception in Sec. IV, chemically reacting mixtures are not treated in the present discussion. In Sec. II we briefly describe the transport linear systems. In Sec. III we introduce the sparse transport matrix $d_b(G)$ and extract the fundamental assumptions leading to convergent asymptotic expansions for the transport coefficients. These assumptions apply to the integral collision operator and to the variational space used for its approximation. Finally, in Sec. IV we show that the theoretical results obtained in this paper can be applied to practical problems.

II. TRANSPORT LINEAR SYSTEMS

A. Enskog expansion and collisional invariants

We consider a gas mixture with n species and we denote by $\mathcal{S}=[1,n]$ the set of species indices. In the well known first-order Enskog expansion, the species distribu-

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tion functions f_k, $k \in \mathcal{S}$, are written as a linear perturbation of the local Maxwellian equilibrium distribution functions f_k^0, $k \in \mathcal{S}$, in the form

$$f_k = f_k^0(1+\phi_k), \quad k \in \mathcal{S} \ . \tag{2.1}$$

The local Maxwellian distributions are constrained to be such that the corresponding species number densities, mass averaged velocity, and energy are those of the exact distributions. It is also assumed that the species distribution functions do not depend on time and position directly but implicitly through the macroscopic variable gradients. The species perturbed distribution functions ϕ_k, $k \in \mathcal{S}$, are then shown to be the solution of integral equations involving the linearized Boltzmann collision operator completed with appropriate integral constraints [1–5].

The linear integral equations can be written in the compact form

$$\mathfrak{I}(\phi) = \Psi \ , \tag{2.2}$$

where $\phi = (\phi_k)_{k \in \mathcal{S}}$ is the family of perturbed distribution functions, $\Psi = (\Psi_k)_{k \in \mathcal{S}}$ the family of right members, and $\mathfrak{I} = (\mathfrak{I}_k)_{k \in \mathcal{S}}$ the family of linearized collision operators. It is then convenient to expand the right member Ψ in terms of the macroscopic variable gradients X^μ in the form

$$\Psi = \sum_{\mu} \Psi^\mu \odot X^\mu \ . \tag{2.3}$$

Since X^μ can be either a scalar, a three-dimensional vector, or a three by three matrix, we employ here a general tensor notation with $\odot$ denoting the maximum contracted product between tensors. Note also that in (2.3) Ψ^μ has the same tensorial order as X^μ. We denote by $a = 0$, 1 or 2 the tensorial order of X^μ.

By isotropy, the expansion (2.3) yields a similar expansion for the species perturbed distribution functions, i.e., $\phi = \sum_\mu \phi^\mu \odot X^\mu$, where each expansion coefficient ϕ^μ satisfies the linearized Boltzmann equations

$$\mathfrak{I}(\phi^\mu) = \Psi^\mu \ . \tag{2.4}$$

The integral equations (2.4) must be completed with the constraints

$$\langle\langle \phi^\mu, \xi \rangle\rangle = 0, \quad \xi \in \mathcal{I} \ , \tag{2.5}$$

where $\langle\langle \ , \ \rangle\rangle$ is a scalar product between tensor quantities. For two families of functions $\xi = (\xi_k)_{k \in \mathcal{S}}$ and $\zeta = (\zeta_k)_{k \in \mathcal{S}}$, this scalar product is given by

$$\langle\langle \xi, \zeta \rangle\rangle = \sum_{k \in \mathcal{S}} \langle \xi_k \odot \zeta_k \rangle_k \ , \tag{2.6}$$

where $\langle \ \rangle_k$ is an averaging operator involving the local Maxwellian distribution function f_k^0 of the kth species. In (2.5), we have also introduced the space of tensorial collisional invariants of the mixture

$$\mathcal{I} = \mathrm{span}\{ \mathcal{T}_\nu \widehat{\psi}^l, \ (l,\nu) \in [1, n+4] \times [1, \tau] \} \ , \tag{2.7}$$

where $\mathcal{T}_\nu$, $\nu \in [1, \tau]$, with $\tau = 3^a$, denotes the canonical basis of the space of tensors of order a over the three-dimensional space $\mathbb{R}^3$. In addition, $\widehat{\psi}^l$ are the $n+4$ linearly independent, scalar collisional invariants of the mixture. These collisional invariants are chosen to be orthogonal with respect to the scalar product $\langle\langle \ , \ \rangle\rangle$. In the case of mixtures, the collisional invariants are families of scalar functions, i.e., we have $\widehat{\psi}^l = (\widehat{\psi}_k^l)_{k \in \mathcal{S}}$. The n collisional invariants $\widehat{\psi}^l$ for $l \in \mathcal{S}$, are associated with species type conservation and are given by $\widehat{\psi}^l = (\delta_{kl})_{k \in \mathcal{S}}$, where δ_{kl} is the Kronecker symbol. On the other hand, the collisional invariants $\widehat{\psi}^{n+\nu}$, $\nu = 1,2,3$, are associated with momentum conservation and $\widehat{\psi}^{n+4}$ with energy conservation.

B. The bracket product

Between two tensor families ξ and ζ, we define the bracket operator

$$[\xi, \zeta] = \langle\langle \xi, \mathfrak{I}(\zeta) \rangle\rangle \ . \tag{2.8}$$

As a result of the symmetry property of the collision cross sections, we now make the fundamental assumption that the bracket product can be cast into the form

$$[\xi, \zeta] = \sum_{k,l \in \mathcal{S}} \tfrac{1}{4} n_k n_l \{ \xi_k - \xi_k' + \widetilde{\xi}_l - \widetilde{\xi}_l', \zeta_k - \zeta_k' + \widetilde{\zeta}_l - \widetilde{\zeta}_l' \}_{kl} \ , \tag{2.9}$$

where primes denote values after collision and the extra superscript ~ is used to distinguish one of the collision partners from the other in the case where k and l are the same. In addition, n_k denotes the number density of the kth species and $\{ \ , \ \}_{kl}$ is a symmetric positive definite bilinear form which only involves species pair (k,l). We assume that the bracket operator is symmetric, positive semidefinite, and that its kernel is spanned by the collisional invariants. More specifically, we assume the following.

(i) Symmetry: $[\xi, \zeta] = [\zeta, \xi]$.

(ii) Positivity: $[\xi, \xi] \geq 0$ and $[\xi, \xi] = 0 \Longleftrightarrow \xi \in \mathcal{I}$.

As a consequence, the bracket bilinear form $[\ , \ ]$ is positive definite on the functional subspace associated with (2.5). Assuming that the right-hand side Ψ^μ is orthogonal to its kernel, i.e., $\langle\langle \Psi^\mu, \xi \rangle\rangle = 0$, $\xi \in \mathcal{I}$, the integral equation system (2.4) and (2.5) is generally well posed [2,7].

C. Transport fluxes and transport coefficients

In the first-order Enskog expansion, the macroscopic conservation equations are written in terms of transport fluxes. These transport fluxes are denoted by J^μ and may be written, up to a multiplicative factor, as

$$J^\mu = \langle\langle \Psi^\mu, \phi \rangle\rangle \ . \tag{2.10}$$

Using the expansion $\phi = \sum_\mu \phi^\mu \odot X^\mu$ for ϕ, we can directly relate the transport fluxes to the macroscopic variable gradients

$$J^\mu = \sum_{\mu'} L^{\mu\mu'} \odot X^{\mu'} \ , \tag{2.11}$$

where the tensor elements $L^{\mu\mu'}$ are the transport coefficients. In this study we only consider isotropic gas mixtures so that the transport coefficient $L^{\mu\mu'}$ is nonzero only if J^μ and $X^{\mu'}$ have the same tensorial rank.

The transport coefficients are directly related to the bracket operator introduced in Sec. II B since we have

$$L^{\mu\mu'}=[\phi^\mu,\phi^{\mu'}] \ . \tag{2.12}$$

To complete the discussion it is interesting to evaluate the entropy production σ for the near-equilibrium gas mixture. We indeed have

$$\sigma=-\sum_{\mu} J^\mu \odot X^\mu=-\sum_{\mu,\mu'} L^{\mu\mu'}X^\mu \odot X^{\mu'} \ , \tag{2.13}$$

from which positivity properties of the various transport coefficients are readily obtained.

D. Matrix approximations for the inverse collision operator

The linear integral Eqs. (2.4) and (2.5) are solved approximately by using a variational procedure. More specifically, a finite-dimensional functional space $\mathcal{A}$ is first selected,

$$\mathcal{A}=\operatorname{span}\{\xi^{rk},(r,k)\in\mathcal{B}\} \ , \tag{2.14}$$

where ξ^{rk}, $(r,k)\in\mathcal{B}$, are basis functions and where $\mathcal{B}$ is a set of basis function indices. The set of basis function indices is such that $\mathcal{B}\subset\mathcal{I}\times\mathcal{S}$ where $\mathcal{I}$ denotes a set of function type indices. We denote by ω the dimension of the functional space $\mathcal{A}$.

The species perturbed distribution functions ϕ^μ are next expanded in the form

$$\phi^\mu=\sum_{(r,k)\in\mathcal{B}} \alpha_k^r \xi^{rk} \ , \tag{2.15}$$

where the α_k^r are scalars. A classical Galerkin approach is used by requiring that the difference between the approximated $\mathfrak{X}(\phi^\mu)$ and Ψ^μ be orthogonal to the variational space $\mathcal{A}$. With this approach, the scalars α_k^r are then shown to be the solution of constrained linear systems which will be written in compact form in Sec. III.

III. MATHEMATICAL STRUCTURE OF THE TRANSPORT LINEAR SYSTEMS

A. Compact notation

The components with respect to the basis ξ^{rk}, $(r,k)\in\mathcal{B}$, of the functions $\xi=\sum_{(r,k)\in\mathcal{B}}x_k^r\xi^{rk}$ of $\mathcal{A}$ now form a vector of $\mathbb{R}^\omega$ denoted by $x=(x_k^r)_{(r,k)\in\mathcal{B}}$. Ordering the set $\mathcal{B}$ with the lexicographical order, the components of any vector $x\in\mathbb{R}^\omega$ are correspondingly denoted by $x=(x_k^r)_{(r,k)\in\mathcal{B}}$, thereby identifying $\mathbb{R}^\omega$ and $\mathbb{R}^{\mathcal{B}}$. The set $\mathcal{B}$ can then be used as a natural indexing set. For $x,y\in\mathbb{R}^\omega$, the scalar product $\langle x,y\rangle$ is given by $\langle x,y\rangle=\sum_{(r,k)\in\mathcal{B}}x_k^r y_k^r$. For $G\in\mathbb{R}^{\omega,\omega}$, we write $G=(G_{kl}^{rs})_{(r,k),(s,l)\in\mathcal{B}}$ the coefficients of the matrix G. The nullspace and range of the matrix G are denoted by $N(G)$ and $R(G)$, respectively.

Making use of the new formalism, the expansion coefficients α_k^r, $(r,k)\in\mathcal{B}$, form a vector $\alpha=(\alpha_k^r)_{(r,k)\in\mathcal{B}}\in\mathbb{R}^\omega$ which is shown to be the solution of a constrained linear system in the form

$$\begin{aligned} &G\alpha=\beta \ , \\ &\alpha\in\mathcal{C} \ , \end{aligned} \tag{3.1}$$

where we have defined

$$G_{kl}^{rs}=[\xi^{rk},\xi^{sl}], \quad \beta_k^r=\langle\langle \xi^{rk},\Psi^\mu\rangle\rangle \ . \tag{3.2}$$

In addition, we have introduced the constraint space $\mathcal{C}$ given by

$$\mathcal{C}=(\operatorname{span}\{\mathcal{G}^{l\nu},(l,\nu)\in[1,n+4]\times[1,\tau]\})^\perp \ , \tag{3.3}$$

where the constraint vectors have components given by

$$\begin{aligned} &\mathcal{G}_k^{rl\nu}=\langle\langle \xi^{rk},\mathcal{T}_\nu\hat{\psi}^l\rangle\rangle, \quad (l,\nu)\in[1,n+4]\times[1,\tau], \\ &(r,k)\in\mathcal{B} \ . \end{aligned} \tag{3.4}$$

Finally, the transport coefficients are evaluated from the products $[\phi^\mu,\phi^{\mu'}]$ in the form

$$[\phi^\mu,\phi^{\mu'}]=\langle\langle \phi^\mu,\Psi^{\mu'}\rangle\rangle=\langle\alpha,\beta'\rangle \ , \tag{3.5}$$

where the vector β' has components $\beta_k'^r=\langle\langle \xi^{rk},\Psi^{\mu'}\rangle\rangle$ for $(r,k)\in\mathcal{B}$.

B. Properties of G and B

The symmetry of the matrix $G\in\mathbb{R}^{\omega,\omega}$ is directly deduced from the symmetry of the bracket operator since

$$G_{kl}^{rs}=[\xi^{rk},\xi^{sl}]=[\xi^{sl},\xi^{rk}]=G_{lk}^{sr} \ . \tag{3.6}$$

Furthermore, G is positive semidefinite on $\mathbb{R}^\omega$ and positive definite on the constraint space $\mathcal{C}$. Indeed let now $x\in\mathbb{R}^\omega$, $x=(x_k^r)_{(r,k)\in\mathcal{B}}$, let ξ be its associated function $\xi=\sum_{(r,k)\in\mathcal{B}}x_k^r\xi^{rk}$, and consider the scalar product $\langle Gx,x\rangle$. We have

$$\begin{aligned} \langle Gx,x\rangle &= \sum_{\substack{(r,k)\in\mathcal{B}\\(s,l)\in\mathcal{B}}} G_{kl}^{rs}x_k^r x_l^s \\ &= \sum_{\substack{(r,k)\in\mathcal{B}\\(s,l)\in\mathcal{B}}} [\xi^{rk},\xi^{sl}]x_k^r x_l^s=[\xi,\xi] \ , \end{aligned} \tag{3.7}$$

which is non-negative so that G is positive semidefinite. Moreover, we have $\langle Gx,x\rangle=0$ if and only if ξ is a collisional invariant, so that the nullspace of G is spanned by the components in the basis ξ^{rk}, $(r,k)\in\mathcal{B}$, of the collisional invariants that are in $\mathcal{A}$

$$N(G)=\left\{x\in\mathbb{R}^\omega, \quad \sum_{(r,k)\in\mathcal{B}} x_k^r\xi^{rk}\in\mathcal{I}\cap\mathcal{A}\right\} \ . \tag{3.8}$$

From (3.8) it is then straightforward to establish that

$$N(G)\cap\mathcal{C}=\{0\} \ , \tag{3.9}$$

which shows that G is positive definite on $\mathcal{C}$.

Finally, the right member β is in the range of the matrix G. Indeed, from the property $\langle\!\langle \Psi^{\mu}, \mathcal{T}_{\nu}\hat{\psi}^{l} \rangle\!\rangle = 0$, $(l,\nu) \in [1, n+4] \times [1, \tau]$, it is easily shown that $\langle x, \beta \rangle = 0$ for $x \in N(G)$. Thus, $\beta \in N(G)^{\perp}$ and hence $\beta \in R(G)$ since G is symmetric.

C. Properties of the constraint space $\mathcal{C}$

It is well known that the constrained linear system (3.1) is well posed, i.e., admits a unique solution α for any $\beta \in R(G)$, if and only if the nullspace of G and the constraint space $\mathcal{C}$ are complementary spaces

$$N(G) \oplus \mathcal{C} = \mathbb{R}^{\omega} \; . \tag{3.10}$$

As a consequence, the property (3.10) will be termed "the well posedness condition."

We now establish that the well posedness condition (3.10) holds if and only if the space $\mathcal{I}$ is perpendicular to $\mathcal{A}$

$$\mathcal{I} = \mathcal{I} \cap \mathcal{A} \oplus \mathcal{I} \cap \mathcal{A}^{\perp} \; , \tag{3.11}$$

where $\mathcal{I} \cap \mathcal{A}^{\perp}$ denotes the elements of $\mathcal{I}$ that are orthogonal to $\mathcal{A}$ with respect to the bilinear form $\langle\!\langle \; , \; \rangle\!\rangle$

$$\mathcal{I} \cap \mathcal{A}^{\perp} = \{ \xi \in \mathcal{I}, \; \forall \zeta \in \mathcal{A} \;\; \langle\!\langle \xi, \zeta \rangle\!\rangle = 0 \} \; . \tag{3.12}$$

The property (3.11) will be termed "the perpendicularity property."

For brevity, we only establish here that the well posedness condition (3.10) is a consequence of the perpendicularity property (3.11). The converse property is proved as in [3]. For $x = (x_k^r)_{(r,k) \in \mathcal{B}} \in \mathbb{R}^{\omega}$ and $\xi = \sum_{(r,k) \in \mathcal{B}} x_k^r \xi^{rk}$, we first note that the perpendicularity property implies that

$$x \in \mathcal{C} \Longleftrightarrow \forall \zeta \in \mathcal{I} \cap \mathcal{A}, \quad \langle\!\langle \zeta, \xi \rangle\!\rangle = 0 \; . \tag{3.13}$$

Therefore, $\mathcal{C}$ has the same dimension as the subspace of $\mathcal{A}$ which is orthogonal to $\mathcal{I} \cap \mathcal{A}$ with respect to the positive definite quadratic form $\langle\!\langle \; , \; \rangle\!\rangle$, so that

$$\dim(\mathcal{C}) = \omega - \dim(\mathcal{I} \cap \mathcal{A}) \; . \tag{3.14}$$

On the other hand, from (3.8), we already know that the spaces $N(G)$ and $\mathcal{I} \cap \mathcal{A}$ have the same dimension, so that $\dim[N(G)] + \dim(\mathcal{C}) = \omega$. Finally, we have $N(G) \cap \mathcal{C} = \{0\}$ from (3.9) so that $N(G) \oplus \mathcal{C} = \mathbb{R}^{\omega}$.

D. The sparse transport matrix $d_b(G)$ and the species localization property

We now introduce the basic block decomposition of the matrix $G = (G_{kl}^{rs})_{(r,k),(s,l) \in \mathcal{B}}$ in $\mathbb{R}^{\omega,\omega}$. For convenience, we define the subset $\mathcal{S}_r$ as the set of species indices for which the rth function type is selected, and we denote by ω_r the number of elements of $\mathcal{S}_r$. Consider now a pair of function type indices $r,s \in \mathcal{T}$ and the associated block $G^{rs} = (G_{kl}^{rs})_{k \in \mathcal{S}_r, l \in \mathcal{S}_s}$ of size $\omega_r \times \omega_s$. Then the matrix G can be partitioned into the blocks $G = (G^{rs})_{r,s \in \mathcal{T}}$. We denote by $d_b(G) \in \mathbb{R}^{\omega,\omega}$ the matrix formed by the diagonals of all the rectangular blocks G^{rs}, $r,s \in \mathcal{T}$, of G, so that

$$d_b(G)_{kl}^{rs} = G_{kl}^{rs} \delta_{kl}, \quad (r,k),(s,l) \in \mathcal{B} \; . \tag{3.15}$$

The sparse transport matrix $d_b(G)$ plays a fundamental role in the asymptotic expansion of the transport coefficients. More specifically, we want to know whether the matrices $d_b(G)$ and $2d_b(G) - G$ are positive definite on $\mathbb{R}^{\omega}$. Under sufficient conditions, we establish that $2d_b(G) - G$ is positive definite when $n \geq 3$ and we identify its nullspace when $n = 2$ and $n = 1$. Similarly, we establish that $d_b(G)$ is positive definite when $n \geq 2$ and we identify its nullspace when $n = 1$. In the following sections we only consider the case $n \geq 2$ since for $n = 1$ we have $d_b(G) = G$ and $2d_b(G) - G = G$.

Since we are now interested in some specific coefficients of the matrix G, that is to say, in the sparse transport matrix $d_b(G)$ and not in the full matrix G, the properties of $d_b(G)$ and $2d_b(G) - G$ will depend on the particular choice of the basis functions ξ^{rk}, $(r,k) \in \mathcal{B}$. Indeed, we now assume that the basis functions are "localized with respect to the species"

$$\xi_l^{rk} = 0 \;\; \text{for} \;\; l \neq k \; , \tag{3.16}$$

so that $\xi^{rk} = (\xi_k^{rk} \delta_{kl})_{l \in \mathcal{S}}$. Under the assumption (3.16), we first establish that the matrix $2d_b(G) - G$ is positive semidefinite and we characterize its kernel.

E. Positive semidefiniteness of $2d_b(G) - B$ and characterization of $N(2d_b(G) - G)$

After some algebra, we get that

$$\langle (2d_b(G) - G)x, x \rangle = \sum_{\substack{k,l \in \mathcal{S} \\ k \neq l}} \tfrac{1}{4} n_k n_l \{ \xi_k - \xi_k' - \tilde{\xi}_l + \tilde{\xi}_l', \xi_k - \xi_k' - \tilde{\xi}_l + \tilde{\xi}_l' \}_{kl} + \sum_{k \in \mathcal{S}} \tfrac{1}{4} n_k^2 \{ \xi_k - \xi_k' + \tilde{\xi}_k - \tilde{\xi}_k', \xi_k - \xi_k' + \tilde{\xi}_k - \tilde{\xi}_k' \}_{kk} . \tag{3.17}$$

This is a sum of non-negative terms, so that the symmetric matrix $2d_b(G) - G$ is positive semidefinite on $\mathbb{R}^{\omega}$. It is interesting to point out that in (3.17) the terms in the simple sum indexed by $k \in \mathcal{S}$ are equal to the corresponding term in the expression of $\langle Gx, x \rangle$ whereas the terms in the double sum indexed by $k,l \in \mathcal{S}$, $k \neq l$, differ in the sign of the contributions from the collision partner l.

Keeping in mind that the number densities are positive, we deduce from (3.17) that $x \in N(2d_b(G) - G)$ if and only if for all species pair (k,l), with $k \neq l$, the subfamily

$(\xi_k,-\xi_l)$ is a collisional invariant of the binary submixture (k,l). Denoting by $\mathcal{I}_{kl}$ the space of collisional invariants of the submixture (k,l), we have thus established that for $n\geq 2$

$$N(2d_b(G)-G)=\{x\in\mathbb{R}^\omega,\ \forall k,l\in\mathcal{S},\ k\neq l,\ (\xi_k,-\xi_l)\in\mathcal{I}_{kl}\}\ . \tag{3.18}$$

As a comparison it is interesting to note that for $n\geq 2$ we also have

$$N(G)=\{x\in\mathbb{R}^\omega,\ \forall k,l\in\mathcal{S},k\neq l,\ (\xi_k,\xi_l)\in\mathcal{I}_{kl}\}\ . \tag{3.19}$$

The space $\mathcal{I}_{kl}$ is spanned by the functions

$$\mathcal{I}_{kl}=\mathrm{span}\{\mathcal{T}_\nu\hat{\psi}^m_{kl},\ (m,\nu)\in(\{k,l\}\cup[n+1,n+4])\times[1,\tau]\}\ , \tag{3.20}$$

where $\hat{\psi}^m_{kl}$, $m\in\{k,l\}\cup[n+1,n+4]$, are the scalar collisional invariants of the submixture (k,l). These collisional invariants are related to the full mixture collisional invariants $\hat{\psi}^m$, $m\in\{k,l\}\cup[n+1,n+4]$, since we have

$$\hat{\psi}^m_{kl}=(\hat{\psi}^m_k,\hat{\psi}^m_l),\quad m\in\{k,l\}\cup[n+1,n+4]\ , \tag{3.21}$$

by definition of collisional invariants.

F. The species orthogonality property and the nullspace $N(2d_b(G)-G)$

We now identify the nullspace $N(2d_b(G)-G)$ by using (3.18). This is done by assuming that the perpendicularity property (3.11) holds, that the species localization property (3.16) holds, and that the variational space $\mathcal{A}$ is orthogonal to the constant functions. In other words, we assume that the species collisional invariants $\mathcal{T}_\nu\hat{\psi}^l$, $l\in\mathcal{S}$ and $\nu\in[1,\tau]$, are in $\mathcal{I}\cap\mathcal{A}^\perp$, that is,

$$\mathcal{T}_\nu\hat{\psi}^l\in\mathcal{I}\cap\mathcal{A}^\perp\ \text{ for } (l,\nu)\in\mathcal{S}\times[1,\tau]\ . \tag{3.22}$$

This property will be termed "the species orthogonality property."

Assume first that $n\geq 3$, let $x=(x^r_k)_{(r,k)\in\mathcal{B}}\in\mathbb{R}^\omega$ and $\xi=\sum_{(r,k)\in\mathcal{B}}x^r_k\xi^{rk}$, and assume that $\langle(2d_b(G)-G)x,x\rangle=0$. From (3.18), we know that for any given species pair (k,l) with $k\neq l$, there exist scalars $u_{j\nu}$ such that

$$(\xi_k,-\xi_l)=\sum_{\substack{j\in\{k,l\}\\ \nu\in[1,\tau]}}u_{j\nu}\mathcal{T}_\nu\hat{\psi}^j_{kl}+\sum_{\substack{j\in[n+1,n+4]\\ \nu\in[1,\tau]}}u_{j\nu}\mathcal{T}_\nu\hat{\psi}^j_{kl}\ . \tag{3.23}$$

Introducing the corresponding full mixture collisional invariant

$$\mathcal{U}=\sum_{\substack{j\in\{k,l\}\\ \nu\in[1,\tau]}}u_{j\nu}\mathcal{T}_\nu\hat{\psi}^j+\sum_{\substack{j\in[n+1,n+4]\\ \nu\in[1,\tau]}}u_{j\nu}\mathcal{T}_\nu\hat{\psi}^j\ , \tag{3.24}$$

and identifying the kth and lth components yields that $\xi_k=\mathcal{U}_k$ and $\xi_l=-\mathcal{U}_l$. From $\xi_k=\mathcal{U}_k$ and the species localization property (3.16), we obtain that $\langle\langle\mathcal{U},\mathcal{T}_\nu\hat{\psi}^k\rangle\rangle=\langle\langle\xi,\mathcal{T}_\nu\hat{\psi}^k\rangle\rangle$ and therefore that $\langle\langle\mathcal{U},\mathcal{T}_\nu\hat{\psi}^k\rangle\rangle=0$ thanks to $\xi\in\mathcal{A}$ and the species orthogonality property (3.22). Since $\langle\langle\mathcal{U},\mathcal{T}_\nu\hat{\psi}^k\rangle\rangle=u_{k\nu}\langle\langle\mathcal{T}_\nu\hat{\psi}^k,\mathcal{T}_\nu\hat{\psi}^k\rangle\rangle$, we deduce that $u_{k\nu}=0$, for $\nu\in[1,\tau]$. Similarly, one can show that $u_{l\nu}=0$, for $\nu\in[1,\tau]$, so that we have

$$\mathcal{U}=\sum_{\substack{j\in[n+1,n+4]\\ \nu\in[1,\tau]}}u_{j\nu}\mathcal{T}_\nu\hat{\psi}^j\ . \tag{3.25}$$

Since $n\geq 3$ we can consider k, l, and m such that $k\neq l$, $l\neq m$, and $m\neq k$. From the preceding analysis, we can first write for the species pair (k,l) that $\xi_k=\mathcal{U}_k$ and $\xi_l=-\mathcal{U}_l$ where $\mathcal{U}$ is a collisional invariant of the full mixture. Similarly, for the species pair (l,m), we can then write that $\xi_l=\mathcal{V}_l$ and $\xi_m=-\mathcal{V}_m$ where $\mathcal{V}$ is another collisional invariant of the full mixture in the form

$$\mathcal{V}=\sum_{\substack{j\in[n+1,n+4]\\ \nu\in[1,\tau]}}v_{j\nu}\mathcal{T}_\nu\hat{\psi}^j\ . \tag{3.26}$$

We now point out that for $l\in\mathcal{S}$, the scalar functions $\mathcal{T}_\nu\hat{\psi}^j_l$, $(j,\nu)\in[n+1,n+4]\times[1,\tau]$, are associated with either momentum or energy conservation and are thus linearly independent. From $\mathcal{U}_l=-\xi_l=-\mathcal{V}_l$ we then deduce that $u_{j\nu}=-v_{j\nu}$, $(j,\nu)\in[n+1,n+4]\times[1,\tau]$, so that we indeed have $\mathcal{U}=-\mathcal{V}$. By repeating the argument for the species pair (m,k), we finally obtain that $\xi_k=\mathcal{U}_k=-\mathcal{U}_k=0$. This now implies that $\xi_k=0$ for any $k\in\mathcal{S}$, so that $x=0$ and $2d_b(G)-G$ is positive definite if $n\geq 3$.

In the special case $n=2$, it is not possible to change signs an odd number of times. In this case we have $x\in N(2d_b(G)-G)$ if and only if $x^*\in N(G)$, where we have defined

$$x^{*r}_k=(-1)^k x^r_k,\quad (r,k)\in\mathcal{B}\ , \tag{3.27}$$

for $k\in\mathcal{S}=\{1,2\}$.

G. Summary of the properties of G and of the sparse transport matrix $d_b(G)$

In Secs. III B and III C, we have established that whenever the perpendicularity property (3.11) holds, then the following are true.

(a) G is symmetric positive semidefinite.

(b) $N(G)=\{x\in\mathbb{R}^\omega,\ \sum_{(r,k)\in\mathcal{B}}x^r_k\xi^{rk}\in\mathcal{I}\cap\mathcal{A}\}$.

(c) G is positive definite on $\mathcal{C}$.

(d) $N(G)\oplus\mathcal{C}=\mathbb{R}^\omega$.

(e) $\beta\in R(G)$.

In Secs. III D–III F, we have shown that whenever the perpendicularity property (3.11), the species localization property (3.16), and the species orthogonality property (3.22) hold, then we have the following.

(f) The matrix $2d_b(G)-G$ is symmetric positive semidefinite for $n\geq 1$, (i) $2d_b(G)-G$ is positive definite for $n \geq 3$, (ii) $N(2d_b(G)-G)=\{x^*, x \in N(G)\}$ for $n=2$, (iii) $N(2d_b(G)-G)=N(G)$ for $n=1$, where x^* is defined in (3.27).

Finally the properties of the sparse transport matrix $d_b(G)$ result from $d_b(G)=\frac{1}{2}(2d_b(G)-G)+\frac{1}{2}G$. After some algebra, it is then possible to establish that whenever the perpendicularity property (3.11), the species localization property (3.16), and the species orthogonality property (3.22) hold, then we have the following.

(g) The matrix $d_b(G)$ is symmetric positive semidefinite for $n \geq 1$, (i) $d_b(G)$ is positive definite for $n \geq 2$, (ii) $N(d_b(G))=N(G)$ for $n=1$.

Remark. The structure of the matrix $2d_b(G)-G$ reveals that the general case for mixtures is $n \geq 3$ and that binary mixtures are a degenerate case.

H. Asymptotic expansions for the transport coefficients

In order to obtain asymptotic expansions for the transport coefficients, we use the theory of projective iterative methods for constrained singular linear systems. Several mathematical results have been recently derived in the framework of multicomponent transport [3,8]. The main result of interest here is the following theorem.

Asymptotic expansion. Let $G \in \mathbb{R}^{\omega,\omega}$ be a singular symmetric positive semidefinite matrix and let $\mathscr{C}$ be a subspace complementary to $N(G)$, i.e., $N(G) \oplus \mathscr{C} = \mathbb{R}^{\omega}$. Consider a symmetric matrix M such that the matrix $2M-G$ is positive definite, and define $T=I-M^{-1}G$. Let P be the oblique projector onto the subspace $\mathscr{C}$ along $N(G)$. Let also $\beta \in R(G)$, $x_0 \in \mathbb{R}^{\omega}$, $y_0 = Px_0$, and consider for $i \geq 0$ the iterates $x_{i+1}=Tx_i+M^{-1}\beta$ and $y_{i+1}=PTy_i+PM^{-1}\beta$. Then $y_i=Px_i$ for all $i \geq 0$, the matrices T and PT are convergent, and we have the following limits:

$$\lim_{i\to\infty} y_i = P(\lim_{i\to\infty} x_i) = \alpha \ , \tag{3.28}$$

where α is the unique solution of (3.1). Furthermore, upon defining the approximate transport coefficients

$$L_{[i]}^{\mu\mu'} = \left\langle \sum_{j=0}^{i} (PT)^j PM^{-1}P^t\beta, \beta' \right\rangle \ , \tag{3.29}$$

we have

$$\lim_{i\to\infty} L_{[i]}^{\mu\mu'} = \left\langle \sum_{j=0}^{\infty} (PT)^j PM^{-1}P^t\beta, \beta' \right\rangle = L^{\mu\mu'} \ . \tag{3.30}$$

With the results of Sec. III G, it is clear that an appropriate choice for the splitting matrix M is the sparse transport matrix $d_b(G)$ in the general case $n \geq 3$, whereas in the particular cases $n=1$ or 2, the main diagonal of $db(G)$ needs to be weighted by some positive coefficients. We also point out that the projector matrix P in (3.29) ensures that at each iteration the approximation to the species perturbed distribution functions satisfies the physical constraints.

Finally, we note that another approach for expanding the transport coefficients is to consider the conjugate gradient algorithm. This algorithm converges in a finite number of steps. Numerical results on practical problems also show that high convergence rates are obtained when this algorithm is preconditioned by the sparse transport matrix $d_b(G)$. Details of the algorithm with application to multicomponent transport are given in [3,8] and are omitted here.

IV. APPLICATION TO MULTICOMPONENT TRANSPORT

In the framework of either the semiclassical, the classical, or the quantum mechanical isotropic kinetic theory, the resulting bracket operator satisfies the symmetry and positivity properties discussed in Sec. II B. The perturbed distribution functions $\phi=(\phi_k)_{k\in\mathscr{S}}$ are expanded in the form

$$\phi_k = -\phi_k^{\eta}:\nabla v - \phi_k^{\kappa}(1/3)\nabla\cdot v - \sum_{l\in\mathscr{S}} \phi_k^{D_l}\cdot(\nabla\bar{p}_l - \rho Y_l b_l) - \phi_k^{\lambda'}\cdot\nabla(1/k_B\bar{T}) \ , \tag{4.1}$$

where ϕ_k^{η} is a traceless symmetric matrix function, ϕ_k^{κ} a scalar function, and $\phi_k^{D_l}$ and $\phi_k^{\lambda'}$ are vector functions. In addition, v is the macroscopic mass averaged velocity of the mixture, $\bar{p}_l$ the partial pressure of the lth species, Y_l its mass fraction, b_l the external force acting on the lth species, ρ the density, k_B the Boltzmann constant, and $\bar{T}$ the temperature.

To the first approximation in the Enskog expansion, the transport fluxes are the pressure tensor Π, the species diffusion velocities V_k, and the heat flux vector q given by

$$\Pi = \bar{p}I - (\kappa - \tfrac{2}{3}\eta)(\nabla\cdot v)I - \eta[\nabla v + (\nabla v)^t] \ , \tag{4.2}$$

$$V_k = -\sum_{l\in\mathscr{S}} D_{kl}d_l - \theta_k \nabla \ln\bar{T}, \quad k \in \mathscr{S} \ , \tag{4.3}$$

$$q = \sum_{k\in\mathscr{S}} \rho h_k Y_k V_k - \lambda'\nabla\bar{T} - \sum_{k\in\mathscr{S}} \theta_k d_k \ , \tag{4.4}$$

where $\bar{p}$ is the thermodynamic pressure, I the identity matrix, κ the volume viscosity—also termed bulk viscosity— η the shear viscosity, $D=(D_{kl})_{k,l\in\mathscr{S}}$ the diffusion matrix, d_k the diffusion driving force for the kth species, $\theta=(\theta_k)_{k\in\mathscr{S}}$ the thermal diffusion vector, h_k the enthalpy per unit mass of the kth species, and λ' the partial thermal conductivity. The diffusion driving forces are given by

$$d_k = \nabla X_k + (X_k - Y_k)\frac{\nabla\bar{p}}{\bar{p}} + \frac{\rho}{\bar{p}}\sum_{l\in\mathscr{S}} Y_k Y_l (b_l - b_k), \quad k \in \mathscr{S} \ , \tag{4.5}$$

where X_k is the mole fraction of the kth species. Furthermore, the following expressions are obtained for the transport coefficients:

$$\eta = \frac{k_B\bar{T}}{10}[\phi^{\eta},\phi^{\eta}], \quad \kappa = \frac{k_B\bar{T}}{9}[\phi^{\kappa},\phi^{\kappa}] \ , \tag{4.6}$$

$$D_{kl}=\frac{\bar{p}k_B\bar{T}}{3}[\phi^{D_k},\phi^{D_l}],\quad k,l\in\mathcal{S}\ ,$$

$$\lambda'=\frac{1}{3k_B\bar{T}^2}[\phi^{\lambda'},\phi^{\lambda'}]\ , \tag{4.7}$$

$$\theta_k=-\tfrac{1}{3}[\phi^{D_k},\phi^{\lambda'}],\quad k\in\mathcal{S}\ . \tag{4.8}$$

By construction, the diffusion matrix is symmetric and we also have the mass conservation constraints $\sum_{k\in\mathcal{S}}Y_kD_{kl}=0$, $l\in\mathcal{S}$, and $\sum_{k\in\mathcal{S}}Y_k\theta_k=0$. It is worthwhile to point out that the above results can be generalized to the case of reactive mixtures in the "tempered reaction regime," where the chemical characteristic times are larger by an order of magnitude than the mean free time of the molecules. We refer to [3] for a more detailed discussion.

Matrix approximations for the inverse collision operator are obtained by considering expansion functions ξ^{rk}, $(r,k)\in\mathcal{B}$. In the isotropic classical case for linear molecules, the expansion functions are generally chosen as simple linear combinations of the functions ϕ^{a0cdk} [5,9] defined by

$$\phi^{a0cdk}(w_k,J_k)=\{\mathcal{L}^c_{a+1/2}(w_k\cdot w_k)\mathcal{L}^d_0(J_k^2)[\oplus^a w_k]\delta_{kl}\}_{l\in\mathcal{S}}\ . \tag{4.9}$$

Here, a, c, and d are integers, w_k denotes the reduced velocity of the particles of the kth species, and J_k their reduced angular momentum. Furthermore, $\mathcal{L}^p_q$ is the associated Laguerre polynomial of order p with parameter q and $[\otimes^a w_k]$ a tensor of rank a with respect to the three-dimensional space [1].

In the semiclassical case [1,4,5], the expansion functions are generally chosen as simple linear combinations of the functions ϕ^{a0cdk} defined by

$$\phi^{a0cdk}(w_k,\epsilon_{kK})=\{\mathcal{L}^c_{a+1/2}(w_k\cdot w_k)\times W^d_k(\epsilon_{kK})[\otimes^a w_k]\delta_{kl}\}_{l\in\mathcal{S}}\ , \tag{4.10}$$

where W^d_k is the Wang Chang and Uhlenbeck polynomial of order d for the kth species and ϵ_{kK} the reduced internal energy of the kth species in the Kth internal energy state. In the isotropic quantum mechanical case, the expansion functions are given, for instance, in [5] and are omitted for brevity.

In the following sections we verify that the perpendicularity property and the species orthogonality property are satisfied with usual choices for the approximation space $\mathcal{A}\subset\mathrm{span}\{\phi^{a0cdk},\ (cd,k)\in\mathcal{B}\}$.

A. The scalar case

In the scalar case, we assume that the indexing set $\mathcal{B}$ does not contain the indices $(00,k)$ for $k\in\mathcal{S}$. One may then easily establish that $\mathcal{T}_\nu\hat{\psi}^l\in\mathcal{J}\cap\mathcal{A}^\perp$ for $(l,\nu)\in[1,n+3]\times\{1\}$ and that the energy collisional invariant satisfies

$$\hat{\psi}^{n+4}=\sum_{k\in\mathcal{S}}\phi^{0010k}+\sum_{k\in\mathcal{P}}\phi^{0001k}\ , \tag{4.11}$$

where $\mathcal{P}$ denotes the set of species with internal energy. Assuming that $\hat{\psi}^{n+4}\in\mathcal{A}$, we obtain the decomposition

$$\mathcal{J}=\mathbb{R}\hat{\psi}^{n+4}\oplus\mathcal{J}\cap\mathcal{A}^\perp\ . \tag{4.12}$$

The perpendicularity property and the species orthogonality property are thus satisfied. As a consequence, if the basis functions of $\mathcal{A}$ are also localized with respect to the species, we conclude that the results of Sec. III apply.

Remark. First-order disequilibrium of several, mechanically independent, internal energy modes, e.g., rotation and vibration, can be treated by generalizing the semiclassical expansion functions (4.9). In this case it is possible to consider Wang Chang and Uhlenbeck polynomials in each of the internal energy modes. Denoting by $\mathfrak{E}$ the set of indices for the internal energy modes, an energy state of a particle is now referred to by a multiindex $K=(K^e)_{e\in\mathfrak{E}}$ with reduced internal energies $\epsilon^e_{kK^e}$. One can then verify that

$$\hat{\psi}^{n+4}=\sum_{k\in\mathcal{S}}\phi^{0010\hat{0}k}+\sum_{e\in\mathfrak{E}}\sum_{k\in\mathcal{P}}\phi^{000\hat{e}k}\ , \tag{4.13}$$

with $\phi^{0010\hat{0}k}=\{\mathcal{L}^1_{1/2}(w_k\cdot w_k)\delta_{kl}\}_{l\in\mathcal{S}}$ and $\phi^{100\hat{e}k}=\{W^1_k(\epsilon^e_{kK^e})\delta_{kl}\}_{l\in\mathcal{S}}$.

B. The vector case

In the vector case, one may easily establish that we have $\mathcal{T}_\nu\hat{\psi}^l\in\mathcal{J}\cap\mathcal{A}^\perp$ for $(l,\nu)\in([1,n]\cup\{n+4\})\times[1,3]$ and $(l,\nu)=(n+\nu',\nu)$, with $\nu,\nu'\in[1,3]$ and $\nu\neq\nu'$. Moreover, by isotropy we also have $\mathcal{T}_1\hat{\psi}^{n+1}-\mathcal{T}_2\hat{\psi}^{n+2}\in\mathcal{J}\cap\mathcal{A}^\perp$ and $\mathcal{T}_2\hat{\psi}^{n+2}-\mathcal{T}_3\hat{\psi}^{n+3}\in\mathcal{J}\cap\mathcal{A}^\perp$. In addition, one can easily verify that

$$\mathcal{T}_1\hat{\psi}^{n+1}+\mathcal{T}_2\hat{\psi}^{n+2}+\mathcal{T}_3\hat{\psi}^{n+3}=\sum_{k\in\mathcal{S}}(2k_B\bar{T}m_k)^{1/2}\phi^{1000k}\ , \tag{4.14}$$

where m_k is the mass of the particles of the kth species. Assuming then that $\mathcal{T}_1\hat{\psi}^{n+1}+\mathcal{T}_2\hat{\psi}^{n+2}+\mathcal{T}_3\hat{\psi}^{n+3}\in\mathcal{A}$, we obtain the decomposition

$$\mathcal{J}=\mathbb{R}(\mathcal{T}_1\hat{\psi}^{n+1}+\mathcal{T}_2\hat{\psi}^{n+2}+\mathcal{T}_3\hat{\psi}^{n+3})\oplus\mathcal{J}\cap\mathcal{A}^\perp\ . \tag{4.15}$$

The perpendicularity property and the species orthogonality property are thus satisfied. As a consequence, if the basis functions of $\mathcal{A}$ are also localized with respect to the species, we conclude that the results established in Sec. III apply.

Another interesting case arises when considering the thermal conductivity and the thermal diffusion ratios. These transport efficients can be defined directly within a variational framework independently of the partial thermal conductivity, the thermal diffusion vector, and the diffusion matrix [10]. In this case we have

$$\mathcal{J}=\mathcal{J}\cap\mathcal{A}^\perp\ . \tag{4.16}$$

C. The matrix case

In the matrix case, one may easily establish that $\mathcal{T}_\nu\hat{\psi}^l\in\mathcal{J}\cap\mathcal{A}^\perp$ for any $(l,\nu)\in[1,n+4]\times[1,9]$, so that

$$\mathcal{J}=\mathcal{J}\cap\mathcal{A}^{\perp}\,. \tag{4.17}$$

The perpendicularity property and the species orthogonality property are thus satisfied. As a consequence, if the basis functions of $\mathcal{A}$ are also localized with respect to the species we conclude that the results established in Sec. III apply.

V. CONCLUSIONS

In this paper we have investigated the structure of transport linear systems in dilute isotropic gas mixtures. In particular, we have introduced the sparse transport matrix $d_b(G)$. We have then shown from the kinetic theory that the sparse transport matrix satisfies a set of fundamental properties and that the transport coefficients can be expanded as convergent series. Finally, we have verified that the present theory can be applied to practical problems in either a classical, semiclassical or quantum mechanical framework.

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