Time-structure invariance criteria for closure approximations

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In many areas of physics, time evolution equations for moments of distributions are expressed in terms of higher-order moments. Closure approximations are then introduced in an *ad hoc* fashion to reduce the higher-order moments to functions of the lower-order ones. Herein, the time-structure invariance of the Poisson bracket as manifested through the Jacobi identity is used to derive constraint relationships on these approximations. These constraints severely limit the allowable functionality of general closures and help to define the boundaries within which future investigations should concentrate. [S1063-651X(97)10410-X]

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INTRODUCTION

In many areas of physics, time evolution equations for probability distributions are typically expressed in terms of moments averaged over the probability space. These areas include polymer dynamics [1], quantum field theory [2], suspension and colloid fluid mechanics [3,4], kinetics of phase transitions and spinodal decomposition [5,6], the mechanics of turbulence [7–9], liquid-crystalline dynamics [1,6], the fluid mechanics of immiscible blends [10,11], and statistical mechanics [12,13]. These moment evolution equations are much simpler to solve computationally than the full probability distribution evolution equation, but typically involve higher-order moments appearing in the equations for the lower-order moments. Closure approximations are then introduced in order to reduce the moments of higher order in terms of lower-order ones, but, in many cases, very little physical guidance is used in the construction of these closure approximations, with the result that many of those used display aphysical behavior under certain circumstances. Furthermore, with the few genuine restrictions on the allowable functional forms of these approximations currently in use, an infinite variety of them are available, each of which must be tested individually, which is an even more computationally intensive process than solving the full evolution equation for the probability distribution itself, e.g., by simulation techniques [14]. The magnitude of literature devoted to testing closure approximations is voluminous and testifies not only to the importance of the issue, but also to the lack of physical guidance that is available for their construction.

As a specific example of the use of closures in the mechanics of a suspension of particles in an incompressible Newtonian fluid, the evolution equation for the second moment of the probability density function $\mathbf{P}(\mathbf{x},t)$ is given by [4]

$$\begin{aligned}\frac{DP_{\alpha\beta}}{Dt} &= \tfrac{1}{2}(\nabla_\gamma v_\alpha - \nabla_\alpha v_\gamma)P_{\gamma\beta} + \tfrac{1}{2}(\nabla_\gamma v_\beta - \nabla_\beta v_\gamma)P_{\alpha\gamma} \\ &\quad + \tfrac{1}{2}\lambda(\nabla_\gamma v_\alpha + \nabla_\alpha v_\gamma)P_{\gamma\beta} + \tfrac{1}{2}\lambda(\nabla_\gamma v_\beta + \nabla_\beta v_\gamma)P_{\alpha\gamma} \\ &\quad - 2\lambda Q_{\alpha\beta\gamma\varepsilon}\nabla_\gamma v_\varepsilon + 2D(\delta_{\alpha\beta} - 3P_{\alpha\beta}), \end{aligned} \quad (1)$$

$\nabla\mathbf{v}$ being the velocity gradient tensor, λ the particle shape factor, D the rotational diffusivity, and $D(*)/Dt$ the material derivative of $*$. The quantities $\mathbf{P}$ and $\mathbf{Q}$ are defined as

$$P_{\alpha\beta} \equiv \int p_\alpha p_\beta \psi \, d^3p, \quad Q_{\alpha\beta\gamma\varepsilon} \equiv \int p_\alpha p_\beta p_\gamma p_\varepsilon \psi \, d^3p, \quad (2)$$

$\mathbf{p}$ being the unit vector pointing in the direction of the major axis of a given particle and $\psi(\mathbf{p},\mathbf{x},t)$ the probability density function. With these definitions, normalization of the distribution function at any position $\mathbf{x}$ and time t requires that $P_{\alpha\alpha}=1$ and $Q_{\alpha\alpha\gamma\varepsilon}=P_{\gamma\varepsilon}$.

Equation (1) would be a straightforward expression to solve, even under general circumstances, were it not for the fourth moment of the distribution $\mathbf{Q}$ appearing on its right-hand side. Similarly to the procedure outlined above, an evolution equation for the fourth moment can also be derived, but it turns out to depend upon the sixth moment and so on, *ad infinitum*. Thus, in order to gain any practical advantages from Eq. (1) it is necessary to devise a closure approximation for the fourth moment in terms of the second.

Another tangible example of the use of closure approximations is provided by the Doi-Ohta theory of incompressible, immiscible blends [10]. In this theory, the droplets of the dispersed phase are taken to have a surface area (per unit volume) Q given by

$$Q = \int f(\mathbf{n},\mathbf{x},t) d^3n, \quad (3)$$

where $\mathbf{n}$ is the outwardly directed unit vector normal to the droplet interface. The conservative dynamics of the second moment of the interphase density function f follow an evolution equation of the form

$$\frac{DN_{\alpha\beta}}{Dt} = -N_{\alpha\gamma}\nabla_\beta v_\gamma - N_{\beta\gamma}\nabla_\alpha v_\gamma + Z_{\alpha\beta\gamma\varepsilon}\nabla_\gamma v_\varepsilon, \quad (4)$$

where

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$$N_{\alpha\beta}\equiv\int n_\alpha n_\beta f(\mathbf{n},\mathbf{x},t)d^3n,$$

$$Z_{\alpha\beta\gamma\varepsilon}\equiv\int n_\alpha n_\beta n_\gamma n_\varepsilon f(\mathbf{n},\mathbf{x},t)d^3n. \tag{5}$$

Again, one is faced with obtaining an approximation for the fourth moment appearing in Eq. (4) in terms of the second moment; however, this time the normalization condition requires that $N_{\alpha\alpha}=Q$, so that we no longer have a constraint upon the first invariant (i.e., the trace) of the second moment.

Unfortunately, there is very little physical guidance available to use when motivating a particular closure. Typically, one would like the closure $\mathbf{R}$ to be approximately equal to $\mathbf{Q}$ or $\mathbf{Z}$ in arbitrary flow fields. Since $\mathbf{R}=f(\mathbf{A})$ (with $\mathbf{A}$ denoting either $\mathbf{P}$ or $\mathbf{N}$), it is expected that $\mathbf{R}$ should possess some of the inherent symmetries of $\mathbf{Q}$ or $\mathbf{Z}$, say,

$$R_{\alpha\beta\gamma\varepsilon}=R_{\gamma\varepsilon\alpha\beta}=R_{\beta\alpha\gamma\varepsilon}=R_{\alpha\beta\varepsilon\gamma}, \tag{6}$$

for example. Another typical requirement ensures the correct normalization, $R_{\alpha\alpha\gamma\varepsilon}=A_{\gamma\varepsilon}$. These two restrictions, in most studies, represent most of the physical information that is brought to bear upon the choice of the closure. In sophisticated studies, such as that of Hinch and Leal [4], limiting forms of the equations are examined, where the closures can be fitted exactly. *Ad hoc* interpolations are then constructed for intermediate circumstances.

More recent investigations [15,16] are beginning to provide adequate approximations for the majority of the few, simple, homogeneous flow fields examined. In these studies, very general approximations are written down in terms of arbitrary scalar functions of the invariants of the second moment [17] and these functions are evaluated by fitting the approximation to the exact solution involving the distribution function for a few well-defined flow problems. However, no other physical guidance is available to aid in the selection of these functions and their evaluation based upon obtaining the distribution function is, although on a much more limited scale, solving the problem one was trying to avoid in the first place. Furthermore, whether or not, and to what extent, these closures will work in inhomogeneous flow fields should be a major concern.

Although the future development of closure approximations will probably, by necessity, follow along the lines outlined in the preceding paragraph, the work effort may be significantly reduced if one has an *a priori* idea, based upon some meaningful physical guidance, as to what the allowable forms of the arbitrary functionals are in order to ensure that the overall dynamical structure of the total system of equations is preserved. Clearly, more physical criteria are needed to filter the excessive functionality imbedded in the mathematical nature of closure approximations. In this article we offer assistance in this regard by formulating general guidelines for the selection of these closures based upon the time-structure invariance inherent to the reversible dynamics of physical systems. The application of this technique will be illustrated for the examples cited above, which are representative of similar equations in other areas of physics. For simplicity in the following analysis, we shall set the particle shape factor λ equal to unity, corresponding to very long, thin particles in the first example. The first step, however, is to formulate and explain time-structure invariance.

TIME-STRUCTURE INVARIANCE

The principle of time-structure invariance is imbedded in the Poisson bracket of Hamiltonian mechanics. In this framework, the reversible dynamics of an arbitrary functional F are governed by the expression

$$\frac{dF}{dt}=\{F,H\}, \tag{7}$$

where $\{,\}$ denotes the Poisson bracket and H is another functional, typically called the Hamiltonian, which acts to generate the system dynamics. The Poisson bracket is antisymmetric, $\{F,H\}=-\{H,F\}$, so that identifying the Hamiltonian with the total system energy then requires that $dH/dt=\{H,H\}=0$, thus guaranteeing the conservation of energy. Another key feature of the Poisson bracket is that any reversible dynamics, generated by H, will preserve the structure of the bracket for all times t. If F_t is defined as the time-dependent F, which is the solution of

$$\frac{dF_t}{dt}=\{F_t,H\}, \tag{8}$$

then the structure of the Poisson bracket is preserved for two arbitrary functionals when

$$\{F,G\}_t=\{F_t,G_t\}. \tag{9}$$

Taking the time derivative of this expression according to Eq. (8),

$$\{\{F,G\}_t,H\}=\{\{F_t,H\},G_t\}+\{F_t,\{G_t,H\}\}, \tag{10}$$

and then substituting Eq. (9) into this expression, after rearrangement, one obtains

$$\{\{G_t,F_t\},H\}+\{\{F_t,H\},G_t\}+\{\{H,G_t\},F_t\}=0. \tag{11}$$

This expression is known as the *Jacobi identity*. More details concerning Poisson brackets and their properties may be found in any textbook on classical mechanics.

The Poisson bracket plays a central role in the dynamics of all physical systems. Although it can only describe the reversible dynamics, it still contributes to dissipative systems when these are expressed in Hamiltonian form. This expression of dissipative dynamics in Hamiltonian form allows the extension of many of the benefits of classical Hamiltonian mechanics to real systems and thus this idea has attracted much attention in recent years. The product of this attention is the emergence of elegant and powerful formalisms for the description of dissipative systems [18–20]. For the present analysis, it must be realized that once one expresses the reversible dynamics of any system in the form of a bracket structure, it is required that this bracket possess the properties inherent to a Poisson bracket, i.e., antisymmetry and satisfaction of the Jacobi identity.

APPLICATION OF TIME-STRUCTURE INVARIANCE TO DYNAMIC PROBLEMS

In order to apply time-structure invariance to a particular problem, one must first recast it in Hamiltonian form. To accomplish this, one must determine the Poisson bracket corresponding to the reversible dynamics of the system under consideration, with reference to the antisymmetry property. This is not, in principle, difficult to do, as many forms of Poisson brackets for various physical systems have already been worked out [11,21–26]. Constraint equations may then be obtained by requiring satisfaction of the Jacobi identity (11).

In the following analysis, only the two examples mentioned above are considered, which are representative of many such constructions encountered in the various subfields of physics [27]. The Poisson bracket corresponding to the reversible dynamics [28] of the first example (suspension fluid mechanics) for two functionals $F[\mathbf{v},\mathbf{P}]$ and $H[\mathbf{v},\mathbf{P}]$ is

$$\begin{aligned}\{F,H\}=-\int [&F_{v_\gamma}H_{v_\beta}\nabla_\beta v_\gamma - H_{v_\gamma}F_{v_\beta}\nabla_\beta v_\gamma\\ &+F_{P_{\alpha\beta}}H_{v_\gamma}\nabla_\gamma P_{\alpha\beta}-H_{P_{\alpha\beta}}F_{v_\gamma}\nabla_\gamma P_{\alpha\beta}\\ &+P_{\gamma\beta}H_{P_{\alpha\beta}}\nabla_\gamma F_{v_\alpha}-P_{\gamma\beta}F_{P_{\alpha\beta}}\nabla_\gamma H_{v_\alpha}\\ &+P_{\gamma\alpha}H_{P_{\alpha\beta}}\nabla_\gamma F_{v_\beta}-P_{\gamma\alpha}F_{P_{\alpha\beta}}\nabla_\gamma H_{v_\beta}\\ &+2F_{P_{\alpha\beta}}R_{\alpha\beta\gamma\varepsilon}\nabla_\varepsilon H_{v_\gamma}-2H_{P_{\alpha\beta}}R_{\alpha\beta\gamma\varepsilon}\nabla_\varepsilon F_{v_\gamma}]d^3x.\end{aligned} \tag{12}$$

For the second example,

$$\begin{aligned}\{F,H\}=-\int [&F_{v_\gamma}H_{v_\beta}\nabla_\beta v_\gamma - H_{v_\gamma}F_{v_\beta}\nabla_\beta v_\gamma\\ &+F_{N_{\alpha\beta}}H_{v_\gamma}\nabla_\gamma N_{\alpha\beta}-H_{N_{\alpha\beta}}F_{v_\gamma}\nabla_\gamma N_{\alpha\beta}\\ &+N_{\gamma\beta}F_{N_{\alpha\beta}}\nabla_\alpha H_{v_\gamma}-N_{\gamma\beta}H_{N_{\alpha\beta}}\nabla_\alpha F_{v_\gamma}\\ &+N_{\gamma\alpha}F_{N_{\alpha\beta}}\nabla_\beta H_{v_\gamma}-N_{\gamma\alpha}H_{N_{\alpha\beta}}\nabla_\beta F_{v_\gamma}\\ &+H_{N_{\alpha\beta}}R_{\alpha\beta\gamma\varepsilon}\nabla_\varepsilon F_{v_\gamma}-F_{N_{\alpha\beta}}R_{\alpha\beta\gamma\varepsilon}\nabla_\varepsilon H_{v_\gamma}]d^3x,\end{aligned} \tag{13}$$

where the functionals F and H now depend on $\mathbf{v}$ and $\mathbf{N}$. For the present analysis, there is no need to specify the functionals F and H, except to recognize the Hamiltonian as H, which is the proper generator for the system dynamics (and hence $dH/dt=0$), and to define the Volterra derivatives appearing in Eqs. (12) and (13) in the proper manner [18]:

$$F_{v_\gamma}\equiv\frac{\delta F}{\delta v_\gamma},\quad F_{P_{\alpha\beta}}\equiv\frac{\delta F}{\delta P_{\alpha\beta}},\quad F_{N_{\alpha\beta}}\equiv\frac{\delta F}{\delta N_{\alpha\beta}}. \tag{14}$$

When $\mathbf{R}=\mathbf{0}$ in Eq. (12), that expression reduces to the Poisson bracket for a contravariant deformation tensor, which was discovered by Grmela [23] and derived from Hamilton's principle by Edwards and Beris [26]. Through the method of its derivation, it should retain both of the properties of a Poisson bracket, and this has been verified by direct substitutions [23,24]. When $\mathbf{R}=\mathbf{0}$ in Eq. (13), there results the Poisson bracket for a covariant deformation tensor, which was discovered in [24] and derived from Hamilton's principle in [18]. It too should satisfy the properties of a Poisson bracket through its construction, and this has been verified directly [24].

The brackets (12) and (13) were obtained by extending known results [11,18,23,24,26] to the present cases, where $\mathbf{R}$ is a function of $\mathbf{P}$ or $\mathbf{N}$, as consistent with the antisymmetry property of a Poisson bracket. This extension is unique and explicitly required for all of the reversible dynamics of the system to be described by a Poisson bracket. It may be verified that these brackets do indeed generate the evolution equations for the second moments (1) and (4) for a prototypical Hamiltonian [18]

$$H[\mathbf{v},\mathbf{A}]=\int\left(\frac{1}{2}\rho v_\gamma v_\gamma+h_0(\mathbf{A})\right)d^3x. \tag{15}$$

Through the antisymmetry property of the Poisson bracket, we find an immediate relationship between the evolution equation for the second moment and the reversible contributions to the kinematic properties of the fluids. By evaluating the evolution equation for the velocity vector field from each bracket, one can obtain explicit relationships for the reversible contributions to the extra stress tensor field involving $\mathbf{R}$:

$$\begin{aligned}\sigma_{\alpha\beta}&=2P_{\beta\gamma}H_{P_{\gamma\alpha}}-2R_{\gamma\varepsilon\alpha\beta}H_{P_{\gamma\varepsilon}},\\ \sigma_{\alpha\beta}&=-2N_{\alpha\gamma}H_{N_{\gamma\beta}}+R_{\gamma\varepsilon\alpha\beta}H_{N_{\gamma\varepsilon}},\end{aligned} \tag{16}$$

where σ is defined from the momentum equation

$$\rho\frac{\partial v_\alpha}{\partial t}=-\rho v_\beta\nabla_\beta v_\alpha-\nabla_\alpha p+\nabla_\beta\sigma_{\alpha\beta}, \tag{17}$$

p being the isotropic pressure and ρ the fluid mass density. Hence any closure approximation that is chosen for a particular second moment evolution equation must be incorporated into the stress tensor, according to expressions such as Eq. (16), in order to obtain an internally consistent prediction of kinematical properties.

It thus remains to examine the full forms of Eqs. (12) and (13) to find under what conditions the Jacobi identity is satisfied for each bracket via a direct substitution and subsequent elimination. Although this method is straightforward, it is quite tedious. The results of this calculation are two constraint equations, for each bracket, which place severe restrictions on the functionality of $\mathbf{R}$. These constraint equations are

$$R_{\alpha\beta\gamma\varepsilon}=P_{\eta\zeta}\frac{\partial R_{\alpha\beta\gamma\varepsilon}}{\partial P_{\eta\zeta}}, \tag{18}$$

$$\begin{aligned}&R_{\zeta\beta\eta\varepsilon}\delta_{\gamma\alpha}-R_{\varepsilon\beta\gamma\zeta}\delta_{\alpha\eta}+R_{\alpha\zeta\eta\varepsilon}\delta_{\beta\gamma}-R_{\alpha\varepsilon\gamma\zeta}\delta_{\eta\beta}+R_{\alpha\beta\eta\zeta}\delta_{\varepsilon\gamma}\\ &\quad-R_{\alpha\beta\gamma\varepsilon}\delta_{\zeta\eta}+P_{\varepsilon\rho}\frac{\partial R_{\alpha\beta\gamma\zeta}}{\partial P_{\eta\rho}}-P_{\zeta\rho}\frac{\partial R_{\alpha\beta\eta\varepsilon}}{\partial P_{\gamma\rho}}+P_{\rho\varepsilon}\frac{\partial R_{\alpha\beta\gamma\zeta}}{\partial P_{\rho\eta}}\\ &\quad-P_{\rho\zeta}\frac{\partial R_{\alpha\beta\eta\varepsilon}}{\partial P_{\rho\gamma}}+2R_{\theta\rho\gamma\zeta}\frac{\partial R_{\alpha\beta\eta\varepsilon}}{\partial P_{\theta\rho}}-2R_{\theta\rho\eta\varepsilon}\frac{\partial R_{\alpha\beta\gamma\zeta}}{\partial P_{\theta\rho}}=0\end{aligned} \tag{19}$$

for the first bracket (12) and Eq. (18), with $\mathbf{N}$ replacing $\mathbf{P}$, and

$$R_{\alpha\beta\eta\zeta}\delta_{\gamma\varepsilon}-R_{\alpha\beta\gamma\varepsilon}\delta_{\eta\zeta}+R_{\eta\beta\gamma\zeta}\delta_{\alpha\varepsilon}-R_{\gamma\beta\eta\varepsilon}\delta_{\alpha\zeta}+R_{\alpha\eta\gamma\zeta}\delta_{\beta\varepsilon}-R_{\alpha\gamma\eta\varepsilon}\delta_{\beta\zeta}+N_{\gamma\rho}\frac{\partial R_{\alpha\beta\eta\varepsilon}}{\partial N_{\zeta\rho}}-N_{\eta\rho}\frac{\partial R_{\alpha\beta\gamma\zeta}}{\partial N_{\varepsilon\rho}}+N_{\rho\gamma}\frac{\partial R_{\alpha\beta\eta\varepsilon}}{\partial N_{\rho\zeta}}-N_{\rho\eta}\frac{\partial R_{\alpha\beta\gamma\zeta}}{\partial N_{\rho\varepsilon}}+R_{\theta\rho\eta\varepsilon}\frac{\partial R_{\alpha\beta\gamma\zeta}}{\partial N_{\theta\rho}}-R_{\theta\rho\gamma\zeta}\frac{\partial R_{\alpha\beta\eta\varepsilon}}{\partial N_{\theta\rho}}=0 \quad (20)$$

for the second one (13).

The constraint relationship of Eq. (18) defines **R** as a homogeneous function of **N** or **P** of degree one, with all of the associated properties of such functions. This fact is potentially very useful for the illumination of restrictions on the form of **R** in general circumstances, as will be explored below. Expressions (18)–(20) are the fundamental results of this paper; in essence, by expressing **R** in a form compatible with these constraints, one can satisfy the time-structure invariance criterion of the reversible dynamics. Furthermore, any **R** that does not meet these requirements should be regarded with caution and as possibly being ill-formulated physically.

Now that the constraint equations have been obtained, one can apply them to the general form of a closure approximation, which is consistent with the Cayley-Hamilton theorem,

$$\begin{aligned}R_{\alpha\beta\gamma\varepsilon}=&\beta_{11}\delta_{\alpha\beta}\delta_{\gamma\varepsilon}+\beta_{12}\delta_{\alpha\gamma}\delta_{\beta\varepsilon}+\beta_{13}\delta_{\alpha\varepsilon}\delta_{\beta\gamma}+\beta_{21}\delta_{\alpha\beta}A_{\gamma\varepsilon}+\beta_{22}\delta_{\gamma\varepsilon}A_{\alpha\beta}+\beta_{23}\delta_{\beta\varepsilon}A_{\alpha\gamma}+\beta_{24}\delta_{\alpha\gamma}A_{\beta\varepsilon}+\beta_{25}\delta_{\beta\gamma}A_{\alpha\varepsilon}\\&+\beta_{26}\delta_{\alpha\varepsilon}A_{\beta\gamma}+\beta_{31}A_{\alpha\beta}A_{\gamma\varepsilon}+\beta_{32}A_{\alpha\gamma}A_{\beta\varepsilon}+\beta_{33}A_{\alpha\varepsilon}A_{\beta\gamma}+\beta_{41}\delta_{\alpha\beta}A^2_{\gamma\varepsilon}+\beta_{42}\delta_{\gamma\varepsilon}A^2_{\alpha\beta}+\beta_{43}\delta_{\beta\varepsilon}A^2_{\alpha\gamma}+\beta_{44}\delta_{\alpha\gamma}A^2_{\beta\varepsilon}\\&+\beta_{45}\delta_{\beta\gamma}A^2_{\alpha\varepsilon}+\beta_{46}\delta_{\alpha\varepsilon}A^2_{\beta\gamma}+\beta_{51}A_{\alpha\beta}A^2_{\gamma\varepsilon}+\beta_{52}A_{\gamma\varepsilon}A^2_{\alpha\beta}+\beta_{53}A_{\beta\varepsilon}A^2_{\alpha\gamma}+\beta_{54}A_{\alpha\gamma}A^2_{\beta\varepsilon}+\beta_{55}A_{\beta\gamma}A^2_{\alpha\varepsilon}\\&+\beta_{56}A_{\alpha\varepsilon}A^2_{\beta\gamma}+\beta_{61}A^2_{\alpha\beta}A^2_{\gamma\varepsilon}+\beta_{62}A^2_{\alpha\gamma}A^2_{\beta\varepsilon}+\beta_{63}A^2_{\alpha\varepsilon}A^2_{\beta\varepsilon},\end{aligned} \quad (21)$$

with $A^2_{\gamma\varepsilon}\equiv A_{\gamma\eta}A_{\eta\varepsilon}$. In this expression, the β_{ij}'s are scalar functions of the invariants of **A**. Various degrees of symmetrization imply various equalities between the β_{ij}'s appearing in Eq. (21). For example, if one enforces full symmetry, then it is clear that all of the β_{ij}'s are equal to each other for each given value of $i=1,\ldots,6$, In this case, there are only six arbitrary functions and the natural closure approximation of Verleye and Dupret [15] is obtained.

The application of constraint (18) to the closure (21) implies that one can immediately write down the allowed functionality of the β_{ij}'s for any valid closure,

$$\beta_{1j}=I_1 f_{1j}(x,y), \quad \beta_{2j}=f_{2j}(x,y),$$

$$\beta_{3j}=\frac{1}{I_1}f_{3j}(x,y), \quad \beta_{4j}=\frac{1}{I_1}f_{4j}(x,y), \quad (22)$$

$$\beta_{5j}=\frac{1}{I_1^2}f_{5j}(x,y), \quad \beta_{6j}=\frac{1}{I_1^3}f_{6j}(x,y),$$

$j=1,2,3$ or $1,\ldots,6$, where the f_{ij}'s are arbitrary functions of x and y, $I_1\equiv\mathrm{tr}\mathbf{A}$, $I_2\equiv\frac{1}{2}[(\mathrm{tr}\mathbf{A})^2-\mathrm{tr}(\mathbf{A}\cdot\mathbf{A})]$, and $I_3\equiv\det\mathbf{A}$ are the invariants of **A**, and $x\equiv I_2/I_1^2$, $y\equiv I_3/I_1^3$. The action of constraint (18) is to reduce 27 functions of three variables I_1, I_2, and I_3 to 27 functions of two variables x and y. Although not particularly limiting in a strict sense, the satisfaction of this constraint is straightforward to guarantee.

It is obvious that Eqs. (19) and (20) impose severe restrictions on the allowable functionality of the closure. Further progress depends on the exact definition of the degree of symmetrization assumed, as well as on the functionalities chosen for the f_{ij}'s. By substitution of the chosen form of Eq. (21) into the second constraint (19) or (20) and then equating to zero independently the various orders with respect to the second moment [29], additional restrictions are realized that relate the functions appearing in Eq. (22) to each other. As for the various closures that have been used during investigation of the two examples, one can draw some definite conclusions [30].

For the suspension example, Advani and Tucker [31] present seven different closures for Eq. (1) in tabular form, ranging from the closures of Hand [3] and Hinch and Leal [4] to their own hybrid form. Rather than repeat this table here, the reader is referred to [31], p. 373. Only the quadratic ($S1$) closure satisfies both constraints (18) and (19). In the few homogeneous flow fields tested thus far, only the quadratic closure and one other (the Advani-Tucker hybrid) have never displayed an aphysical behavior [31]. Thus it seems plausible that the more sophisticated investigations involving fitted approximations [15,16] would benefit by consideration of the constraints imposed upon the closure by time-structure invariance.

The closure approximation introduced by Doi and Ohta [10] for the fourth moment in their evolution equation for the second moment is

$$R_{\alpha\beta\gamma\varepsilon}=\frac{1}{\mathrm{tr}\mathbf{N}}N_{\alpha\beta}N_{\gamma\varepsilon}. \quad (23)$$

This closure satisfies both constraints imposed by the Jacobi identity and hence Eq. (23) is dynamically consistent with time-structure invariance. This is in accord with our experience since it is known that this closure gives a reasonably good approximation of the fourth moment and is well behaved physically, at least in shear flow [10]. However, it now becomes clearer what types of extensions of Eq. (23) are allowable in order to obtain, we hope, a more accurate approximation, as described below.

As alluded to in [28], one must not confuse the closure approximation of Eq. (23), which is purely convective, with a similar one, common in liquid-crystalline theories, that arises through the coupling of the constant length constraint from the rigid objects and dissipative effects associated with the rotational diffusivity [32]. This latter closure approximation is known to cause a suppression of the rich dynamical behavior of liquid-crystalline materials that is inherent in the evolution equation for the distribution function [33]. However, it is evident that this suppression of dynamical behavior is due solely to the dissipative effects on the closure. The constraints discussed herein do not apply to closures with dissipative contributions and no conclusions can be reached concerning them in this analysis. These dissipative closures must be addressed as discussed in [28].

In order to examine some more general forms of closure approximations one may concentrate on the second example (the Doi-Ohta theory), with constraints (18) and (20). The functional dependences of the f_{ij}'s are now not specified prior to the application of the constraints, but it is still necessary to choose a particular degree of symmetrization for the closure of Eq. (21). Three particular cases of symmetrization will be examined here, ranging from a low form of symmetrization to full symmetrization. Due to the inherent symmetry of the second-rank tensor **N**, a low form of symmetry for a general closure is $R_{\alpha\beta\gamma\varepsilon}=R_{\beta\alpha\gamma\varepsilon}=R_{\alpha\beta\varepsilon\gamma}$, a simple example of Eq. (21) for this particular case being

$$R_{\alpha\beta\gamma\varepsilon}=I_1 f_1 \delta_{\alpha\beta}\delta_{\gamma\varepsilon}+f_2\delta_{\alpha\beta}N_{\gamma\varepsilon}+\frac{1}{I_1}f_3 N_{\alpha\beta}N_{\gamma\varepsilon} +\frac{1}{I_1}f_4\delta_{\alpha\beta}N^2_{\gamma\varepsilon}+\frac{1}{I_1^2}f_5 N_{\alpha\beta}N^2_{\gamma\varepsilon}+\frac{1}{I_1^3}f_6 N^2_{\alpha\beta}N^2_{\gamma\varepsilon}, \tag{24}$$

where all β_{ij}, $j\neq 1$, are taken as zero [34]. Constraint (20) can now be applied to this closure and a set of constraint equations can be derived by equating to zero the various orders of the tensor **N** appearing in the resulting expression. These equations give explicit relationships between the f_i's appearing in Eq. (24). When applying this procedure, it is necessary to reduce to lower order all third and higher moments using the Cayley-Hamilton theorem, as mentioned above. Furthermore, since for general functions one does not know *a priori* the order of the derivatives of the f_i's with respect to **N**, these must be assumed to possess a general form as well:

$$\frac{\partial f_i}{\partial N_{\alpha\beta}}=\Psi_i\delta_{\alpha\beta}+\Xi_i N_{\alpha\beta}+\Omega_i N_{\alpha\gamma}N_{\gamma\beta} \quad \text{for all } i. \tag{25}$$

In these expressions, Ψ_i, Ξ_i, and Ω_i must also be evaluated using the constraint equations derived in the above-stated manner, as well as the additional constraints

$$\frac{\partial f_i}{\partial N_{\alpha\beta}}N_{\alpha\beta}=0 \quad \text{for all } i, \tag{26}$$

$$1=9f_1+3f_2+f_3+3f_4(1-2x)+f_5(1-2x)+f_6(1-2x)^2, \tag{27}$$

the former arising automatically from the realization that the f_i's are functions of x and y and the latter from the normalization condition [35].

For the closure of Eq. (24), the constraint relationships derived are

$$\Xi_3+\frac{2}{I_1^2}f_5-\frac{1}{I_1}\Psi_5=0, \tag{28a}$$

$$I_1\Psi_5+(I_1^2-2I_2)\Xi_5=0, \tag{28b}$$

$$I_1\Psi_3+(I_1^2-2I_2)\Xi_3=0, \tag{28c}$$

$$f_3+(1-2x)f_5=1, \tag{28d}$$

with all other f_i, Ψ_i, Ξ_i, and Ω_i required to vanish. These constraints define the allowed functionality of the remaining f_i's, and since the four equations involve six unknowns, in general they cannot be solved without additional physical requirements. However, since both Ω_3 and Ω_5 vanish, f_3 and f_5, according to Eq. (25), cannot depend upon the determinant, requiring that these entities are functions of x only [36]. One would be tempted to use higher-order normalization conditions [37] to obtain additional constraint equations, thus closing the system, but these turn out always to involve y, which is no longer included in the allowed functionality of the remaining f_i's. Hence the net result of the application of the constraint (20) to the closure (24) is the reduction of six functions of two variables to two functions of one variable, with four explicit relationships between the remaining f_i's that must be satisfied. In fact, by taking the derivative of the normalization constraint (28d) with respect to $N_{\alpha\beta}$ and subsequently using Eq. (28b), it is straightforward to show that any two functions satisfying the normalization constraint will automatically satisfy the remaining constraint equations. This implies that only the normalization condition affects the values of f_3 and f_5 and that only a single additional constraint condition is required in order to determine both functions uniquely. In the absence of the normalization condition, the first three constraints of Eqs. (28) represent the more general functionality allowed from consideration of time-structure invariance alone.

One can also examine two limits where the constraint equations reduce to a closed set, arising when either remaining f_i is set equal to zero. When $f_5=0$, f_3 is required to be a constant and this constant is required from the normalization condition to be unity. This is the closure approximation introduced by Doi and Ohta [10]. When f_3 is set equal to zero, $f_5=1/(1-2x)$, which is consistent with all of the constraints (28) imposed by the Jacobi identity.

The next closure approximation to be examined possesses an intermediate degree of symmetry, as dictated by Eq. (6):

$$R_{\alpha\beta\gamma\varepsilon}=I_1 f_1\delta_{\alpha\beta}\delta_{\gamma\varepsilon}+f_2(\delta_{\alpha\beta}N_{\gamma\varepsilon}+N_{\alpha\beta}\delta_{\gamma\varepsilon})+\frac{1}{I_1}f_3 N_{\alpha\beta}N_{\gamma\varepsilon} +\frac{1}{I_1}f_4(\delta_{\alpha\beta}N^2_{\gamma\varepsilon}+N^2_{\alpha\beta}\delta_{\gamma\varepsilon})+\frac{1}{I_1^2}f_5 \times(N_{\alpha\beta}N^2_{\gamma\varepsilon}+N^2_{\alpha\beta}N_{\gamma\varepsilon})+\frac{1}{I_1^3}f_6 N^2_{\alpha\beta}N^2_{\gamma\varepsilon}. \tag{29}$$

This expression results from Eq. (21) by assuming that all $\beta_{ij}=0$, except $\beta_{11},\beta_{31},\beta_{61},\beta_{21}=\beta_{22}$, $\beta_{41}=\beta_{42}$, and $\beta_{51}=\beta_{52}$. Applying constraint (20) to this closure using the procedure described above, one can calculate that the only allowable nonzero function f_i is f_3, which must be equal to a constant. Hence, in this case, the net result of applying Eq. (20) to Eq. (29) is that six functions of two variables are reduced to a single arbitrary constant. Again, the normalization condition can be used to assign this constant the value of unity, for which Eq. (23) results.

The last case to examine is that of full symmetry, which means that **R** is invariant to any permutations of the component indices. The closure approximation is now that which is expressed by Eq. (21) with all the β_{ij}'s being equal to β_i for each value of $i=1,\ldots,6$. In this case, one can calculate, with the aid of a symbolic manipulator, that all of the f_i's must vanish. It is now apparent that as the degree of symmetrization increases, the flexibility in the choice of the functionality of the f_i's decreases. This is not surprising since it is evident that an increased degree of symmetry acts to restrict the functionality of the general closure (21). Any further restrictions imposed on the system by time-structure invariance can only do the same. Once full symmetry is obtained, the restrictions placed upon the closure approximation (for this particular example) by time-structure invariance become so severe as to allow no consistent closure whatsoever.

At first glance the above-stated observation seems to contradict physical intuition and to invalidate some well-defined probability distributions. For example, the fourth moment of a Gaussian distribution may be expressed exactly in terms of the second moments as

$$Z_{\alpha\beta\gamma\varepsilon}=N_{\alpha\beta}N_{\gamma\varepsilon}+N_{\alpha\gamma}N_{\beta\varepsilon}+N_{\alpha\varepsilon}N_{\beta\gamma}. \tag{30}$$

It must be realized, however, that the fourth moment does not enter into the intrinsic equations of motion of the system, so that the fact that all of the arbitrary functions are zero is entirely consistent with the Gaussian distribution.

For other distributions in the present example, it should be recognized that the full symmetry of **R** may need to be sacrificed in order to obtain a closure consistent with other physical requirements on the system. There seems to be a price to be paid for reducing a fourth-rank tensor (with a higher-order of symmetry) to a product of second-rank tensors (with lower-order symmetry). It is not conclusively clear at this point whether or not full symmetry is more or less important than the other physical requirements. However, as mentioned earlier when discussing the suspension example, many of the closures that respected other physical criteria (including the symmetries) of the fourth moment exhibited an aphysical behavior under some circumstances, whereas the only closure that respected time-structure invariance was always well behaved [31]. This provides one crude indication that respecting time-structure invariance may be the most important of the various physical requirements. Furthermore, as evident from the Doi-Ohta evolution equations, the different components of the fourth moment (and hence the closure) play very different roles in the evolution equations for the components of the second moment; this implies again that full symmetry might not be a strong physical requirement for the closure.

The problem with symmetrization and time-structure invariance overconstraining the problem may be endemic to the simplicity of the case (the Doi-Ohta equation) examined. For the more complicated case of a fluid suspension, it may turn out that the presence of a non-unit shape factor could impart an extra degree of freedom to the analysis, thus rendering feasible full compatibility with all of the constraints that are now accepted on the functionality of the closure. Hence the trade-off between normalization, degree of symmetrization, and time-structure invariance may only be an issue for simple systems such as the Doi-Ohta case.

The final issue to be addressed in this article is to try to make some deductions as to what are some additional equations that can be used to close the set of constraints imposed by normalization and the Jacobi identity. Unfortunately, this issue must remain largely unsolved, but help may be offered in general form. For instance, if the system is compressible, one may consider an isotropic expansion (for which **N** is diagonal, with all nonzero components being equal, i.e., $\mathbf{N}=I_1\boldsymbol{\delta}/3$) by taking the trace of the evolution equation for **N**, Eq. (4), with the closure of Eq. (24):

$$\frac{\partial N_{\alpha\alpha}}{\partial t}=-2N_{\gamma\varepsilon}\nabla_\gamma v_\varepsilon+f_3N_{\gamma\varepsilon}\nabla_\gamma v_\varepsilon+\frac{f_5}{N_{\alpha\alpha}}N_{\gamma\eta}N_{\eta\varepsilon}\nabla_\gamma v_\varepsilon. \tag{31}$$

For an isotropic expansion, it is required that

$$\frac{\partial N_{\alpha\alpha}}{\partial t}=-\frac{N_{\alpha\alpha}}{3}\nabla_\gamma v_\gamma, \tag{32}$$

which adds another constraint to the set of equations (28):

$$f_3+\tfrac{1}{3}f_5=1. \tag{33}$$

This expression, coupled with the normalization condition, requires that $f_5=0$ and $f_3=1$. Of course, for an incompressible fluid system this constraint on the evolution equation for the second moment is not required. Perhaps other physically meaningful constraints may be constructed in a similar manner.

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[27] In this analysis, the normalization constraint $\mathrm{tr}\mathbf{P}=1$ is not imposed in order to keep the analysis as general as possible, i.e., applicable to a general, as opposed to a constrained, deformation tensor.

[28] The term multiplied by the rotational diffusivity in Eq. (1) is a dissipative term, which therefore will not show up in the Poisson bracket. It may be included in the dynamical description of the system through a dissipation bracket [18], but presently will be ignored as being extraneous to the main issue. In reference to this, one must now realize the limitations on the constraints proposed in this article. Any higher-moment averages appearing as a result of dissipative forces, such as the Brownian force, are not required to possess the structure embodied in the Poisson bracket. These higher moments will appear in the dissipation bracket, and constraints upon them will arise through Onsager-Casimir reciprocity, material objectivity, the entropy inequality, and so on.

[29] Note that higher-order terms, such as $\mathbf{P}\cdot\mathbf{P}\cdot\mathbf{P}$, must be reduced to lower-order ones, via the Cayley-Hamilton theorem, when applying this procedure (see below).

[30] Strictly speaking, the constraint equations (18) and (19) should not be applied to constrained systems such as the first example where $\mathrm{tr}\mathbf{P}=1$. Alternative constraints should be derived using the constrained version of the Poisson bracket which arises under a suitable transformation, as developed by Edwards, Beris, and Grmela [18,25]. Such an action entails an excessive degree of tedium. Nevertheless, applying Eqs. (18) and (19) to a constrained system might still produce the correct restrictions provided one sets $\mathrm{tr}\mathbf{P}=1$ only after the constraint equations have been applied fully. Assuming this action to be reasonable, the approximations used to close the evolution equations for the second moment of particle suspensions may be evaluated based upon the constraints (18) and (19).

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[35] This equation is obtained by taking the trace of $\mathbf{R}$, $R_{\alpha\alpha\gamma\varepsilon}$, and multiplying it by $\delta_{\gamma\varepsilon}$.

[36] Note that as long as f_3 and f_5 are functions of x only, the middle two constraints are satisfied automatically.

[37] For instance, by multiplying the trace of $\mathbf{R}$ by $N_{\gamma\varepsilon}$.