Molecular to fluid dynamics: The consequences of stochastic molecular motion

Stefan Heinz*

Technische Universität München, Fachgebiet Strömungsmechanik, Boltzmannstrasse 15, D-85747 Garching, Germany

The derivation of fluid dynamic equations from molecular equations is considered. This is done on the basis of a stochastic model for the molecular motion which can be obtained by a projection of underlying deterministic equations. The stochastic model is used to derive fluid dynamic equations where the molecular stress tensor and heat flux appear as unknowns. However, the stochastic model also implies transport equations for these quantities. Combined with the assumption of a local equilibrium state, these transport equations can be used to derive a hierarchy of algebraic expressions for the molecular stress tensor and heat flux. A scaling analysis then explains the range of applicability of the Navier-Stokes model. The latter is relevant, for example, to simulations of high-Mach-number turbulent flows.

I. INTRODUCTION

A comprehensive understanding of the range of applicability of the Navier-Stokes equations is important with regard to a rigorous foundation of the theory of turbulence, and it is relevant to fundamental studies of the physics of turbulent flows. Such investigations are based, in general, on the direct solution of the Navier-Stokes equations (direct numerical simulation), but sufficient data for a (complete) assessment of simulation results are unavailable in general. One needs, therefore, strong theoretical arguments to support results obtained by direct numerical simulation. This is relevant, for example, to simulations of high-Mach-number flows which may involve shocks. For strong shocks, the presence of large gradients and the highly nonequilibrium state of the fluid suggest that the applicability of the Navier-Stokes model may become questionable [1]. The need to address the suitability of fluid dynamic equations also appears with regard to simulations of two-phase or viscoelastic flows where the Navier-Stokes model is known to fail in many cases [2].

The question of the validity of the Navier-Stokes equations can be considered in a variety of ways. A first way is given by the use of concepts of extended irreversible thermodynamics, where (in contrast to the simple flux-gradient relationships of ordinary irreversible thermodynamics [3]) transport equations for the molecular stress tensor and heat flux are constructed [2]. However, the problem of that approach is given by the fact that coefficients of corrections to the Navier-Stokes model cannot be obtained as a part of the construction of these equations. Thus, such equations are not unique so that their use cannot be seen as a general method for the assessment of the validity of the Navier-Stokes equations.

Another method is given by the use of equations for the molecular motion which then imply equations for the fluid and thermodynamics. Mostly, this is performed on the basis of the Boltzmann equation; see, for example, Cercignani [4], Bird [5], and Esposito *et al.* [6] and references therein. However, this approach is also faced with several questions. The first problem arises from the fact that such equations are related to the consideration of rarefied gases, whereas one is interested in dense fluids (gases and liquids) in fluid mechanics in general. The second problem arises from the fact that the derivation of the fluid dynamic equations in this way poses a nontrivial problem. By adopting this approach one may find a variety of equations, and the physical relevance of such extensions of the Navier-Stokes equations cannot be seen to be fully clarified until now [4–7].

Instead of using the Boltzmann equation [4] or its stochastic versions [5] for the derivation of equations for the fluid and thermodynamics, one may also apply stochastic diffusion models for the molecular motion. One way to address this problem is to assume that the position of a molecule represents a stochastic diffusion process [8]. However, this assumption is only valid for time steps that are large compared to a characteristic time scale for molecular velocity correlations (τ_m; see Sec. III); this means one applies a relatively crude model [9]. Hence, this approach also has several shortcomings. Macroscopic transport coefficients are not calculated from the molecular dynamics but they have to be introduced as external variables. Further, due to the fact that dynamics of velocity fluctuations are only asymptotically described, this analysis had to be limited to incompressible flows such that corrections to the Navier-Stokes model could not be obtained. A better way to develop stochastic models for the molecular motion is to assume that position and velocity of a molecule represent a stochastic diffusion process, which was pioneered by Kirkwood and co-workers [10–14]. Compared to molecular position models, the consideration of a velocity model has the significant advantage that macroscopic transport equations (coefficients) may be obtained as a consequence. Nevertheless, there are a couple of relevant questions related to the previous use of this approach. First, Kirkwood [10] introduced such a molecular velocity model as a heuristic model for liquids (with a fixed Prandtl number; see below). This leads to the question (i) of how it is possible to explain the range of applicability of this model (its applicability to flows with different

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*Present address: Department of Mathematics, University of Wyoming, 1000 E. University Ave., Laramie, WY 82071, USA. Electronic address: heinz@uwyo.edu

Prandtl numbers) on the basis of first principles. A second question is related to the fact that Kirkwood's model was applied previously only to recover the Navier-Stokes model [10–14]. There is, therefore, the question (ii) of how physically consistent extensions of the Navier-Stokes model can be obtained, which may be used to control its applicability and to extend it in order to simulate, for example, high-Mach-number flows. This is a nontrivial problem; see, e.g, the discussion of Cercignani [4] with regard to corresponding extensions on the basis of the Boltzmann equation.

Questions (i) and (ii) will be addressed here. The fluid dynamic variables will be defined in Sec. II. In Sec. III, a projection of the underlying deterministic equations for the molecular motion is used to obtain a stochastic molecular model. This results in a molecular acceleration model which generalizes Kirkwood's velocity model. However, the study of this model is relatively complex. Thus, the model analysis will be performed in two steps: the consequences of the implied simpler velocity model will be investigated first, and modifications of the resulting equations for the fluid and thermodynamics, which arise from the consideration of the more general acceleration model, will be presented in the Appendix. The transport equations for fluid dynamic variables which follow from the stochastic model considered are also presented in Sec. III. These equations are unclosed due to the appearance of the unknown molecular stress tensor and heat flux. However, the stochastic molecular model also implies transport equations for the latter quantities. Combined with the assumption of a local equilibrium state, these transport equations can be used for the calculation of the molecular stress tensor and heat flux (see Fig. 1 for an illustration). This will be shown in Sec. IV. The closure of the equations for the fluid and thermodynamics will be addressed in Sec. V on the basis of a scaling analysis. Section VI deals with conclusions of this analysis.

II. FLUID DYNAMICS VARIABLES

To prepare the derivation of equations for the fluid and thermodynamics in the following sections, it is convenient to define first the fluid dynamic variables.

A. Conditional ensemble means

Fluid dynamic variables can be defined by adopting conditional ensemble means. We define the mean of any function Q of molecular properties (e.g., velocities) conditioned on the position $\mathbf{x}$ in physical space by the expression

$$\bar{Q}(\mathbf{x},t) = \frac{1}{\rho(\mathbf{x},t)} \langle \rho^{(m)}(\mathbf{x},t)Q \rangle. \qquad (2.1)$$

The symbol $\langle \cdots \rangle$ denotes an ensemble mean. $\rho^{(m)}$ refers to the instantaneous molecular mass density,

$$\rho^{(m)}(\mathbf{x},t) = M\delta(\mathbf{x}*(t) - \mathbf{x}), \qquad (2.2)$$

and the mean mass density $\rho(\mathbf{x},t)$ is defined as the ensemble mean of $\rho^{(m)}$:
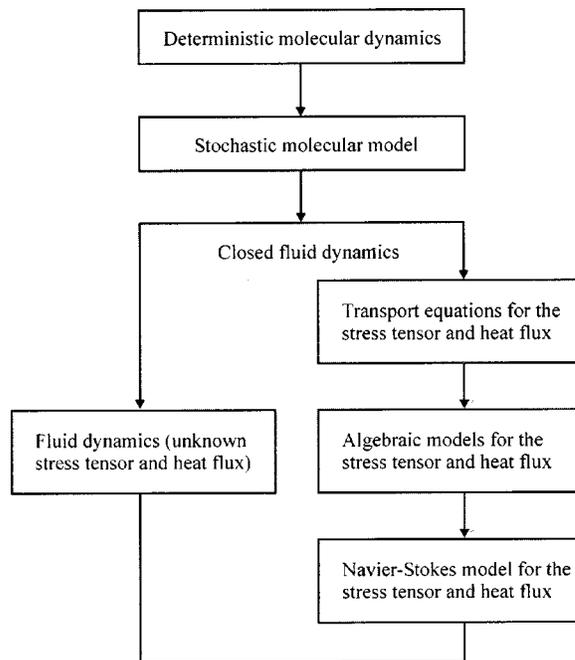
FIG. 1. An illustration of the derivation of fluid dynamic equations from molecular equations presented here. The projection operator technique can be used to reduce coupled deterministic equations for all the molecules to a stochastic acceleration (velocity) equation for one molecule. The stochastic model considered implies transport equations for the fluid mass density, velocity, and energy, where the molecular stress tensor and heat flux appear as unknowns. Additionally, the stochastic model implies transport equations for these quantities. Algebraic models for the molecular stress tensor and heat flux can be found by adopting the local equilibrium approximation. The Navier-Stokes model is then recovered if τ_m becomes small compared to the fluid dynamic time scale S^{-1}.

$$\rho(\mathbf{x},t) = \langle \rho^{(m)}(\mathbf{x},t) \rangle. \qquad (2.3)$$

In Eq. (2.2), $\delta(\cdots)$ is the delta function and $\mathbf{x}*(t)$ denotes the position of a molecule at time t. It is worth noting that $\rho^{(m)}$ (and therefore ρ) is not normalized to unity. Relation (2.2) shows that $\rho^{(m)}$ integrates to the total mass M of molecules within the domain considered:

$$M = \int d\mathbf{x} \rho^{(m)}(\mathbf{x},t). \qquad (2.4)$$

By invoking the ergodic theorem [15], the ensemble averaging considered here may be seen as a spatial filter procedure where the filter width is much smaller than the domain considered but such that a very large number of molecules is involved into the calculation of means at $\mathbf{x}$. Thus, such ensemble-averaged variables describe the properties of a continuum: they represent fluid dynamic variables as, for instance, the fluid mass density or velocity; see Sec. II C.

B. Conditioned velocity probability density function

The conditional mean (2.1) may also be represented as a mean of a probability density function (PDF). This relation reads

$$\bar{Q}(\mathbf{x},t) = \int d\mathbf{w} Q(\mathbf{w},\mathbf{x},t) F^{(m)}(\mathbf{w},\mathbf{x},t), \qquad (2.5)$$

where the conditional PDF of molecular velocities is given by

$$F^{(m)}(\mathbf{w},\mathbf{x},t) = \frac{1}{\rho(\mathbf{x},t)} \langle \rho^{(m)}(\mathbf{x},t) \delta(\mathbf{V}*(\mathbf{x}*(t),t) - \mathbf{w}) \rangle. \qquad (2.6)$$

Here, $\mathbf{V}*(\mathbf{x}*(t),t) = (V_1*, V_2*, V_3*)$ is the velocity of a molecule. The consistency of the definitions (2.1) and (2.5) for $\bar{Q}$ may be seen by inserting Eq. (2.6) into Eq. (2.5). The use of the shifting property of δ functions and integration over the velocity sample space then recovers relation (2.1).

The definition (2.6) represents a relation between Eulerian and Lagrangian variables: the Eulerian PDF $F^{(m)}$ at position $\mathbf{x}$ is defined in terms of the Lagrangian particle properties $\mathbf{x}*(t)$ and $\mathbf{V}*(t)$. This relationship will be used below to derive the fluid dynamics as a consequence of molecular motion: this means that a model for $\mathbf{x}*(t)$ and $\mathbf{V}*(t)$ will be used to calculate $F^{(m)}$. The knowledge of this PDF then enables the calculation of fluid dynamic variables. To see this we write Eq. (2.6) in the following way:

$$F^{(m)}(\mathbf{w},\mathbf{x},t) = \overline{\delta(\mathbf{V}(\mathbf{x},t) - \mathbf{w})}. \qquad (2.7)$$

Here, the Lagrangian particle velocity is written $\mathbf{V}*(\mathbf{x}*(t),t) = \mathbf{V}*(\mathbf{x},t) = \mathbf{V}(\mathbf{x},t)$. The replacement of $\mathbf{x}*(t)$ by $\mathbf{x}$ in $\mathbf{V}*$ is a consequence of the properties of δ functions. $\mathbf{V}$ is then used to denote the corresponding Eulerian velocity, which is nothing but the velocity of that molecule which has the property $\mathbf{x}*(t) = \mathbf{x}$ (we consider a continuum where one finds a molecule at each position). According to Eq. (2.7), all the fluid dynamic variables may be obtained from $F^{(m)}$ by multiplying this PDF with the corresponding variables and integration over the sample space. This may be seen in Sec. II C with regard to the fluid velocity and energy.

C. Fluid dynamic variables

Relation (2.7) will be used now to define fluid dynamic variables in addition to the mass density ρ that is defined by Eq. (2.3). The fluid velocity $U_i(i=1,3)$ is given by the mean velocity of molecules in an infinitesimal vicinity of $\mathbf{x}$:

$$U_i(\mathbf{x},t) = \overline{V_i(\mathbf{x},t)} = \int d\mathbf{w} w_i F^{(m)}(\mathbf{w},\mathbf{x},t). \qquad (2.8)$$

The deviations of $V_i(\mathbf{x},t)$ from their mean velocities $U_i(\mathbf{x},t)$ will be denoted by lowercase letters:

$$v_i(\mathbf{x},t) = V_i(\mathbf{x},t) - \overline{V_i(\mathbf{x},t)}. \qquad (2.9)$$

These fluctuations v_i vanish in the mass-weighted mean, $\overline{v_i}(\mathbf{x},t) = 0$, but they do not vanish in the ensemble mean in general, $\langle v_i \rangle \neq 0$. The intensity of fluctuations v_i is characterized by the specific (normalized by mass) kinetic energy e, which is defined by

$$e(\mathbf{x},t) = \frac{1}{2}\overline{v_i v_i} = \frac{1}{2}\int d\mathbf{w}(w_i - U_i)(w_i - U_i) F^{(m)}(\mathbf{w},\mathbf{x},t). \qquad (2.10)$$

Further fluid dynamic variables will be considered below in conjunction with the consideration of transport equations for the calculation of ρ, U_i, and e.

III. FROM THE MOLECULAR TO FLUID DYNAMICS

A way to address the questions (i) and (ii) described in the Introduction is the use of a systematic procedure for the construction of stochastic equations. Such a methodology is given by the projection operator technique. The basic idea of this technique is to extract the dynamics of relevant variables from any (complete) deterministic dynamics. This results in contributions to the dynamics of relevant variables that are explicit deterministic functions of the relevant variables (which may involve memory effects) and remaining contributions that involve the influence of all the other variables. The latter terms have the properties of stochastic forces. The projection operator technique may be applied in various variants: see, for instance, Grabert [16], Lindenberg and West [17], and Zubarev *et al.* [18,19]. One way is to derive a PDF transport equation for relevant variables, which has the structure of a generalized Fokker-Planck equation. Another way is to derive equations for the instantaneous dynamics of relevant variables [20]. This approach will be used here.

A. Stochastic molecular models

We will restrict the attention to the case of monatomic fluids which do not have internal degrees of freedom (rotational or vibrational energy). Thus, the state of each molecule is completely described by its position x_i*, velocity V_i*, and acceleration A_i*. The projection operator technique then provides the following equations if $(\mathbf{x}*, \mathbf{V}*, \mathbf{A}*)$ is considered as a Markov process [20]:

$$\frac{dx_i*}{dt} = V_i*, \qquad (3.1a)$$

$$\frac{dV_i*}{dt} = A_i*, \qquad (3.1b)$$

$$\frac{dA_i*}{dt} = \frac{1}{\tau_a}\left\{ -A_i* - \frac{1}{\tau_m}(V_i* - U_i) + F_i^{(m)} + \sqrt{\frac{4e}{3\tau_m}}\frac{dW_i}{dt} \right\}. \qquad (3.1c)$$

The fluid velocity U_i, kinetic energy e, external force $F_i^{(m)}$ (which may be caused, for example, by gravity), and the characteristic time scales for molecular acceleration and velocity correlations, τ_a and τ_m, respectively, may depend on position and time. The relation between τ_m and intermolecular forces was described by Kirkwood [10], and the relation of τ_a and τ_m to fluid dynamic parameters will be considered below. dW_i/dt refers to a Gaussian process with vanishing

means and uncorrelated values at different times (δ_{ij} is the Kronecker delta):

$$\left\langle \frac{dW_i}{dt} \right\rangle = 0, \quad \left\langle \frac{dW_i}{dt}(t)\frac{dW_j}{dt'}(t') \right\rangle = \delta_{ij}\delta(t-t'). \quad (3.2)$$

The incorporation of the white-noise process dW_i/dt in the acceleration equation (3.1c) (which is a significantly weaker assumption than to apply such a term in a velocity model: see below) represents the most relevant assumption related to the construction of Eqs. (3.1). Compared to the application of the Boltzmann equation, the appearance of such a stochastic force may be seen as the effect of the interaction of long-range forces between the molecules, which produce a continuous sequence of small and almost stochastic velocity changes. Such an approach is, therefore, a valid concept for the modeling of a dense fluid. The molecules are assumed to move independently. This corresponds to the consideration of a perfect fluid.

Equations (3.1) can be applied to derive the equations of the fluid and thermodynamics. However, this results in relatively complex developments, as will be shown in the Appendix. To keep the explanation of essential steps simple we will do the following. First, we use a simplified version of Eqs. (3.1) to construct transport equations for fluid dynamic variables. A corresponding analysis of Eqs. (3.1) will be performed in the Appendix, which will be used to discuss the modifications of fluid dynamic equations, which arise from the consideration of the more general acceleration model. Correspondingly, we now assume that τ_a is small, which corresponds [with regard to the discrete version of Eq. (3.1c)] to the consideration of time steps $\Delta t \gg \tau_a$. In this case, Eqs. (3.1) reduce to

$$\frac{dx_i^*}{dt} = V_i^*, \quad (3.3a)$$

$$\frac{dV_i^*}{dt} = -\frac{1}{\tau_m}(V_i^* - U_i) + F_i^{(m)} + \sqrt{\frac{4e}{3\tau_m}}\frac{dW_i}{dt}. \quad (3.3b)$$

The assumption of a small time scale τ_a of acceleration fluctuations is equivalent to the neglect of correlations of forces due to intermolecular interactions. This is a valid assumption if one is mainly interested in fluid dynamic motions which are independent of details of the intermolecular collision process. One obtains in this way a local model for velocities, which is sufficient to generalize the Navier-Stokes equations (see the following sections). Equations (3.3) are equivalent to the molecular velocity model that was suggested by Kirkwood. The difference is given by the fact that Kirkwood introduced this model as a heuristic model for liquids [10], whereas it is derived here as asymptotic limit of the acceleration model, Eqs. (3.1), which is a consequence of underlying deterministic molecular dynamics.

B. Unclosed fluid dynamic equations

The stochastic model (3.3) can be rewritten into an equivalent Fokker-Planck equation [21]. The definition (2.6) of $F^{(m)}$ reveals that $F^{(m)}\rho/M$ is the joint PDF of particle

positions and velocities. We multiply the transport equation for $F^{(m)}\rho/M$, which is implied by Eqs. (3.3), with M to obtain

$$\frac{\partial \rho F^{(m)}}{\partial t} = -\frac{\partial \rho w_i F^{(m)}}{\partial x_i} - \frac{\partial}{\partial w_i}\left[-\frac{1}{\tau_m}(w_i - U_i) + F_i^{(m)} \right]\rho F^{(m)}$$

$$+ \frac{2e}{3\tau_m}\frac{\partial^2 \rho F^{(m)}}{\partial w_j \partial w_j}. \quad (3.4)$$

The multiplication of Eq. (3.4) with the corresponding variables and integration over the velocity space results in transport equations for the fluid dynamic variables. For the mass density $\rho(\mathbf{x},t)$, fluid velocity $U_i(\mathbf{x},t)$ and specific kinetic energy $e(\mathbf{x},t)$ we obtain in this way:

$$\frac{\partial \rho}{\partial t} + \frac{\partial \rho U_i}{\partial x_i} = 0, \quad (3.5a)$$

$$\frac{\partial \rho U_j}{\partial t} + \frac{\partial \rho U_i U_j}{\partial x_i} + \frac{\partial p_{ij}}{\partial x_i} = \rho F_j, \quad (3.5b)$$

$$\frac{\partial \rho e}{\partial t} + \frac{\partial \rho U_k e}{\partial x_k} + \frac{\partial q_k}{\partial x_k} + p_{jk}\frac{\partial U_j}{\partial x_k} = \rho Q_B. \quad (3.5c)$$

On the right-hand sides, we applied for source terms due to external forces the abbreviations

$$F_j = \overline{F_j^{(m)}}, \quad Q_B = \overline{f_k v_k}, \quad (3.6)$$

where $f_k = F_k^{(m)} - F_k$ are fluctuations of external forces. On the left-hand sides of Eqs. (3.5b) and (3.5c), we defined the stress tensor p_{ij} and heat flux vector q_k via the relations

$$p_{ij} = \overline{\rho v_i v_j}, \quad (3.7a)$$

$$q_k = \frac{1}{2}\overline{\rho v_k v_n v_n}. \quad (3.7b)$$

Equations (3.5) were derived here as a consequence of the stochastic model (3.3), but they are independent of all the model details (with the exception of the appearance of the known external force $F_i^{(m)}$). The same equations (3.5) can be obtained by adopting the Boltzmann equation for the molecular motion; see, for instance, Cercignani [4]. Therefore, the physics is embedded in the unknown quantities of Eqs. (3.5b) and (3.5c): p_{ij} and q_k (the source term Q_B is assumed to be known).

C. Molecular stress tensor and heat flux equations

The calculation of p_{ij} and q_k can be addressed on the basis of their transport equations, which are implied by the stochastic model (3.3). In correspondence to the derivation of Eqs. (3.5), one obtains for p_{ij} the equation

$$\frac{\partial p_{ij}}{\partial t} + \frac{\partial U_k p_{ij}}{\partial x_k} + \frac{\partial \overline{\rho v_k v_i v_j}}{\partial x_k} + p_{jk}\frac{\partial U_i}{\partial x_k} + p_{ik}\frac{\partial U_j}{\partial x_k}$$

$$= -\frac{2}{\tau_m}\pi_{ij} + \rho\overline{f_i v_j} + \rho\overline{f_j v_i}. \quad (3.8)$$

With regard to the developments made in Sec. IV, it is con-

venient to split p_{ij} into an isotropic and deviatoric part:

$$p_{ij} = p\delta_{ij} + \pi_{ij}. \tag{3.9}$$

The isotropic part, which is the pressure, is related to ρ and e via

$$p = \frac{p_{ii}}{3} = \rho\frac{2e}{3}. \tag{3.10}$$

Hence, the equation for p follows from Eq. (3.5c). According to Eq. (3.8), π_{ij} satisfies the equation

$$\frac{\partial \pi_{ij}}{\partial t} + \frac{\partial U_k \pi_{ij}}{\partial x_k} + \frac{\partial \overline{\rho v_k (v_i v_j - v_n v_n \delta_{ij}/3)}}{\partial x_k}$$

$$- \rho\left(\overline{f_i v_j} + \overline{f_j v_i} - \frac{2}{3}\overline{f_n v_n}\delta_{ij}\right)$$

$$= -\frac{2}{\tau_m}\pi_{ij} - 2pS_{ij}{}^d - \pi_{jk}\frac{\partial U_i}{\partial x_k} - \pi_{ik}\frac{\partial U_j}{\partial x_k} + \frac{2}{3}\pi_{nk}\frac{\partial U_n}{\partial x_k}\delta_{ij}. \tag{3.11}$$

Here,

$$S_{ij}{}^d = \frac{1}{2}\left(\frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i} - \frac{2}{3}\frac{\partial U_n}{\partial x_n}\delta_{ij}\right) \tag{3.12}$$

refers to the deviatoric part of the rate-of-strain tensor:

$$S_{ij} = \frac{1}{2}\left(\frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i}\right). \tag{3.13}$$

A transport equation for q_i may be obtained on the basis of the PDF transport equation (3.4) in the same way as the transport equation for π_{ij}. However, this equation involves triple correlations which appear as unknowns. Therefore, the most convenient way to address the determination of q_i is to consider its calculation as a special case of the calculation of triple correlations. Their transport equation is given according to Eq. (3.4) by

$$\frac{\partial \overline{\rho v_i v_j v_k}}{\partial t} + \frac{\partial \rho U_m \overline{v_i v_j v_k}}{\partial x_m} - \rho\overline{f_i v_j v_k} - \rho\overline{f_j v_i v_k} - \rho\overline{f_k v_i v_j}$$

$$= -\frac{\partial \overline{\rho v_m v_i v_j v_k}}{\partial x_m} - \rho\frac{\partial U_i}{\partial x_m}\overline{v_m v_j v_k} - \rho\frac{\partial U_j}{\partial x_m}\overline{v_m v_i v_k}$$

$$- \rho\frac{\partial U_k}{\partial x_m}\overline{v_m v_i v_j} + \frac{\partial \overline{\rho v_i v_m}}{\partial x_m}\overline{v_j v_k} + \frac{\partial \overline{\rho v_j v_m}}{\partial x_m}\overline{v_i v_k}$$

$$+ \frac{\partial \overline{\rho v_k v_m}}{\partial x_m}\overline{v_i v_j} - \frac{3\rho}{\tau_m}\overline{v_i v_j v_k}. \tag{3.14}$$

Equation (3.11) for π_{ij} and Eq. (3.14) for triple correlations have to be complemented by a model for the characteristic correlation time scale τ_m of molecular velocity fluctuations [or, alternatively, for the kinematic viscosity ν; see relation (4.4) below]. Then, Eqs. (3.11) and (3.14) are still unclosed due to the appearance of third-order correlations in Eq. (3.11) and fourth-order correlations in Eq. (3.14). Approximations for these correlations may be obtained by reducing their transport equations to algebraic expressions or by adopting

the corresponding Gaussian expressions for these terms: see the developments made in Sec. IV.

The equations for π_{ij} and q_i obtained here differ significantly from corresponding equations which are postulated within the framework of the extended irreversible thermodynamics. Within that approach one does only obtain a certain class of transport equations for p_{ij} and q_i where several unknown parameters appear (see the discussion of Jou *et al.* [2]). In contrast to that, the use of the stochastic model (3.3) combined with a model for τ_m or ν corresponds to a consistent choice of parameters that are unknown within the approach of extended irreversible thermodynamics.

IV. ASSUMPTION OF A LOCAL EQUILIBRIUM

To obtain closed equations (3.5) and to discuss the difference to the Navier-Stokes equations, we will apply now the usual assumption that there exists a local equilibrium [2]. In this case, Eqs. (3.11) and (3.14) can be reduced to algebraic expressions for the molecular stress tensor and heat flux, as will be shown next.

A. Calculation of the deviatoric stress tensor

To derive an algebraic expression for π_{ij} one has to neglect the first two gradient terms on the left-hand side of Eq. (3.11). This corresponds to the assumption that there exists an equilibrium state where production, dissipation and molecular diffusion of π_{ij} balance each other locally. The third term on the left-hand of relation (3.11) represents an anisotropic contribution due to molecular diffusion: see, for instance, Eq. (4.7) below. Such diffusion terms are often found to be of minor relevance in turbulence models, and the same may be assumed regarding the molecular scale. Thus, we apply a Gaussian approximation for this term (which implies its neglect). Correspondingly, we may neglect the last three terms on the left-hand side of Eq. (3.11), which involve the anisotropic part of the production by external forces. Such external force-velocity correlations are found to be of minor relevance under many conditions. The resulting algebraic relation for π_{ij} then reads

$$\frac{2}{\tau_m}\pi_{ij} = -\frac{4e}{3}\rho S_{ij}{}^d - \pi_{jk}\frac{\partial U_i}{\partial x_k} - \pi_{ik}\frac{\partial U_j}{\partial x_k} + \frac{2}{3}\pi_{nk}\frac{\partial U_n}{\partial x_k}\delta_{ij}. \tag{4.1}$$

This equation for π_{ij} can be solved by adopting a technique that was pioneered by Pope [22–24] with respect to the construction of corresponding algebraic models for the stress tensor in turbulence models. The resulting general expression for π_{ij} can be written in terms of contributions of growing order in velocity gradients. A simple method to obtain the low-order terms of this general expression for π_{ij} is given by the following successive approximation.

In the first order of approximation we assume that π_{ij} is a linear function of velocity gradients. This is equivalent to the neglect of all the terms that involve π_{ij} multiplied by velocity gradients in Eq. (4.1). In this case, we find for π_{ij} the expression

$$\pi_{ij}^{(1)} = -2\mu S_{ij}^{\ d}, \tag{4.2}$$

where we introduced the viscosity

$$\mu = \rho\nu. \tag{4.3}$$

The kinematic viscosity ν, which appears as a diffusion coefficient in Eq. (4.2), is an abbreviation for

$$\nu = \frac{\tau_m e}{3}. \tag{4.4}$$

By introducing the mean thermal molecular velocity $u_T = (2e)^{1/2}$ and mean free path $\lambda = \tau_m (e/2)^{1/2}$, Eq. (4.4) may also be brought into the form which is used in kinetic gas theory, $\nu = u_T \lambda/3$. The analysis of the acceleration model (3.1) in the Appendix shows that the consideration of finite τ_a does not result in a change of the structure of Eq. (4.2), but expression (4.4) is generalized by Eq. (A7).

In the second order of approximation, we apply the first-order approximation (4.2) in the terms that involve π_{ij} on the right-hand side of Eq. (4.1). This leads to an expression for π_{ij} which involves velocity gradients of second order:

$$\pi_{ij}^{(2)} = \pi_{ij}^{(1)} - \pi_{jk}^{(1)}\frac{\tau_m}{2}\frac{\partial U_i}{\partial x_k} - \pi_{ik}^{(1)}\frac{\tau_m}{2}\frac{\partial U_j}{\partial x_k} + \pi_{nk}^{(1)}\frac{\tau_m}{3}\frac{\partial U_n}{\partial x_k}\delta_{ij}. \tag{4.5}$$

Higher-order approximations can be constructed by repeating this successive approximation.

The comparison of relation (4.5) with models derived within the framework of extended irreversible thermodynamics (see the corresponding discussion of Jou *et al.* [2]) reveals the same advantage as pointed out above with regard to equations for the transport of the molecular stress tensor and heat flux: the coefficients of nonlinear terms are completely determined here. This fact is relevant to simulations of high-Mach-number flows because experimental data for the assessment of simulation results are hardly available for Mach numbers $N_{\text{Ma}} > 4$ [2]. It is worth noting that the construction of nonlinear models for the (subgrid-scale) stress tensor in filtered equations for turbulence also leads to the question of how the coefficients of nonlinear contributions have to be chosen [25,26]. With regard to that, it has been shown recently that the explanation of the dynamics of turbulent fluctuations by a stochastic model represents a way to overcome this problem [27].

B. Heat flux calculation

With the same arguments as applied above, we neglect all the terms on the left-hand side of Eq. (3.14). Further, we approximate the velocity correlation of fourth order by the corresponding Gaussian expression, which is a requirement to be in consistency with the use of the Gaussian approximation for the triple correlation term in Eq. (3.11). In combination with the assumption of isotropic velocity variances this leads to the relation

$$\frac{3}{\tau_m}\overline{v_i v_j v_k} = -\frac{4e}{9}\frac{\partial e}{\partial x_m}(\delta_{im}\delta_{jk} + \delta_{jm}\delta_{ik} + \delta_{km}\delta_{ij})$$
$$-\frac{\partial U_i}{\partial x_m}\overline{v_m v_j v_k} - \frac{\partial U_j}{\partial x_m}\overline{v_m v_i v_k} - \frac{\partial U_k}{\partial x_m}\overline{v_m v_i v_j}. \tag{4.6}$$

In analogy to the derivation of Eq. (4.2), the first-order approximation for triple correlations is obtained by neglecting the last three terms (related to the production by velocity gradients) in Eq. (4.6):

$$\overline{v_i v_j v_k}^{(1)} = -\frac{4\nu}{9}\frac{\partial e}{\partial x_m}(\delta_{im}\delta_{jk} + \delta_{jm}\delta_{ik} + \delta_{km}\delta_{ij}). \tag{4.7}$$

The second order of approximation is found by inserting Eq. (4.7) into the last three terms of Eq. (4.6):

$$\overline{v_i v_j v_k}^{(2)} = \overline{v_i v_j v_k}^{(1)} - \frac{\tau_m}{3}$$
$$\times\left(\frac{\partial U_i}{\partial x_m}\overline{v_m v_j v_k}^{(1)} + \frac{\partial U_j}{\partial x_m}\overline{v_m v_i v_k}^{(1)} + \frac{\partial U_k}{\partial x_m}\overline{v_m v_i v_j}^{(1)}\right). \tag{4.8}$$

According to its definition (3.7b), the heat flux q_i is then given in the first order of approximation by the corresponding reduction of Eq. (4.7):

$$q_i^{(1)} = -\kappa\frac{\partial e}{\partial x_i}. \tag{4.9}$$

Here, we introduced the heat conduction coefficient κ that is determined by

$$\kappa = \frac{10}{9}\rho\nu. \tag{4.10}$$

The ratio of the diffusion coefficients $\mu = \rho\nu$ in Eq. (4.2) and κ in Eq. (4.9) determines the Prandtl number N_{Pr} in the specific kinetic energy equation, which is defined by

$$N_{\text{Pr}} = \frac{\mu}{\kappa}. \tag{4.11}$$

The use of expression (4.10) in the definition (4.11) implies then the value

$$N_{\text{Pr}} = \frac{9}{10}, \tag{4.12}$$

as a consequence of the stochastic model (3.3). This result agrees with the corresponding consequence of the Chapman-Enskog method [14]. The explanation of Prandtl number variations requires consideration of finite τ_a: see expression (A10) in the Appendix.

The calculation of q_i in the second order of approximation leads by means of Eq. (4.8) to

$$q_i^{(2)} = q_i^{(1)} - \frac{\tau_m}{3}\left(\frac{\partial U_i}{\partial x_n}q_n^{(1)} + \frac{\partial U_k}{\partial x_n}\overline{\rho v_n v_i v_k}^{(1)}\right). \quad (4.13)$$

The last term can be rewritten by replacing the gradient of the kinetic energy e in Eq. (4.7) by means of expression (4.9):

$$\overline{v_n v_i v_k}^{(1)} = \frac{2}{5\rho}q_m^{(1)}(\delta_{nm}\delta_{ik} + \delta_{im}\delta_{nk} + \delta_{km}\delta_{ni}). \quad (4.14)$$

This results then in the following expression for the heat flux q_i in the second order of approximation:

$$q_i^{(2)} = q_i^{(1)} - \frac{\tau_m}{3}\frac{\partial U_i}{\partial x_n}q_n^{(1)} - \frac{2\tau_m}{15}\frac{\partial U_k}{\partial x_n}q_m^{(1)}(\delta_{nm}\delta_{ik} + \delta_{im}\delta_{nk}$$
$$+ \delta_{km}\delta_{ni}). \quad (4.15)$$

Higher-order approximations may be found by repeating this successive approximation. Essentially, the discussion of the advantages of Eq. (4.5) for the molecular stress tensor also applies to Eq. (4.15).

V. SCALING ANALYSIS

A way to assess the range of applicability of various models (of the Navier-Stokes model) for π_{ij} and q_i is to rescale the fluid dynamic equations. This will be done next by adopting a reference density ρ_0, velocity U_0, kinetic energy e_0, and length L_0 (the reference time scale is L_0/U_0).

A. Scaling parameters

The only parameter of the stochastic model (3.3) is τ_m. By invoking relation (4.4), Eqs. (3.5) then only depend on ν. By adopting the scaling variables U_0 and L_0, the kinematic viscosity ν can be made dimensionless by introducing the Reynolds number

$$N_{\text{Re}} = \frac{U_0 L_0}{\nu}. \quad (5.1)$$

The coupling between the velocity and energy equations can be assessed by introducing the speed of sound $a_0 = (\gamma p_0/\rho_0)^{1/2}$ via the following parametrization of e_0:

$$e_0 = \frac{3p_0}{2\rho_0} = \frac{3}{2\gamma}a_0^2. \quad (5.2)$$

The middle expression determines a reference pressure p_0 according to Eq. (3.10). The parameter $\gamma = c_p/c_v$ refers to the ratio of the constant-pressure to constant-volume specific heats (for monatomic fluids, γ has the value $\gamma = 5/3$). By applying a_0, the ratio between the characteristic molecular velocity scale U_0 to the velocity scale a_0 related to the energy equation can be determined by means of the Mach number

$$N_{\text{Ma}} = \frac{U_0}{a_0}, \quad (5.3)$$

which will be used below to assess the relevance of corrections to the Navier-Stokes model.

B. Scaling of the first-order approximations

By scaling Eqs. (3.5) in terms of the scaling parameters introduced above, one obtains the following expressions for the scaled deviatoric part π_{ij} of the stress tensor and heat flux q_i in the first order of approximation:

$$\frac{\pi_{ij}^{(1)}}{\rho_0 U_0^2} = -\frac{2}{N_{\text{Re}}}\rho^+ S_{ij}^{d^+}, \quad (5.4a)$$

$$\frac{q_i^{(1)}}{\rho_0 U_0 e_0} = -\frac{10}{9N_{\text{Re}}}\rho^+\frac{\partial e^+}{\partial x_i^+}. \quad (5.4b)$$

The plus refers to scaled quantities; this means we applied in Eqs. (5.4) the abbreviations $\rho^+ = \rho/\rho_0$, $S_{ij}^{d^+} = S_{ij}^d L_0/U_0$, and $e^+ = e/e_0$.

These expressions recover the known scaling of the first-order approximations with the inverse Reynolds number: these terms (and all higher-order approximations, see below) become very small in the case of a high Reynolds number (a small kinematic viscosity ν). However, their neglect would result in the reduction of Eqs. (3.5b) and (3.5c) to partial differential equations of first order, which has consequences with regard to the boundary conditions that can be applied [24].

C. Scaling of the second-order approximations

The expressions (4.5) and (4.15), which were obtained for the deviatoric part π_{ij} of the stress tensor and heat flux q_i in the second order of approximation, reveal that the dimensionless quantity $\tau_m \partial U_i/\partial x_j$ controls the appearance of contributions in addition to first-order approximations. By adopting Eq. (4.4) for τ_m, this quantity can be written

$$\tau_m\frac{\partial U_i}{\partial x_j} = \frac{3\nu}{e}\frac{\partial U_i}{\partial x_j} = \frac{2\gamma N_{\text{Ma}}^2}{e^+}\frac{1}{N_{\text{Re}}}\frac{\partial U_i^+}{\partial x_j^+} = \frac{2\gamma}{e^+}N_{\text{Re}}N_{\text{Kn}}^2\frac{\partial U_i^+}{\partial x_j^+}.$$
$$(5.5)$$

This expression makes use of the definition of the Knudsen number

$$N_{\text{Kn}} = \frac{N_{\text{Ma}}}{N_{\text{Re}}} = \frac{e^+}{\sqrt{6\gamma}}\frac{\tau_m\sqrt{e_0}}{L_0}. \quad (5.6)$$

The last expression in Eq. (5.6) results from the application of the definitions of N_{Ma} and N_{Re} combined with Eq. (4.4) for ν and Eq. (5.2) for e_0.

The combination of Eqs. (5.4) and (5.5) shows that second-order approximations scale with the squared Knudsen number. Expression (5.5) can be used to assess the requirement to involve terms in addition to first-order approximations for π_{ij} and q_i. Compared to first-order contributions, second-order contributions scale with $N_{\text{Re}}N_{\text{Kn}}^2 = N_{\text{Ma}}^2/N_{\text{Re}}$, which is shown in Fig. 2 for different values of N_{Ma}. The influence of the Mach number can be investigated by rescaling the fluid dynamic equations such that positions are normalized on the mean free path $\lambda = \tau_m(e/2)^{1/2}$. This was done by Levermore *et al.* [1] who investigated the relevance of corrections to the Navier-Stokes model by comparing sta-
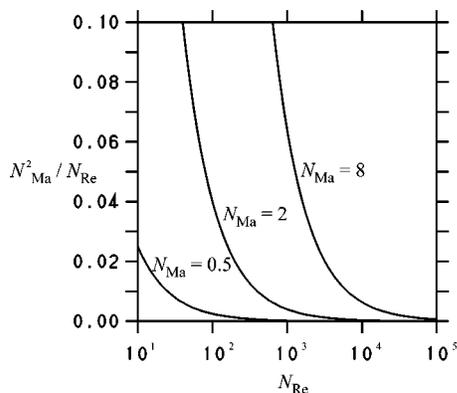
FIG. 2. The parameter N_{Ma}^2/N_{Re} in relation (5.5) against the Reynolds number N_{Re} for different values of the Mach number N_{Ma}.

tionary planar shock profiles calculated by the Navier-Stokes model and the direct simulation Monte Carlo method developed by Bird [5]. According to their results, the applicability of first-order approximations for π_{ij} and q_i is well justified for $N_{Ma} \lesssim 2$. For $N_{Ma}=3$, they observed deviations of 10%–30%, and they found an unacceptable performance of first-order approximations for the case $N_{Ma}=4$. On this basis, one may estimate by means of Fig. 2 that the Navier-Stokes model is well applicable in cases where $N_{Ma} \lesssim 2$ and N_{Re} exceeds the critical Reynolds number for the onset of turbulence ($N_{Re} > 400$).

VI. SUMMARY AND CONCLUDING REMARKS

The relation between the molecular and fluid dynamics is relevant to a rigorous foundation of the theory of turbulence, but methods applied previously did not provide sufficient answers to this question. Lebowitz, for example, stated: "It is no secret that there does not exist at present anything resembling a rigorous derivation of the hydrodynamic equations governing the time evolution of macroscopic variables from the laws governing the dynamics of their microscopic constituents" [28]. With regard to this question, it is of interest that the analysis of the stochastic motion of molecules provides a relatively simple view of the relation between the molecular and fluid dynamics. The basis for that is given by deriving the stochastic acceleration model (3.1) for the molecular dynamics of dense fluids from any (complete) deterministic dynamics by means of the projection operator technique [9,20]. For simplicity, the analysis of this acceleration model was performed such that the implications of its asymptotic limit [of the velocity model (3.3)] were studied first, and the investigation of Eqs. (3.1) in the Appendix was used then to explain modifications which arise from the consideration of the more general acceleration model.

In both variants, this means by adopting the molecular acceleration or velocity model, the Navier-Stokes model can be derived as the result of three steps, see the illustration in Fig. 1. In a first step, transport equations for the molecular stress tensor and heat flux were obtained, which appear as unknowns in Eqs. (3.5). These transport equations can be closed by adopting parametrizations for velocity correlations

of third and fourth order; see Sec. IV. In a second step we applied the usual local-equilibrium assumption in the equations for the molecular stress tensor and heat flux, which implies algebraic corrections to the Navier-Stokes model of growing order in velocity gradients. All these expressions, which may be derived by adopting an integrity basis [22–24], are fully determined (additional parameters are not involved). In a third step it was shown that the Navier-Stokes model is recovered provided that $\tau_m \partial U_i / \partial x_j$ becomes small (due to the fact that $\tau_a \lesssim \tau_m$ it is sufficient to consider a constraint for τ_m). This condition implies the need that $Mg = \tau_m S \ll 1$. Here, $Mg = \tau_m / S^{-1}$ represents a dimensionless time scale ratio where $S = (2S_{ik}^{\ d}S_{ki}^{\ d})^{1/2}$ refers to the characteristic shear rate of fluid motion. Hence, the applicability of the Navier-Stokes model requires that the scales of molecular and fluid motions be well separated. It is of interest to note that the time scale ratio Mg, which controls the relevance of corrections to the Navier-Stokes model (Mach number effects), has the same structure as the gradient Mach number that is used as an indicator for the relevance of compressibility effects within the framework of turbulence models [29,30].

In that way, answers to questions (i) and (ii) described in the Introduction were presented. With regard to question (i), it was shown that any deterministic dynamics imply a generalization of Kirkwood's velocity model: the acceleration model (3.1). This model may be applied to flows with different Prandtl numbers; this mean to both dense gases and liquids. With regard to question (ii), the benefit of the findings presented here is given by the possibility to control the applicability of the Navier-Stokes model. This can be done by monitoring Mg. This number is available in simulations, as may be seen by replacing τ_m by ν via relation (4.4). It is worth emphasizing that the use of the condition $Mg \ll 1$ is significantly simpler than the application of certain matrices from moments of the molecular velocity PDF, as suggested by Levermore *et al.* [1]. Corrections to the Navier-Stokes model and nonequilibrium effects can be involved (without any need for the introduction of additional parameters) if the applicability of the Navier-Stokes model is not well justified, as given, for example, with regard to high-Mach-number flows with shocks.

APPENDIX

In analogy to the consideration of consequences of the stochastic velocity model (3.3) in Secs. III and IV, we analyze here the implications of the more general acceleration model (3.1). This can be done on the basis of the Fokker-Planck equation

$$\frac{\partial \rho f^{(m)}}{\partial t} = -\frac{\partial \rho w_i f^{(m)}}{\partial x_i} - \frac{\partial \rho \alpha_i f^{(m)}}{\partial w_i}$$
$$+ \frac{\partial}{\partial \alpha_i}\left[\alpha_i + \frac{1}{\tau_m}(w_i - U_i) - F_i^{(m)}\right]\frac{\rho}{\tau_a}f^{(m)}$$
$$+ \frac{2e}{3\tau_m\tau_a{}^2}\frac{\partial^2 \rho f^{(m)}}{\partial \alpha_j \partial \alpha_j}, \tag{A1}$$

which corresponds to the stochastic model (3.1). Here $f^{(m)}$ refers to the joint acceleration-velocity PDF conditioned on the position $\mathbf{x}$, and $\boldsymbol{\alpha}$ denotes the sample-space acceleration.

By multiplying Eq. (A1) with appropriate variables and integration one may find transport equations for all the moments of the joint acceleration-velocity PDF. The direct integration of Eq. (A1) recovers the continuity equation (3.5a). The equations for the mean velocity U_i and acceleration $\bar{A}_i$ read

$$\frac{DU_i}{Dt} + \frac{\partial \rho \overline{v_m v_i}}{\rho \partial x_m} = \bar{A}_i, \tag{A2a}$$

$$\frac{D\bar{A}_i}{Dt} + \frac{\partial \rho \overline{v_m a_i}}{\rho \partial x_m} = \frac{1}{\tau_a}(F_i - \bar{A}_i), \tag{A2b}$$

where $D/Dt = \partial/\partial t + U_m \partial/\partial x_m$ denotes the substantial derivative. The equations for the moments of second order are given by

$$\frac{D\overline{v_i v_j}}{Dt} + \frac{\partial \rho \overline{v_m v_i v_j}}{\rho \partial x_m} + \overline{v_j v_m}\frac{\partial U_i}{\partial x_m} + \overline{v_i v_m}\frac{\partial U_j}{\partial x_m} = \overline{a_i v_j} + \overline{a_j v_i}, \tag{A3a}$$

$$\frac{D\overline{a_i v_j}}{Dt} + \frac{\partial \rho \overline{v_m a_i v_j}}{\rho \partial x_m} + \overline{v_j v_m}\frac{\partial \bar{A}_i}{\partial x_m} + \overline{a_i v_m}\frac{\partial U_j}{\partial x_m}$$
$$= \overline{a_i a_j} - \frac{1}{\tau_a}\left(\overline{a_i v_j} - \overline{f_i v_j} + \frac{\overline{v_i v_j}}{\tau_m}\right), \tag{A3b}$$

$$\frac{D\overline{a_i a_j}}{Dt} + \frac{\partial \rho \overline{v_m a_i a_j}}{\rho \partial x_m} + \overline{a_j v_m}\frac{\partial \bar{A}_i}{\partial x_m} + \overline{a_i v_m}\frac{\partial \bar{A}_j}{\partial x_m}$$
$$= -\frac{1}{\tau_a}\left(2\overline{a_i a_j} - \overline{f_i a_j} - \overline{f_j a_i} + \frac{\overline{a_i v_j} + \overline{a_j v_i}}{\tau_m} - \frac{4e}{3\tau_a\tau_m}\delta_{ij}\right), \tag{A3c}$$

where a_i and f_i refer to acceleration and body force fluctuations, respectively. Correspondingly, one finds for the moments of third order the transport equations

$$\frac{D\overline{v_i v_j v_k}}{Dt} + \overline{v_j v_k v_m}\frac{\partial U_i}{\partial x_m} + \overline{v_i v_k v_m}\frac{\partial U_j}{\partial x_m} + \overline{v_i v_j v_m}\frac{\partial U_k}{\partial x_m}$$
$$+ \frac{\partial \rho \overline{v_m v_i v_j v_k}}{\rho \partial x_m} - \frac{\partial \rho \overline{v_m v_i}}{\rho \partial x_m}\overline{v_j v_k} - \frac{\partial \rho \overline{v_m v_j}}{\rho \partial x_m}\overline{v_i v_k} - \frac{\partial \rho \overline{v_m v_k}}{\rho \partial x_m}\overline{v_i v_j}$$
$$= \overline{a_i v_j v_k} + \overline{a_j v_i v_k} + \overline{a_k v_i v_j}, \tag{A4a}$$
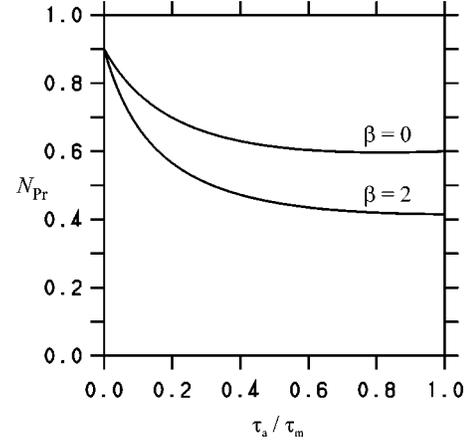
FIG. 3. The dependence of the Prandtl number N_{Pr} on τ_a/τ_m according to Eq. (A10) for different values of the parameter β.

$$\frac{D\overline{a_i v_j v_k}}{Dt} + \overline{v_j v_k v_m}\frac{\partial \bar{A}_i}{\partial x_m} + \overline{a_i v_k v_m}\frac{\partial U_j}{\partial x_m} + \overline{a_i v_j v_m}\frac{\partial U_k}{\partial x_m}$$
$$+ \frac{\partial \rho \overline{v_m a_i v_j v_k}}{\rho \partial x_m} - \frac{\partial \rho \overline{v_m a_i}}{\rho \partial x_m}\overline{v_j v_k} - \frac{\partial \rho \overline{v_m v_j}}{\rho \partial x_m}\overline{a_i v_k} - \frac{\partial \rho \overline{v_m v_k}}{\rho \partial x_m}\overline{a_i v_j}$$
$$= \overline{a_i a_j v_k} + \overline{a_i a_k v_j} - \frac{1}{\tau_a}\left(\overline{a_i v_j v_k} + \frac{\overline{v_i v_j v_k}}{\tau_m} - \overline{f_i v_j v_k}\right), \tag{A4b}$$

$$\frac{D\overline{a_i a_j v_k}}{Dt} + \overline{a_j v_k v_m}\frac{\partial \bar{A}_i}{\partial x_m} + \overline{a_i v_k v_m}\frac{\partial \bar{A}_j}{\partial x_m} + \overline{a_i a_j v_m}\frac{\partial U_k}{\partial x_m}$$
$$+ \frac{\partial \rho \overline{v_m a_i a_j v_k}}{\rho \partial x_m} - \frac{\partial \rho \overline{v_m a_i}}{\rho \partial x_m}\overline{a_j v_k} - \frac{\partial \rho \overline{v_m a_j}}{\rho \partial x_m}\overline{a_i v_k} - \frac{\partial \rho \overline{v_m v_k}}{\rho \partial x_m}\overline{a_i a_j}$$
$$= \overline{a_i a_j a_k} - \frac{1}{\tau_a}\left(2\overline{a_i a_j v_k} + \frac{\overline{a_i v_j v_k} + \overline{a_j v_i v_k}}{\tau_m} - \overline{f_i a_j v_k} - \overline{f_j a_i v_k}\right), \tag{A4c}$$

$$\frac{D\overline{a_i a_j a_k}}{Dt} + \overline{a_j a_k v_m}\frac{\partial \bar{A}_i}{\partial x_m} + \overline{a_i a_k v_m}\frac{\partial \bar{A}_j}{\partial x_m} + \overline{a_i a_j v_m}\frac{\partial \bar{A}_k}{\partial x_m}$$
$$+ \frac{\partial \rho \overline{v_m a_i a_j a_k}}{\rho \partial x_m} - \frac{\partial \rho \overline{v_m a_i}}{\rho \partial x_m}\overline{a_j a_k} - \frac{\partial \rho \overline{v_m a_j}}{\rho \partial x_m}\overline{a_i a_k} - \frac{\partial \rho \overline{v_m a_k}}{\rho \partial x_m}\overline{a_i a_j}$$
$$= -\frac{1}{\tau_a}\left(3\overline{a_i a_j a_k} + \frac{\overline{v_i a_j a_k} + \overline{v_j a_i a_k} + \overline{v_k a_i a_j}}{\tau_m}\right.$$
$$\left. - \overline{f_i a_j a_k} - \overline{f_j a_i a_k} - \overline{f_k a_i a_j}\right). \tag{A4d}$$

First, we will use Eqs. (A3) to generalize the calculation of the deviatoric stress tensor π_{ij} in Sec. IV. For simplicity, the effect of finite τ_a will be only considered with respect to the first-order approximation of π_{ij} (higher-order approximations can be obtained as shown in Sec. IV). By neglecting body force effects and the left-hand sides of Eqs. (A3b) and (A3c), we obtain for $\overline{a_i v_j}$ and $\overline{a_i a_j}$ the expressions

$$\overline{a_i v_j} = -\frac{1}{\rho \tau_m} \frac{\pi_{ij}}{1 + \tau_a/\tau_m}, \quad \overline{a_i a_j} = \frac{1}{\tau_a}\left(\overline{a_i v_j} + \frac{1}{\tau_m} \overline{v_i v_j} \right).$$

$$\text{(A5)}$$

By adopting this expression for $\overline{a_i v_j}$, Eq. (A3a) provides equations for the kinetic energy e and π_{ij}. The equation for e corresponds to Eq. (3.5c) with the exception that $Q_B = 0$ since body force effects are neglected here. By neglecting contributions due to the first two terms on the left-hand side of Eq. (A3a) and adopting isotropic variances in the production terms (which are proportional to velocity gradients), the equation for π_{ij} implies the first-order approximation

$$\pi_{ij}^{(1)} = -2\mu S_{ij}^{d}. \qquad \text{(A6)}$$

Here, the viscosity $\mu = \rho\nu$, where the kinematic viscosity ν is given in generalization of Eq. (4.4) by

$$\nu = \frac{\tau_m e}{3}\left(1 + \frac{\tau_a}{\tau_m}\right). \qquad \text{(A7)}$$

The generalization of the heat flux calculation can be performed in correspondence to the developments made in Sec. IV, where again the attention is restricted to the approximation of first order. We neglect body force effects and the first four terms on the left-hand sides of Eqs. (A4). In addition to that, we approximate correlations of fourth order by their corresponding Gaussian expressions, and we apply $\overline{v_i v_j} = 2e/3 \, \delta_{ij}$, $\overline{a_i v_j} = 0$ and $\overline{a_i a_j} = 2e/(3\tau_a \tau_m)\delta_{ij}$ for the variances: see the relation (A5). In this case, Eqs. (A4) reduce to

$$\frac{4e}{9}\frac{\partial e}{\partial x_m}[\delta_{im}\delta_{jk} + \delta_{jm}\delta_{ik} + \delta_{km}\delta_{ij}] = \overline{a_i v_j v_k} + \overline{a_j v_i v_k} + \overline{a_k v_i v_j},$$

$$\text{(A8a)}$$

$$0 = \tau_a \tau_m [\overline{a_i a_j v_k} + \overline{a_i a_k v_j}] - \tau_m \overline{a_i v_j v_k} - \overline{v_i v_j v_k}, \quad \text{(A8b)}$$

$$\frac{4e}{9}(1+\beta)\frac{\partial e}{\partial x_m}\delta_{km}\delta_{ij} = \tau_a \tau_m \overline{a_i a_j a_k} - 2\tau_m \overline{a_i a_j v_k} - \overline{a_i v_j v_k}$$
$$- \overline{a_j v_i v_k}, \qquad \text{(A8c)}$$

$$0 = 3\tau_m \overline{a_i a_j a_k} + \overline{a_j a_k v_i} + \overline{a_i a_k v_j} + \overline{a_i a_j v_k}, \qquad \text{(A8d)}$$

where $\beta = -(e/\tau_a \tau_m)\partial(\tau_a \tau_m)/\partial e$ is introduced. To derive Eq. (A8c) we used the assumption that τ_a and τ_m may vary in space via a temperature (or, alternatively, kinetic energy) dependence, $\partial(\tau_a \tau_m)/\partial x_m = [\partial(\tau_a \tau_m)/\partial e]\partial e/\partial x_m$. The analysis of Eqs. (A8) then shows that

$$q_i^{(1)} = -\frac{\rho\nu}{N_{\text{Pr}}}\frac{\partial e}{\partial x_i}. \qquad \text{(A9)}$$

ν is determined by Eq. (A7), and the Prandtl number N_{Pr} is given in generalization of Eq. (4.12) by

$$N_{\text{Pr}} = \frac{9}{10}\left(1 + \frac{\tau_a}{\tau_m}\right)\left(1 + 0.5\frac{\tau_a}{\tau_m}\right)\left(1 + (3.5+\beta)\frac{\tau_a}{\tau_m}\right)^{-1}. $$

$$\text{(A10)}$$

Corresponding Prandtl number variations are illustrated in Fig. 3 for different values of β. One observes that N_{Pr} increases with decreasing resolution (if τ_a effects become smaller).

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