Nonlinear viscosity and Grad's method

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The Grad ten-moment approximation (no heat flux) is analyzed for cylindrical symmetry in a stationary situation in which the gradients of the fluxes are assumed to be small. We show that if the collision term in the transport equation, resulting from the ten-moment approximation, is linearized in the fluxes, we can obtain a viscosity (η_l) that depends on the gradient of the velocity with the correct limiting behavior for small gradients. The nonlinear contribution of the fluxes to the collision term are then taken into account to derive an expression for the viscosity (η_{nl}) as a function of the gradient of the velocity. A comparison between η_l and $\eta_{\rm nl}$ is performed finding that the maximum percentage deviation between them is 0.52% when the gradient of the hydrodynamic velocity is positive, but when the gradient is negative the situation changes dramatically. $[S1063-651X(99)00410-9]$

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I. INTRODUCTION

Shear rate-dependent viscosities have been obtained by many authors for specific situations, mainly nonstationary states in simple fluids. There is, for instance, the exact solution to the Boltzmann equation for Maxwellian molecules given by Truesdell and co-workers [1]. Monte Carlo calculations, molecular dynamics calculations, and solutions to the Bhattangar, Gross, and Krook (BGK) equations for rigid spheres have also been discussed in detail in many papers $[2-5]$ where a comparison with Truesdell's work can be found. The concept of nonlinear viscosity (shear ratedependent viscosity) has a long history; for example, Gilbarg and Paolucci $[6]$ made some considerations of this phenomenon in their study of shock waves. A more comprehensive set of references can be found in the works by Truesdell [7].

More recently Karlin *et al.* [8] and Gorban and Karlin [9] have used Grad's method to reach similar conclusions such as those mentioned in the preceding paragraph, for nonstationary situations, but with a longitudinal flow instead of a shear one. Also, Al-Ghoul and Eu $[10]$ have derived explicit expressions for the nonlinear viscosity using a modified moment method and their expressions have been applied to different situations such as the shock wave problem $[11]$. These works can be difficult to understand in part because of their use of the same terminology as other researchers to denote different things; for example, the Rankine-Hugoniot equations according to Al-Ghoul and Eu $[11]$ are the integral form of the conservation equations and not the equations relating the two equilibrium states as is standard. In the recent book by Eu $[10]$ the reader can find many references to the nonlinear viscosity issue as implemented by Eu and collaborators.

While there is no doubt that viscosities that depend on the gradients of the hydrodynamic velocity $[u(\mathbf{r},t)]$ can be obtained from Boltzmann's equation $[1-5,8-10]$, a phenomenon which we refer to as nonlinear viscosity, there are two main points which we would like to address in this work. First, we consider stationary situations using Grad's method. Second, we consider the nonlinear contribution of the fluxes to the collisions. As we will see, there are situations in which, if these nonlinear contributions are not included, the results can be in serious error. We will show that in the usual procedure in which the transport coefficients are reanalyzed in a somewhat more systematic way, there is a dependence of the viscosity on the longitudinal deformation rate $\left[\partial \mathbf{u}_x(\mathbf{r},t)/\partial x\right]$. This is done for Grad's ten-moment equations but avoiding the use of the invariance principle stated by Karlin *et al.* $|8|$.

In order to simplify the calculations and to make the point in the simplest way some restrictions are convenient. The first one is that we can take the heat flux to be zero, a point which is not trivial and certainly deserves more attention. The second one is that the velocity has only one component and does not depend on time $\mathbf{u}(\mathbf{r}) = u(x)\mathbf{i}$. As a specific situation the reader may think of a one dimensional stationary shock wave, where he should, however, notice that in this case the heat flux cannot be taken to be zero since there is a temperature inhomogeneity. We finally assume cylindrical symmetry of the distribution function, a restriction which is mainly a matter of convenience to reduce the expressions and definitions.

II. THE DISTRIBUTION FUNCTION.

We use the expansion of the single particle distribution function in terms of Hermite polynomials and the approximation given by Grad $[12–14]$ in the ten-moment approximation, the weight function being given by the local Maxwellian distribution function $f^{(0)}$:

$$
f^{(0)}(\mathbf{c}, \mathbf{r}, t) = n \left(\frac{m}{2 \pi kT} \right)^{3/2} \exp(-m \mathbf{C}^2 / 2kT), \quad (1)
$$

where **c** is the atomic velocity, *k* Boltzmann's constant, *T* the temperature, *n* the number density, *m* the mass, and $C \equiv c$ $-\mathbf{u}$ the peculiar velocity [13].

The distribution function can be expressed as $[12]$

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$$
f = f^{(0)}(1 + \xi), \tag{2}
$$

where, under cylindrical symmetry,

$$
\xi = \mu_{xx} \left(\mathbf{C}_x^2 - \frac{kT}{m} \right) + \mu_{xy} (\mathbf{C}_x \mathbf{c}_y + \mathbf{C}_x \mathbf{c}_z)
$$

+
$$
\mu_{yy} \left((\mathbf{c}_y^2 + \mathbf{c}_z^2) - \frac{2kT}{m} \right) + \mu_{yz} \mathbf{c}_y \mathbf{c}_z,
$$
 (3)

and Grad's moments can also be written in terms of quantities with a well defined physical meaning, such as the components of the pressure tensor, which are abbreviated as

$$
\mu_{xx} = \frac{m}{2kT} \left(\frac{\mathbf{P}_{xx}}{nkT} - 1 \right),\tag{4}
$$

$$
\mu_{xy} = \frac{m\mathbf{P}_{xy}}{nk^2 T^2},\tag{5}
$$

$$
\mu_{yy} = \frac{m}{2kT} \left(\frac{\mathbf{P}_{yy}}{nkT} - 1 \right),\tag{6}
$$

$$
\mu_{yz} = \frac{mP_{yz}}{nk^2 T^2}.
$$
\n(7)

In these equations **P** is the pressure tensor which is well known to be defined by

$$
\mathbf{P} = \int f(\mathbf{c}, \mathbf{r}, t) m \mathbf{C} \mathbf{C} d\mathbf{C}.
$$
 (8)

The pressure *p* is given by $nkT = (P)/3$, as it should be for a structureless gas in the dilute regime. This definition together with Eqs. (4) and (6) gives a relation between two of the moments, namely,

$$
\mu_{xx} = -2\mu_{yy} \,. \tag{9}
$$

III. THE BOLTZMANN EQUATION

In order to obtain the transport equation let us start with the behavior of the distribution function which is given by the Boltzmann equation,

$$
\mathcal{D}(f) \equiv \frac{\partial f(\mathbf{r}, \mathbf{c}, t)}{\partial t} + \mathbf{c} \cdot \nabla_r f(\mathbf{r}, \mathbf{c}, t) = J(f, f), \qquad (10)
$$

where $J(f, f)$ is the well known collision term [15].

The transport equation is obtained by multiplying the Boltzmann equation by any function of the velocity $\Psi(\mathbf{c})$ and integrating over **c**:

$$
\int d\mathbf{c} \mathcal{D}(f) \Psi(\mathbf{c}) = \int d\mathbf{c} J(f, f) \Psi(\mathbf{c}). \tag{11}
$$

In particular, when we take $\Psi(\mathbf{c}) = (3m/2)\mathbf{C}_x^2$ in Eq. (11) we obtain that for the stationary case [see Eq. $(A7)$],

$$
\frac{\partial}{\partial x} \{ [\mathbf{P}_{xx}(x) - \mathbf{P}_{yy}(x)] u(x) \}
$$

$$
+ 2 \mathbf{P}_{xx}(x) \frac{\partial u(x)}{\partial x} = \int d\mathbf{c} \frac{3m}{2} \mathbf{C}_{x}^{2} J(f, f). \quad (12)
$$

The right hand side of Eq. (12) gives the contribution of the collision term and its calculation is performed by direct substitution of the distribution function given by Eq. (2) . As a result we obtain a bilinear expression in the fluxes $(\mu_{xx}, \mu_{xy}, \mu_{yz})$ which can be linearized. The approximation implied by this method corresponds to a situation in which the Maxwellian form is rather close to the distribution function. When we take the hard sphere model, the collisional term in Eq. (12) is expressed as follows:

$$
\int d\mathbf{c} \frac{3m}{2} \mathbf{C}_x^2 J(f, f) = \frac{3m}{2} \mu_{xx} \Xi,
$$
 (13)

where Ξ is a collision integral whose value for the rigid sphere of diameter σ is [15]

$$
\Xi = \left[\mathbf{C}_x^2; \mathbf{C}_x^2 - \frac{1}{2} (\mathbf{c}_y^2 + \mathbf{c}_z^2) \right] = -\frac{32}{5} \sigma^2 n^2 \sqrt{\pi} \left(\frac{kT}{m} \right)^{5/2}.
$$
\n(14)

The term in square brackets in Eq. (14) is defined for two arbitrary functions of the velocities, say $\Phi(\mathbf{c})$ and $\Theta(\mathbf{c})$, as

$$
\begin{aligned} [\Theta(\mathbf{c}); \Psi(\mathbf{c})] \\ &= \int d\mathbf{c}d\mathbf{c}_1 d\mathbf{e} \Sigma(\chi, g) g f^{(0)}(\mathbf{c}, \mathbf{r}, t) f^{(0)}(\mathbf{c}_1, \mathbf{r}, t) \\ &\times \Theta(\mathbf{c}) \Delta[\Psi(\mathbf{c})], \end{aligned} \tag{15}
$$

where the shorthand notation $\Delta[\Psi(\mathbf{c})]$ means

$$
\Delta[\Psi(\mathbf{c})] = [\Psi(\mathbf{c}') + \Psi(\mathbf{c}'_1)] - [\Psi(\mathbf{c}) + \Psi(\mathbf{c}_1)], \quad (16)
$$

 $\Sigma(\chi, g)$ is the differential cross section, g the magnitude of the relative velocity, *d***e** denotes an integration over the solid angle, and the velocities correspond to their values before and after the collision (primes) $[15]$. The collision integrals that multiply the other fluxes are zero or are neglected according to the linear approximation in the fluxes. Notice also that use of Eq. (9) was made in order to express the linear contribution of μ_{yy} to the collision term only in terms of μ_{xx} .

From Eqs. (12) and (13) we conclude that

$$
\frac{\partial}{\partial x}\{[\mathbf{P}_{xx}(x) - \mathbf{P}_{yy}(x)]u(x)\} + 2\mathbf{P}_{xx}(x)\frac{\partial u(x)}{\partial x} = \frac{3m}{2}\mu_{xx}\Xi,
$$
\n(17)

which in fact can be seen as a constitutive relation for the nonvanishing components of the pressure tensor. If, following Grad $[12]$, we substitute in the left hand side of Eq. (17) the values for the pressure tensor calculated with the Maxwellian ($\mathbf{P}_{xx} = n kT$, $\mathbf{P}_{yy} = n kT$) we obtain, after using Eq. (4) , the result that

$$
\mathbf{P}_{xx} = n kT - \frac{4}{3} \eta_{\text{NS}} \frac{\partial u(x)}{\partial x},\tag{18}
$$

where η_{NS} is the well known Navier-Stokes expression [15] for the shear viscosity:

$$
\eta_{\rm NS} = \frac{5}{16} \left(\frac{mkT}{\pi} \right)^{1/2} / \sigma^2.
$$
 (19)

We now show how a longitudinal deformation rate $(\partial u/\partial x)$ dependence of the viscosity can be obtained from Grad's linear transport equations, Eq. (17) , using a different approximation. For this purpose we assume that we can neglect the gradients of the fluxes, namely, if, for example, the pressure tensor is a function of $\partial u(x)/\partial x$ but not of the higher order derivatives, then the approximation amounts to a situation in which the second order derivatives of the velocity or the products of the gradients of the normal variables can be neglected. Since

$$
\mathbf{P}_{xx} - \mathbf{P}_{yy} = \frac{3}{2} (\mathbf{P}_{xx} - nkT) \tag{20}
$$

is a flux, we obtain from Eqs. (12) and (13) that

$$
\frac{3}{2}(\mathbf{P}_{xx} - nkT)\frac{\partial u}{\partial x} + 2\mathbf{P}_{xx}\frac{\partial u}{\partial x} = \frac{3m}{2}\mu_{xx}\Xi.
$$
 (21)

It is convenient to introduce a dimensionless pressure tensor $\widetilde{\mathbf{P}}_{xx}$ and a reduced longitudinal deformation rate a_i^* as

$$
\widetilde{\mathbf{P}}_{xx} = \frac{\mathbf{P}_{xx}}{nkT}, \quad a_l^* \equiv \frac{1}{\nu} \frac{\partial u(x)}{\partial x}, \tag{22}
$$

where ν is an effective collision frequency defined by

$$
\nu \equiv \frac{n k T}{\eta_{\rm NS}}.\tag{23}
$$

The reduced form of Eq. (21) is then given by

$$
(\widetilde{\mathbf{P}}_{xx}-1)a_t^{\star} + \frac{4}{3}\widetilde{\mathbf{P}}_{xx}a_t^{\star} = -(\widetilde{\mathbf{P}}_{xx}-1),\tag{24}
$$

where the expression for Ξ given by Eq. (14) has been used. Equation (24) can be easily solved to yield

$$
\widetilde{\mathbf{P}}_{xx} = \frac{(1 + a_t^*)}{(1 + 7a_t^*/3)},\tag{25}
$$

which for small a_l^* has the following behavior:

$$
\widetilde{\mathbf{P}}_{xx} = (1 + a_l^{\star}) [1 - 7 a_l^{\star} / 3 + O(a_l^{\star 2})] = [1 - \frac{4}{3} a_l^{\star} + O(a_l^{\star 2})].
$$
\n(26)

Equation (26) shows that we recover the correct limiting case, corresponding to the reduced form of Eq. (18) . However, Eq. (25) is clearly more general than its limiting case. From the derivation of this equation one should expect it to be valid for small a_l^* , yet its range can be extended at least a little bit more than the linear term in a_l^* . Indeed, from it we can derive the region of longitudinal rates for which the Navier-Stokes regime (no longitudinal rate dependence for the viscosity) is valid to a certain percentage. For example, a percentage difference of 5% between the reduced pressure tensor given by Eq. (25) and the reduced pressure tensor at the Navier-Stokes regime (\tilde{P}^{NS}) where P^{NS} given by Eq. (18) can be seen to be valid when $a_l^* \in (-0.11, 0.13)$. On the other hand, it should be pointed out that the results just given were obtained under the assumption that

$$
Q = \frac{u(x)(\partial/\partial x) [\mathbf{P}_{xx} - p(x)]}{[\mathbf{P}_{xx} - p(x)][\partial u(x)/\partial x]} \ll 1.
$$
 (27)

Since from Eq. (25) we can obtain an explicit expression for P_{xx} , namely,

$$
\mathbf{P}_{xx} = p(x) \frac{3p(x) + 3 \eta_{\text{NS}}(x) [\partial u(x)/\partial x]}{3p(x) + 7 \eta_{\text{NS}}(x) [\partial u(x)/\partial x]},
$$
(28)

where η_{NS} is given by Eq. (19), we can evaluate the ratio *Q* defined by Eq. (27) using Eq. (28) to get

$$
Q = 6 \frac{u(x)p(x)[\partial p(x)/\partial x]}{g_1(x)g_2(x)[\partial u(x)/\partial x]} + 3 \frac{u(x)p(x)[\partial \eta_{\text{NS}}(x)/\partial x]}{g_1(x)g_2(x)} + 3 \frac{u(x)p(x)p(x)p_{\text{NS}}(x)[\partial^2 u(x)/\partial x^2]}{g_1(x)g_2(x)[\partial u(x)/\partial x]} + 3 \frac{u(x)p(x)[\partial p(x)/\partial x]}{g_1(x)g_2(x)} - 9 \frac{u(x)p(x)^2[\partial p(x)/\partial x]}{g_1^2(x)g_2(x)[\partial u(x)/\partial x]} - 21 \frac{u(x)p(x)^2[\partial \eta_{\text{NS}}(x)/\partial x]}{g_1^2(x)g_2(x)} - 21 \frac{u(x)p(x)^2 \eta_{\text{NS}}(x)[\partial^2 u(x)/\partial^2 x]}{g_1^2(x)g_2(x)[\partial u(x)/\partial x]} - 9 \frac{u(x)p(x)p_{\text{NS}}(x)[\partial p(x)/\partial x]}{g_1^2(x)g_2(x)} - 21 \frac{u(x)p(x)p_{\text{NS}}(x)[\partial p(x)/\partial x][\partial \eta_{\text{NS}}(x)/\partial x]}{g_1^2(x)g_2(x)} - 21 \frac{u(x)p(x)p_{\text{NS}}(x)[\partial^2 u(x)/\partial^2 x]}{g_1^2(x)g_2(x)} - \frac{3}{2} \frac{u(x)[\partial p(x)/\partial x]}{g_2(x)[\partial u(x)/\partial x]},
$$
\n(29)

where

$$
g_1 = 3p(x) + 7 \eta_{\text{NS}}(x) \frac{\partial u(x)}{\partial x},
$$

$$
g_2 = 3 \frac{p(x)^2}{g_1(x)} + 3 \frac{p(x) \eta_{\text{NS}}(x) [\partial u(x)/\partial x]}{g_1(x)} - \frac{3}{2} p(x).
$$
(30)

From Eqs. (29) and (30) we conclude that our approximation, $Q \ll 1$, holds when $|\partial u / \partial x|$ is very large but the quantities *u*, *p*, *T*, $\partial u/\partial x$, $\partial p/\partial x$, $\partial T/\partial x$, and $\partial^2 u/\partial x^2$ remain finite. These conditions are similar to the ones used by Gorban and Karlin [9] and Karlin *et al.* [8] with which they were able to sum a subseries of the Chapman-Enskog higher order gradient expansion using a direct approach $\lfloor 8 \rfloor$ together with the principle of the invariant manifold $[9]$.

We now compare with the results mentioned in the Introduction but notice that they were obtained for a different situation and thus, in principle, there is no reason why they should correspond. Nevertheless the comparison should at least exhibit whether the order of magnitude and the general trend are correct. Also, we know that in the Chapman-Enskog expansion the transport coefficients are independent of the particular flow considered so that the comparison will be performed at the level of the viscosity. The question now is, how do we define the viscosity when the reduced pressure tensor is given by Eq. (25) ? The natural way of doing this is by assuming a relation of the form (18) with $\eta_{\rm NS}$ replaced by a certain new viscosity η_l . Doing this we obtain for the reduced viscosity (η_l^{\star})

$$
\eta_l^* = \frac{\eta_l}{\eta_{\rm NS}} = \frac{1}{1 + 7a_l^*/3}.\tag{31}
$$

Equation (31) gives for $a_l^* = 1$ the value $\eta_l^* = 0.3$, whereas for the nonstationary shear rate situation considered by other authors $[1,3-5]$ the value for the reduced viscosity, which we denote by η_S^* , is about 0.53 for $a_S^* = 1$ (shear rate situation). The asymptotic behavior of η_l^* for large a_l^* is also different; for the nonstationary situation considered by them, η_s^* goes as $a_{\mathcal{S}}^{\star^{-4/3}}$ while our expression predicts an $a_{l}^{\star^{-1}}$ dependence, so the quantitative behavior for the reduced viscosity is different. We notice further from Eq. (25) that for $a_i^* = 1$ we have \tilde{P}_{xx} =0.6 and so the linear approximation of the collision term, consisting in neglecting the nonlinear contribution of the fluxes, may certainly be questioned since \tilde{P}_{xx} is not near 1. In order to see if the discrepancy may be due to the linear approximation in the collision term we have undertaken some calculations considering the full expression of the collision term.

In the situation considered by Karlin *et al.* [8,9], for a nonstationary longitudinal flow, they obtained for the Maxwell model the value $\eta_l^* = 0.478$ for $a_l^* = 1$ which is different from our result and also from Truesdell's value. The asymptotic behavior of the reduced viscosity for large longitudinal rates can be shown to go as $a_l^{\star-1}$, which is again different from Truesdell's result but is in agreement with our results. It is important to notice that the stationary situation can be obtained from their equation (10) [8], valid for soft spheres, and that the result for the reduced viscosity is in agreement with our results for the case of rigid spheres. We have evaluated the viscosity factor for the nonstationary longitudinal flow considered by Karlin *et al.* [8] for the rigid sphere model following Uribe and Pi \widetilde{na} [16,17] finding that $\overline{\eta}_l^*$ = 0.4781 for a_l^* = 1 which should be compared with the value obtained from Eq. (31), $\eta_l^* = 0.3$ for $a_l^* = 1$.

Finally, we mention that $\partial u(x)/\partial x$ can be positive or negative, that is, a_l^* can be positive or negative. For negative values of a_l^* Eq. (31) predicts a divergence of the reduced viscosity at $a_l^* = -3/7$ and for values of a_l^* less than $-3/7$ the reduced viscosity is negative. Thus, this region is expected to be one in which the nonlinear contributions should be important since negative viscosities are certainly considered nonphysical, an expectation that is confirmed with the results of the next section.

IV. THE NONLINEAR TERMS

We now consider the nonlinear terms in the fluxes, implying that for the rigid sphere model we obtain

$$
\frac{\partial}{\partial x} \{ [\mathbf{P}_{xx}(x) - \mathbf{P}_{yy}(x)] u(x) \} + 2 \mathbf{P}_{xx}(x) \frac{\partial u(x)}{\partial x} \n= \frac{3m}{2} \mu_{xx} \Xi + \frac{3m}{2} \mu_{xx}^2 \Xi_1 / 2 \n+ \frac{3m}{2} \mu_{xy}^2 \Xi_2 / 2 + \frac{3m}{2} \mu_{yz}^2 \Xi_3 / 2.
$$
\n(32)

To obtain a closed system for the fluxes we use the transport equation [Eq. (11)] with $\Psi(c) = mC_x c_y$ and $\Psi(c)$ $=mc_vc_z$ to obtain that

$$
\frac{\partial}{\partial x}\big[u(x)\mathbf{P}_{xy}(x)\big]=m\mu_{xy}\Xi_4+m\mu_{xx}\mu_{xy}\Xi_5+m\mu_{xy}\mu_{yz}\Xi_6,
$$
\n(33)

and

$$
\frac{\partial}{\partial x} [u(x)\mathbf{P}_{yz}(x)] = m\mu_{yz} \Xi_7 + m\mu_{xx}\mu_{yz} \Xi_8 + m\mu_{xy}^2 \Xi_9/2.
$$
\n(34)

The collision integrals that appear in Eqs. (32) , (33) , and (34) have been evaluated for the rigid sphere model, the results being

$$
\Xi_1 = \left\{ \mathbf{C}_x^2 \, ; \Delta^{\star}(f_1(\mathbf{c}), f_1(\mathbf{c}_1)) \right\} = -\frac{32}{35} \sigma^2 n^2 \sqrt{\pi} \left(\frac{kT}{m} \right)^{7/2},\tag{35}
$$

$$
\Xi_2 = \{ \mathbf{C}_x^2 \, ; \Delta^{\star} (f_2(\mathbf{c}), f_2(\mathbf{c}_1)) \} = -\frac{32}{105} \sigma^2 n^2 \sqrt{\pi} \left(\frac{kT}{m} \right)^{7/2},
$$
\n
$$
\Xi_3 = \{ \mathbf{C}_x^2 \, ; \Delta^{\star} (f_3(\mathbf{c}), f_3(\mathbf{c}_1)) \} = \frac{32}{105} \sigma^2 n^2 \sqrt{\pi} \left(\frac{kT}{m} \right)^{7/2},
$$
\n
$$
\Xi_4 = [\mathbf{C}_x^2 \, ; f_2(\mathbf{c})] = -\frac{16}{5} \sigma^2 n^2 \sqrt{\pi} \left(\frac{kT}{m} \right)^{5/2},
$$

 $7/2$

$$
\Xi_{5} = \{ \mathbf{C}_{x} \mathbf{c}_{y} ; \Delta^{\star} (f_{1}(\mathbf{c}), f_{2}(\mathbf{c}_{1})) \} = -\frac{8}{35} \sigma^{2} n^{2} \sqrt{\pi} \left(\frac{kT}{m} \right)^{7/2},
$$
\n
$$
\Xi_{6} = \{ \mathbf{C}_{x} \mathbf{c}_{y} ; \Delta^{\star} (f_{2}(\mathbf{c}), f_{3}(\mathbf{c}_{1})) \} = -\frac{8}{35} \sigma^{2} n^{2} \sqrt{\pi} \left(\frac{kT}{m} \right)^{7/2},
$$
\n
$$
\Xi_{7} = [\mathbf{C}_{x}^{2} ; f_{3}(\mathbf{c})] = -\frac{16}{5} \sigma^{2} n^{2} \sqrt{\pi} \left(\frac{kT}{m} \right)^{5/2},
$$
\n
$$
\Xi_{8} = \{ \mathbf{c}_{y} \mathbf{c}_{z} ; \Delta^{\star} (f_{1}(\mathbf{c}), f_{3}(\mathbf{c}_{1})) \} = \frac{16}{35} \sigma^{2} n^{2} \sqrt{\pi} \left(\frac{kT}{m} \right)^{7/2},
$$
\n
$$
\Xi_{9} = \{ \mathbf{c}_{y} \mathbf{c}_{z} ; \Delta^{\star} (f_{2}(\mathbf{c}), f_{2}(\mathbf{c}_{1})) \} = -\frac{16}{35} \sigma^{2} n^{2} \sqrt{\pi} \left(\frac{kT}{m} \right)^{7/2}.
$$

In Eq. (35) some shorthand notation has been introduced, namely, the functions f_i , $i=1,2,3$ are defined by

$$
f_1(\mathbf{c}) \equiv \mathbf{C}_x^2 - \frac{1}{2} (\mathbf{c}_y^2 + \mathbf{c}_z^2),
$$

\n
$$
f_2(\mathbf{c}) \equiv \mathbf{C}_x(\mathbf{c}_y + \mathbf{c}_z),
$$

\n
$$
f_3(\mathbf{c}) \equiv \mathbf{c}_y \mathbf{c}_z.
$$
\n(36)

 $\Delta^{\star}(\Theta(c), \Lambda(c_1))$ is defined as

$$
\Delta^{\star}(\Theta(\mathbf{c}), \Lambda(\mathbf{c}_1)) \equiv \Theta(\mathbf{c}') \Lambda(\mathbf{c}'_1) + \Theta(\mathbf{c}'_1) \Lambda(\mathbf{c}') - \Theta(\mathbf{c}) \Lambda(\mathbf{c}_1) - \Theta(\mathbf{c}_1) \Lambda(\mathbf{c}),
$$
(37)

where the primes denote the final velocities in the binary collision, and the curly brackets mean

$$
\Phi(\mathbf{c}); \Delta^{\star}(\Theta(\mathbf{c}), \Lambda(\mathbf{c}_1))
$$
\n
$$
\equiv \int d\mathbf{c} d\mathbf{c}_1 d\mathbf{e} \Sigma(\chi, g) g f^{(0)}(\mathbf{c}, \mathbf{r}, t)
$$
\n
$$
\times f^{(0)}(\mathbf{c}_1, \mathbf{r}, t) \Phi(\mathbf{c}) \Delta^{\star}(\Theta(\mathbf{c}), \Lambda(\mathbf{c}_1)). \tag{38}
$$

The evaluation of the collision integrals given by Eqs. (35) , and others that turn out to be zero, is a tedious labor if attempted by hand, which is why we used computer algebra. We have been unable to find results we can compare with, but the general methodology used in evaluating the integrals was checked out in the following particular case; if $f_4(c)$ $=1$ then

$$
\{\Phi(\mathbf{c}); \Delta^{\star}(f_i(\mathbf{c})f_4(\mathbf{c}_1))\} = [\Phi(\mathbf{c}); f_i(\mathbf{c})],
$$
 (39)

where we recall that the square brackets are defined by Eq. (15) . The results from the symbolic algebra program are in agreement with hand calculations for the relevant collision integrals in the linear regime Ξ_4 , Ξ_7 , and Ξ . Also, as we show in Appendix B, a hand calculation was done to evaluate $\{C_x^2, \Delta^{\star}(C_x^2, C_{1x}^2)\}\)$ leading to the same value as the one calculated with the computer algebra code. To this extent we believe the results of this section are reliable.

In terms of the reduced variables introduced in the preceding section and with the approximation that the derivatives of the fluxes can be neglected, the reduced form of Eqs. (32) , (33) , and (34) finally leads to the results that

$$
\begin{aligned} (\widetilde{\mathbf{P}}_{xx} - 1) a_l^{\star} + \frac{4}{3} \widetilde{\mathbf{P}}_{xx} a_l^{\star} \\ &= -(\widetilde{\mathbf{P}}_{xx} - 1) - \frac{1}{28} (\widetilde{\mathbf{P}}_{xx} - 1)^2 - \frac{1}{21} \widetilde{\mathbf{P}}_{xy}^2 + \frac{1}{21} \widetilde{\mathbf{P}}_{yz}^2 \,, \end{aligned} \tag{40}
$$

$$
\widetilde{\mathbf{P}}_{xy}a_t^* = -\widetilde{\mathbf{P}}_{xy} - \frac{1}{28}\widetilde{\mathbf{P}}_{xy}(\widetilde{\mathbf{P}}_{xx} - 1) - \frac{1}{14}\widetilde{\mathbf{P}}_{xy}\widetilde{\mathbf{P}}_{yz},\qquad(41)
$$

$$
\widetilde{\mathbf{P}}_{yz} a_l^{\star} = -\widetilde{\mathbf{P}}_{yz} + \frac{1}{14} \widetilde{\mathbf{P}}_{yz} (\widetilde{\mathbf{P}}_{xx} - 1) - \frac{1}{14} \widetilde{\mathbf{P}}_{xy}^2. \tag{42}
$$

This is a set of closed constitutive equations for the independent components of the pressure tensor written in terms of the longitudinal deformation rate a_l^* .

Equation (41) can be rewritten as

$$
\widetilde{\mathbf{P}}_{xy} \left[1 + a_t^{\star} + \frac{1}{28} (\widetilde{\mathbf{P}}_{xx} - 1) + \frac{1}{14} \widetilde{\mathbf{P}}_{yz} \right] = 0. \tag{43}
$$

From the two solutions of Eq. (43) it is easily seen that $\tilde{\mathbf{P}}_{xy} = 0$ is the only solution giving the correct limiting behavior for small a_l^* . Thus, using $\tilde{P}_{xy} = 0$ in Eq. (42) we infer that \widetilde{P}_{yz} must vanish to be consistent with the correct limiting behavior for small a_l^* . Then substitution of $\tilde{P}_{xy} = 0$ and \tilde{P}_{yz} $=0$ in Eq. (40) leads us to the result that

$$
\frac{1}{28}\tilde{\mathbf{P}}_{xx}^2 + (7a_l^{\star}/3 + 13/14)\tilde{\mathbf{P}}_{xx} - (27/28 + a_l^{\star}) = 0, \quad (44)
$$

from which the solution with the correct limiting behavior follows,

$$
\widetilde{\mathbf{P}}_{xx} = -\frac{98}{3}a_l^{\star} - 13 + \frac{2}{3}\sqrt{2401a_l^{\star^2} + 1974a_l^{\star} + 441}.\tag{45}
$$

The leading terms in a_l^* of the reduced pressure tensor can be shown to be given by

$$
\widetilde{\mathbf{P}}_{xx} = 1 - \frac{4}{3} a_l^{\star} + \frac{64}{21} a_l^{\star^2} + O(a_l^{\star^3}),\tag{46}
$$

which means that we recover the first order Chapman-Enskog expression for the viscosity.

Defining a viscosity ($\eta_{\rm nl}$) through Eq. (18) with $\eta_{\rm nl}$ instead of η_{NS} , as in the preceding section, η_{nl}^{\star} is given by

FIG. 1. Percentage deviation of η_l with respect to η_{nl} versus the reduced longitudinal deformation rate, ϵ vs a_l^* .

$$
\eta_{\text{nl}}^{\star} \equiv \frac{\eta_{\text{nl}}}{\eta_{\text{NS}}} = \frac{49a_l^{\star} + 21 - \sqrt{2401a_l^{\star^2} + 1974a_l^{\star} + 441}}{2a_l^{\star}}.
$$
\n(47)

It is instructive to examine the percentage deviation (ϵ) of the two reduced viscosities as a function of a_i^* :

$$
\epsilon = \frac{(\eta_{\rm nl}^{\star} - \eta_l^{\star})}{\eta_{\rm nl}^{\star}} \times 100\% \,. \tag{48}
$$

In Fig. 1 a plot of ϵ as a function of a_l^* is given, for a_l^* >0 . It is seen that the maximum percentage deviation, in the region shown, is about 0.52%, which means that the nonlinear terms represent a marginal modification to the linear result.

However, for a_l^* < 0, where we expect problems mentioned in the preceding section, the situation is quite different since the percent deviation is very large due to the prediction of negative viscosities of the linear theory. In Fig. 2 the behavior of η_{nl}^* as a function of a_l^* is shown where a thickening of the viscosity can be observed. After an appreciable increase in the reduced viscosity we observe a region in which the viscosity is again nearly independent of the longitudinal deformation rate but its plateau value is about 50 times greater than the Navier-Stokes result. This behavior of the viscosity comes out as a result of considering the nonlinear contributions of the fluxes to the collision term and cannot be predicted by a linear theory.

FIG. 2. Reduced nonlinear viscosity versus the reduced longitudinal deformation rate, η_{nl}^{\star} vs a_l^{\star} .

Finally we would like to know under what conditions the approximation of neglecting the gradients of the fluxes holds. The explicit form of P_{xx} for the nonlinear case is the following:

$$
\mathbf{P}_{xx} = -\frac{98}{3} \eta_{\text{NS}} \frac{\partial u}{\partial x} - 13p(x)
$$

+
$$
\frac{2}{3} \left[2041 \eta_{\text{NS}}^2 \left(\frac{\partial u}{\partial x} \right)^2 + 1974 \eta_{\text{NS}} \frac{\partial u}{\partial x} p(x) + 441p(x)^2 \right]^{1/2}.
$$
 (49)

Evaluating the ratio defined in Eq. (27) (Q_{nl}) , where now the pressure tensor is given by Eq. (49) , we obtain

$$
Q_{nl} = -\frac{98}{3} \frac{u(x)\partial \eta_{NS}/\partial x}{g_2} - \frac{98}{3} \frac{u(x)\eta_{NS}(x)\partial^2 u/\partial^2 x}{g_2 \partial u/\partial x}
$$

$$
-\frac{29}{2} \frac{u(x)\partial p/\partial x}{g_2 \partial u/\partial x}
$$

$$
+\frac{4082}{3} \frac{u(x)\eta_{NS}(x)(\partial u/\partial x)(\partial \eta_{NS}/\partial x)}{g_2 \partial u/\partial x}
$$

$$
+\frac{4082}{3} \frac{u(x)\eta_{NS}^2(x)\partial^2 u/\partial^2 x}{g_2 \sqrt{g_1}} + 658 \frac{u(x)p(x)\partial \eta_{NS}/\partial x}{g_2 \sqrt{g_1}}
$$

+658
$$
\frac{u(x)\eta_{\text{NS}}(x)\partial^2 u/\partial^2 x}{g_2\sqrt{g_1}\partial u/\partial x} + 658 \frac{u(x)\eta_{\text{NS}}(x)\partial p/\partial x}{g_2\sqrt{g_1}}
$$

$$
+294 \frac{u(x)p(x)\partial p/\partial x}{g_2\sqrt{g_1}\partial u/\partial x},\tag{50}
$$

where

$$
g_1 = 2041 \eta_{\text{NS}}^2(x) \left(\frac{\partial u}{\partial x}\right)^2 + 1974 \eta_{\text{NS}}(x) p(x) \frac{\partial u}{\partial x} + 441 p^2(x),
$$

\n
$$
g_2 = -\frac{98}{3} \eta_{\text{NS}}(x) \frac{\partial u}{\partial x} + \frac{29}{2} p(x) + \frac{2}{3} \sqrt{g_1}.
$$
\n(51)

We conclude that the conditions under which the approximation holds, namely, that the gradients of the fluxes can be neglected, are the same as those obtained in the preceding section. This means that the gradients of the fluxes can be neglected when $|\partial u/\partial x|$ is very large but the quantities *u*, *p*, *T*, $\partial u/\partial x$, $\partial p/\partial x$, $\partial T/\partial x$, and $\partial^2 u/\partial x^2$ remain finite.

It is interesting to compare with the results by Eu $[10]$, if there are no temperature gradients. In this case for the stationary case Eu's results (see Eq. 8.69) can be expressed as

$$
\eta_e = \eta_{\rm NS} \bigg(\frac{\sinh^{-1}(\kappa)}{\kappa} \bigg), \tag{52}
$$

where,

$$
\kappa = g_0 (2 \eta_{\text{NS}} [\nabla \mathbf{u}]^{(2)} \odot [\nabla \mathbf{u}]^{(2)})^{1/2}, \tag{53}
$$

where \bf{u} is the hydrodynamic velocity and the superindex (2) means the traceless symmetric tensor $[10]$. We have been unable to find explicit reference to this notation in Eu's book so this is an inference we made based on his Eq. (6.47) when compared with the known Chapman-Enskog $[15]$ result. Also, one must notice that the thermal conductivity defined by Eu is different from the one defined by Chapman and Cowling [15]. The symbol \odot denotes the full contraction of the tensors. Moreover,

$$
g_0 = \frac{(mkT)^{1/4}}{\sqrt{2}p\,\sigma}.\tag{54}
$$

For the rigid sphere model in which $\eta_{\rm NS}$ is given by Eq. (19) and after some transformations Eu's result can be recast in the form

$$
\eta_{\text{Eu}}^{\star} = \frac{\eta_e}{\eta_{\text{NS}}} = \frac{\sinh^{-1}(\xi)}{\xi}, \quad \xi \equiv \sqrt{\frac{32\sqrt{\pi}}{15}} |a_l^{\star}|, \quad (55)
$$

where

$$
a_l^* = \frac{\eta_{\rm NS}}{p} \frac{\partial u}{\partial x}.
$$
 (56)

It is easy to see that for positive longitudinal rates the reduced viscosity is decreasing as for all the nonlinear viscosities that we have discussed, but notice that Eu's reduced viscosity does not depend on the sign of the reduced longitudinal rate, which means that his expression is symmetric

around $a_l^* = 0$ which is in contrast with our result and also with the results by Karlin *et al.* [8]. However, when $a_l^* \rightarrow$ $-\infty$ the nonlinear viscosity given by Eq. (56) goes to zero as happens for the nonlinear viscosity obtained by Karlin *et al.* [8,9], which is in contrast to the plateau found in this section. For $a_l^* = 1$, η_{Eu}^* is equal to 0.7295, in comparison with the results given in Sec. III. The question that springs to mind is the following: What is a more reasonable behavior, from a physical point of view, for large and negative longitudinal rates of the *xx* component of the viscous pressure tensor? In order to get a qualitative understanding let us consider an explosion or implosion of a gas with spherical symmetry. Under this condition the problem is one dimensional, the explosion corresponds to positive longitudinal rates, and for this case all the theories that we have considered predicted that the *xx* component of the reduced stress tensor should go to zero. In the case of an implosion the theories that predict that the viscosity should go to zero when the longitudinal rate goes to $-\infty$ imply that if the compression is large enough the *xx* component of the viscous stress tensor is equal to zero. This means that for large and negative longitudinal rates the *xx* component of the stress tensor is equal to the pressure. Our guess is that it is more reasonable to expect that in an implosion the finite size of the atoms would imply that that it is much harder to compress the system even for large and negative gradients. So, this qualitative argument favors the behavior of the viscosity that we have obtained in this section. Nevertheless it would be convenient to have experiments or simulations to verify our expectations.

V. A GENERAL RESULT

In this section we will prove that the result for the reduced viscosity obtained in Sec. III | see Eq. (31) | is independent of the interatomic potential. To see this we argue as follows: from Grad's ten-moment equations and considering the linearized collision operator, we obtain that

$$
\frac{\partial}{\partial x}\{[\mathbf{P}_{xx}(x) - \mathbf{P}_{yy}(x)]u(x)\} + 2\mathbf{P}_{xx}(x)\frac{\partial u(x)}{\partial x} = \frac{3m}{2}\Xi^G,
$$
\n(57)

where Ξ^G is a collision integral given by

$$
\Xi^G = [\mathbf{C}_x^2; \mathbf{C}_x^2 - \frac{1}{2}(\mathbf{c}_y^2 + \mathbf{c}_z^2)],\tag{58}
$$

and the term in square brackets is defined by Eq. (15) . Notice that in the linearized collision operator the contributions from μ_{xy} and μ_{yz} [see Eq. (3)] are zero due to parity reasons and also that the drift term is independent of the interaction potential.

Let us now assume that by consistency the previous equation should yield the Navier-Stokes constitutive equation for small gradients. Then, following Grad we make the substitution $P_{xx} = P_{yy} = p$ in the left hand side of Eq. (57) (zero order in the Knudsen number) to obtain

$$
2p\frac{\partial u}{\partial x} = \frac{3m^2}{4kT} \Xi^G \bigg(\frac{\mathbf{P}_{xx}}{p} - 1\bigg). \tag{59}
$$

Equation (59) is an algebraic equation for P_{xx} which can be solved to yield

$$
\mathbf{P}_{xx} = p + \frac{8}{3} \frac{p^2 kT}{m^2 \Xi^G} \frac{\partial u}{\partial x}.
$$
 (60)

Equation (60) is consistent with the Chapman-Cowling $|15|$ result,

$$
\mathbf{P}_{xx} = p - \frac{4}{3} \eta_{CC} \frac{\partial u(x)}{\partial x},\tag{61}
$$

where $\eta_{\rm CC}$ is the viscosity, provided that

$$
\eta_{\rm CC} = -2 \frac{p^2 kT}{m^2 \Xi^G}.
$$
\n(62)

Alternatively Eq. (62) may be regarded as an expression for the viscosity in terms of a collision integral.

We now assume that the gradients of the fluxes can be neglected and obtain from Eq. (25) and using Eq. (62) that

$$
\frac{3}{2}(\mathbf{P}_{xx} - nkT) \frac{\partial u}{\partial x} + 2\mathbf{P}_{xx} \frac{\partial u}{\partial x} = \frac{3p^2}{2\eta_{CC}} \left(\frac{\mathbf{P}_{xx}}{p} - 1\right). \tag{63}
$$

In terms of the reduced quantities given by

$$
a_l^* = \frac{\eta_{\rm CC}}{p} \frac{\partial u}{\partial x}, \quad \widetilde{\mathbf{P}}_{xx} = \frac{\mathbf{P}_{xx}}{p}, \tag{64}
$$

Eq. (63) can be rewritten as

$$
(\widetilde{\mathbf{P}}_{xx} - 1)a_t^{\star} + \frac{4}{3}\widetilde{\mathbf{P}}_{xx}a_t^{\star} = -(\widetilde{\mathbf{P}}_{xx} - 1). \tag{65}
$$

Equation (65) is the same as the one that we obtained for the rigid sphere case [see Eq. (24)] and from it we conclude that

$$
\widetilde{\mathbf{P}}_{xx} = \frac{(1+a_t^{\star})}{(1+7a_t^{\star}/3)}, \quad \eta_l^{\star} \equiv \frac{\eta_l}{\eta_{CC}} = \frac{1}{1+7a_l^{\star}/3}.
$$
 (66)

So, in terms of the appropriate variables given by Eq. (64) the form of the reduced pressure tensor and the reduced viscosity, given by Eq. (66) , are independent of the interatomic potential. We would like to stress that this result holds only when using the linear collision operator; for the nonlinear case it is not obvious at this stage if a similar result can be obtained.

VI. DISCUSSION

As mentioned in the Introduction, there is no doubt that there are situations in which the viscosity depends on the gradients of the velocity as first found by Truesdell and coworkers $|1|$. The question now is to have an idea of what the underlying physical mechanism for such a phenomenon is. At this point it is convenient to depart from the situation considered in this work and consider the physical situation in which the viscosity is introduced. Usually it corresponds to the shear rate in a stationary state where the hydrodynamic velocity $[u(y)]$ has only the *x* component which depends on the *y* coordinate.

The viscosity calculation $[18,19]$ uses the concept of the mean free path (*l*) to establish that, by taking into account the net molecular transport of the momentum between two planes whose separation is of the order of the mean free path, a linear relation between the *x*-*y* component of the pressure tensor and the shear rate $(\partial u/\partial y)$ must hold. The constant of proportionality is the viscosity (n) which turns out to be related with the mean free path by

$$
\eta = \frac{1}{3} n \,\overline{v} m l,\tag{67}
$$

where \bar{v} is the mean velocity. Such a derivation contains implicitly the conditions that the higher order gradients of the hydrodynamic velocity can be neglected and that this is the only quantity that depends on *y*. Clausius obtained, for a gas of particles moving with the same absolute value of velocity, an expression for the mean free path, which was later corrected by Maxwell by using a Maxwellian distribution function $[20]$. However, in a non-equilibrium situation, there are corrections to the Maxwellian distribution function which in principle should be taken into account.

Let us calculate the mean free path, or equivalently the collision frequency, for a non-equilibrium situation. We consider the first approximation to the distribution function given by Chapman and Cowling $[15]$ for a shear flow (for simplicity we will not include the heat flow),

$$
f(\mathbf{r}, \mathbf{c}) = f^{(0)} \left[1 - \frac{5}{8n\sqrt{\pi}\sigma^2} \left(\frac{m}{kT} \right)^{3/2} \frac{\partial u}{\partial y} \mathbf{C}_x \mathbf{C}_y \right].
$$
 (68)

For rigid spheres, the number of collisions between the molecules per unit volume and time (N_1) is [15]

$$
N_1 = \frac{1}{8} \int \sin(\chi) d\chi d\epsilon d\mathbf{c}_1 d\mathbf{c}_2 \sigma^2 f(\mathbf{r}, \mathbf{c}_1) f(\mathbf{r}, \mathbf{c}_2). \quad (69)
$$

Using Eq. (68) to evaluate N_1 we obtain

$$
N_1 = 2n^2\sigma^2 \left(\frac{\pi kT}{m}\right)^{1/2} - \frac{5}{384} \left(\frac{m}{\pi kT}\right)^{1/2} \frac{1}{\sigma^2} \left(\frac{\partial u}{\partial y}\right)^2.
$$
 (70)

The collision frequency is defined as N_1/n and then the mean free path can be calculated $[15]$. The main conclusion that we can extract from our calculation is that the collision frequency and the mean free path are expected to be shear rate dependent if the distribution function does not correspond to the Maxwellian. The usual Chapman-Enskog method assumes the gradients are small enough so that the quadratic terms in the shear rate can be neglected. However, in situations where the shear rate is large we can have a shear rate-dependent mean free path which in turn can be expected to give a shear rate-dependent viscosity which is precisely the effect that we have been discussing. While the Chapman-Enskog formulation is incapable of treating these cases, we have the Burnett and higher order equations that have been considered by Chapman and Cowling $[15]$ but these are restricted to small gradients. Regularizations of these expansions can be found in the literature $[8,9,21]$ and the present work shows that Grad's method is able to cope with large longitudinal deformation rates and that a longitudinal deformation rate dependence of the viscosity can be obtained in an almost straightforward way. In these cases the usual derivation of the relation between the viscosity and the mean free path is incorrect since it is assumed that the mean free path does not depend on the shear rate.

Let us now consider a fluid at rest between two parallel planes located at $z=0$ and $z=d$. If the plane at $z=d$ is moving to the right along the *x* axis and the fluid remains at rest, then, the shear rate is infinite; but this could only happen if the fluid does not have viscosity (slip boundary condition), which means that the viscosity must be zero for such a case. Since for small shear rates the viscosity is different from zero it follows that this quantity must be shear dependent.

For a longitudinal flow the necessity of a viscosity depending on the longitudinal deformation rate can be understood in the following way: from the definition of the pressure tensor [see Eq. (8)] it follows that P_{xx} should be greater than zero since the distribution function is non-negative. Assuming that P_{xx} given by Eq. (18) holds for any longitudinal rate, then for $a_l^* \ge 0$ we will have a region of longitudinal rates for which P_{xx} <0 in contradiction with what was previously stated. Notice that the previous argument does not hold if a_i^* < 0 and that positive viscosities, according to the Chapman-Enskog theory, are implicitly assumed.

The nature of the results obtained in this paper merits another comment which is intimately related to the nature of the constitutive equations that are required to obtain a well defined set of hydrodynamic equations. It is well known that the linear laws, Navier-Stokes-Fourier (NSF), give rise to a set of hydrodynamic equations which are of order 2 in the gradients, the corresponding transport coefficients being functions of the *local* equilibrium thermodynamic variables only $[15,22,23]$. A long debated question has been how to go about generalizing these linear laws to yield hydrodynamic equations that include terms of higher order in the gradients, meaning specifically of order higher than 2. According to the kinetic theory based on the Boltzmann equation, the Chapman-Enskog expansion, which is valid for small gradients, leads to one alternative which is well known and its consequences, especially those related to their compatibility with irreversible thermodynamics, have been widely discussed in the literature $[23-27]$. We shall not deal with them here. The point we want to underline is that when the gradients are large, in our case ∇u , then the natural constitutive equation for the viscous stress tensor could be written as

$$
\tau = \eta(\nabla \mathbf{u}) \cdot \nabla \mathbf{u},\tag{71}
$$

indicating that the viscosity is, in general, a nonlinear tensor function of the viscosity gradients. This is, in a macroscopic language, what the results of this paper suggest as the appropriate relationship. But this is not new. Some years ago it was clearly shown $|24-26|$ how one may obtain hydrodynamic equations of arbitrary order in the gradients by constructing a matrix whose elements are transport coefficients such that acting on a vector defined by the thermodynamic forces yields expressions for the corresponding fluxes. If the elements of such a matrix are of a given order in the gradients the resulting fluxes will be one order higher. Equation (71) is an example of this procedure. In the lowest approximation one recovers the NSF hydrodynamics equations and in higher orders, equations which resemble those obtained from the Chapman-Enskog method of solving the Boltzmann equation although in the case of Eq. (71) a series expansion is not justified. Therefore, Eq. (71) may be regarded as a nonlinear constitutive equation which is valid independently of whether ∇u is or is not a small quantity. Moreover, it was also shown that this scheme is perfectly in harmony with the local equilibrium assumption and the second law of thermodynamics, thus providing a solid basis for these types of constitutive equations.

In conclusion we see that it is natural to expect that in situations not near equilibrium, which can be generated by large velocity gradients, the viscosity should depend on the velocity gradients. Also, there are situations $(a_i^* \ge 0)$ in which the nonlinear contribution of the fluxes to the collision term is minor, but for other cases $(a_l^*<0)$ they give a significant contribution. It should be pointed out that the results obtained depend strongly, besides Grad's approximation, on the assumption that the gradients of the fluxes can be neglected, it is only in this case that we can guarantee a viscosity depending only on the first order derivative of the hydrodynamic velocity. As far as we know this is the first time that a thickening in the viscosity followed by a plateau has been reported and also the importance of the nonlinear contributions of the fluxes to the collision term has been clearly shown.

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APPENDIX A: EVALUATION OF THE DRIFT TERMS IN THE TRANSPORT EQUATION

In this appendix we consider the evaluation of the transport equation for $\psi(\mathbf{C}) = 3m/2\mathbf{C}_x^2$, that is, we would like to evaluate the left hand side of the following equation:

$$
\int d\mathbf{c} \frac{3m}{2} \mathbf{C}_x^2 \left(\frac{\partial f}{\partial t} + \mathbf{c} \cdot \nabla_{\mathbf{r}} f \right) = \int d\mathbf{c} \frac{3m}{2} \mathbf{C}_x^2 J(f, f).
$$
\n(A1)

First we evaluate the drift term; for simplicity we will use the conditions used in this work, that is, $\mathbf{c}_0(\mathbf{r},t) \equiv u(x)\mathbf{i}$, then for the stationary case we have

$$
\int d\mathbf{c}m\mathbf{C}_{x}^{2}\mathbf{c}\cdot\mathbf{\nabla}_{\mathbf{r}}f = m \int d\mathbf{c}\mathbf{C}_{x}^{2}\mathbf{c}_{x}\frac{\partial f}{\partial x}
$$

\n
$$
= m \int d\mathbf{c} \left(\frac{\partial f\mathbf{C}_{x}^{2}\mathbf{c}_{x}}{\partial x} - f\mathbf{c}_{x}\frac{\partial \mathbf{C}_{x}^{2}}{\partial x}\right)
$$

\n
$$
= m \frac{\partial}{\partial x} \left(\int d\mathbf{c}f\mathbf{C}_{x}^{3}\right) + m \frac{\partial}{\partial x} \left(\int d\mathbf{c}f\mathbf{C}_{x}^{2}u\right)
$$

\n
$$
+ 2m \frac{\partial u(x)}{\partial x} \left(\int d\mathbf{c}f(\mathbf{C}_{x}^{2} + \mathbf{C}_{x}u)\right)
$$

\n
$$
= \mathbf{Q}_{x} + \frac{\partial}{\partial x} (\mathbf{P}_{xx}u) + 2\mathbf{P}_{xx}\frac{\partial u}{\partial x}.
$$
 (A2)

The first equality follows from the assumption that all the quantities depend only on the *x* coordinate and the second one follows from an integration by parts. The third equality is obtained by using the relation $\mathbf{c}_x = \mathbf{C}_x + u(x)$, interchanging the order for the integration and the partial derivative in the first two terms, and carrying the partial derivative with respect to *x* of the squared peculiar velocity. Finally the fourth equality follows from the definition of the pressure tensor. It is pertinent to stress that the quantity \mathbf{Q}_x defined by

$$
\mathbf{Q}_x \equiv \int d\mathbf{c} f \mathbf{C}_x^3, \tag{A3}
$$

is zero if the distribution function is given by Eq. (2) .

Using similar arguments as the ones used to obtain the previous equation we obtain

$$
\frac{m}{2} \int d\mathbf{c} (\mathbf{c}_y^2 + \mathbf{c}_z^2) \mathbf{c} \cdot \nabla f
$$
\n
$$
= \frac{m}{2} \int d\mathbf{c} (\mathbf{c}_y^2 + \mathbf{c}_z^2) \nabla \cdot (\mathbf{c}f) = \nabla \cdot \frac{m}{2} \int d\mathbf{c} (\mathbf{c}_y^2 + \mathbf{c}_z^2) \mathbf{c}f
$$
\n
$$
= \nabla \cdot \left(\frac{m}{2} \int d\mathbf{c} (\mathbf{C}_x^2 + \mathbf{c}_y^2 + \mathbf{c}_z^2) \mathbf{c}f\right) - \nabla \cdot \left(\frac{m}{2} \int d\mathbf{c} \mathbf{C}_x^2 \mathbf{c}f\right)
$$
\n
$$
= \nabla \cdot \left(\frac{m}{2} \int d\mathbf{c} \mathbf{C}^2 \mathbf{C}f\right) + \nabla \cdot \left(\frac{m}{2} \int d\mathbf{c} \mathbf{C}^2 \mathbf{c}_0 f\right)
$$
\n
$$
- \nabla \cdot \left(\frac{m}{2} \int d\mathbf{c} \mathbf{C}_x^2 \mathbf{c}_0 f\right) - \nabla \cdot \left(\frac{m}{2} \int d\mathbf{c} \mathbf{C}_x^2 \mathbf{C}f\right)
$$
\n
$$
= \frac{\partial \mathbf{q}_x}{\partial x} + \frac{1}{2} \frac{\partial u(x)(\mathbf{P})}{\partial x} - \frac{1}{2} \frac{\partial u(x)\mathbf{P}_{xx}}{\partial x} - \frac{\partial \mathbf{Q}_x}{\partial x} = \frac{\partial \mathbf{q}_x}{\partial x} - \frac{\partial \mathbf{Q}_x}{\partial x}
$$
\n
$$
+ \frac{1}{2} \frac{\partial u(x)(\mathbf{P}_{yy} + \mathbf{P}_{zz})}{\partial x}
$$
\n
$$
= \frac{\partial \mathbf{q}_x}{\partial x} - \frac{\partial \mathbf{Q}_x}{\partial x} + \frac{\partial}{\partial x} (u(x)\mathbf{P}_{yy}), \qquad (A4)
$$

where **q** is the heat flux which is zero if the distribution function given by Eq. (2) is used and in the last equality we have used cylindrical symmetry.

From Eqs. $(A1)$, $(A2)$, and $(A4)$ we obtain, for the distribution function given by Eq. (2) ,

$$
\int d\mathbf{c} \left(m\mathbf{C}_{x}^{2} - \frac{m}{2} (\mathbf{c}_{y}^{2} + \mathbf{c}_{z}^{2}) \right) J(f, f)
$$

$$
= \frac{\partial}{\partial x} (\mathbf{P}_{xx} u(x)) + 2 \mathbf{P}_{xx} \frac{\partial u}{\partial x} - \frac{\partial}{\partial x} (u(x) \mathbf{P}_{yy}).
$$
(A5)

Finally using the relation $\mathbf{c}_y^2 + \mathbf{c}_z^2 = \mathbf{C}^2 - \mathbf{C}_x^2$ in the left hand side of Eq. $(A5)$ and remembering that C^2 is a collision invariant so that

$$
\int d\mathbf{c} \mathbf{C}^2 J(f, f) = 0,\tag{A6}
$$

we obtain

$$
\int d\mathbf{c} \frac{3m}{2} \mathbf{C}_x^2 J(f, f) = \frac{\partial}{\partial x} [(\mathbf{P}_{xx} - \mathbf{P}_{yy}) u(x)] + 2 \mathbf{P}_{xx} \frac{\partial u}{\partial x}.
$$
\n(A7)

APPENDIX B: EVALUATION OF THE COLLISION INTEGRALS

Here we show how to evaluate the collision integrals that result from considering the nonlinear contribution of the fluxes in the collision term. We will evaluate the collision integral given by

$$
\begin{aligned} \{\mathbf{C}_{x}^{2}, \Delta^{\star}(\mathbf{C}_{x}^{2}, \mathbf{C}_{1x}^{2})\} \\ &= 2 \int d\mathbf{c}d\mathbf{c}_{1} d\mathbf{e} \Sigma(\chi, g)gf^{(0)}(\mathbf{c}, \mathbf{r}, t)f^{(0)}(\mathbf{c}_{1}, \mathbf{r}, t) \\ &\times \mathbf{C}_{x}^{2}(\mathbf{C}_{x}^{\prime 2}\mathbf{C}_{1x}^{\prime 2} - \mathbf{C}_{x}^{2}\mathbf{C}_{1x}^{2}). \end{aligned} \tag{B1}
$$

For rigid spheres we have $\Sigma(\chi, g) = \sigma^2/4$ [15], where σ is the rigid sphere diameter so that using the expression for the Maxwellian distribution function given by Eq. (1) and the center of mass and relative coordinates defined as

$$
\mathbf{C} = \mathbf{G} - \mathbf{g}/2, \quad \mathbf{C}_1 = \mathbf{G} + \mathbf{g}/2,
$$

$$
\mathbf{C}' = \mathbf{G} - \mathbf{g}'/2, \quad \mathbf{C}'_1 = \mathbf{G} + \mathbf{g}'/2,
$$
 (B2)

we obtain the following expression for Θ $\equiv \{C_x^2, \Delta^{\star}(C_x^2, C_{1x}^2)\}$:

$$
\Theta = 2\Omega \int d\mathbf{G} d\mathbf{g} d\mathbf{e}g \exp(-m\mathbf{g}^2/4kT) \exp(-m\mathbf{G}^2/kT)
$$

×
$$
(\mathbf{G}_x - \mathbf{g}_x/2)^2 [(\mathbf{G}_x - \mathbf{g}_x'/2)^2 (\mathbf{G}_x + \mathbf{g}_x'/2)^2
$$

$$
-(\mathbf{G}_x - \mathbf{g}_x/2)^2 (\mathbf{G}_x + \mathbf{g}_x/2)^2],
$$
(B3)

where

$$
\Omega = \frac{n^2 \sigma^2}{32 \pi^3} \left(\frac{m}{kT}\right)^3.
$$
 (B4)

The integration over the dispersion angles, namely, *d***e** $\sin(\chi)d\epsilon d\chi$, can be done using the following relation [15]:

$$
\mathbf{g}'_{x} = g \cos(\theta) \cos(\phi) \sin(\chi) \cos(\epsilon)
$$

- $g \sin(\phi) \sin(\chi) \sin(\epsilon) + g \cos(\phi) \cos(\chi) \sin(\theta)$, (B5)

where θ and ϕ are the polar angles of **g**. (Alternatively, in doing the integration over the dispersion angles, it is possible to choose **g** along the *z* axis so that the polar angles of g' are the dispersion angles. Then, Eq. $(B5)$ reduces a bit and it is simpler to carry out the integration). We obtain for Θ

$$
\Theta = 2\pi\Omega \int d\mathbf{G}d\mathbf{g}g \exp(-m\mathbf{g}^2/4kT) \exp(-m\mathbf{G}^2/kT)
$$

$$
\times (\mathbf{G}_x - \mathbf{g}_x/2)^2 \left(\frac{g^4}{20} + 2\mathbf{G}_x^2 \mathbf{g}_x^2 - \frac{\mathbf{g}_x^4}{4} - \frac{2}{3}\mathbf{G}_2 \mathbf{g}_x^2\right). \tag{B6}
$$

The integrations over \mathbf{G}_y and \mathbf{G}_z can be easily carried out, so that

$$
\Theta = 2 \frac{kT}{m} \pi^2 \Omega \int d\mathbf{g} g \exp(-m\mathbf{g}^2/4kT) \exp(-m\mathbf{G}_x^2/kT)
$$

$$
\times \left(\frac{g^4 \mathbf{G}_x^2}{20} + \frac{g^4 \mathbf{g}_x^2}{80} + 2\mathbf{g}_x^2 \mathbf{G}_x^4 + \frac{\mathbf{g}_x^4 \mathbf{G}_x^2}{4} - \frac{\mathbf{g}_x^6}{16} - \frac{2g^2 \mathbf{G}_x^4}{3} - \frac{g^2 \mathbf{g}_x^2 \mathbf{G}_x^2}{6}\right). \tag{B7}
$$

The integrations over G_x are Gaussian and do not represent a problem. If in addition we use polar coordinates for **g** and carry out the angular integrations we are led to

$$
\Theta = 2 \frac{kT}{m} \pi^2 \Omega \int_0^\infty dg g^3 \exp(-mg^2/4kT)
$$

\n
$$
\times \left[\frac{g^4}{20} \frac{\sqrt{\pi}}{2} \left(\frac{kT}{m} \right)^{3/2} (4\pi) + \frac{g^6}{80} \sqrt{\pi} \left(\frac{kT}{m} \right)^{1/2} \left(\frac{4}{3\pi} \right) \right.
$$

\n
$$
+ 2g^2 \frac{3\sqrt{\pi}}{4} \left(\frac{kT}{m} \right)^{5/2} \left(\frac{4}{3\pi} \right) + \frac{g^4}{4} \frac{\sqrt{\pi}}{2} \left(\frac{kT}{m} \right)^{3/2} \left(\frac{4}{5\pi} \right)
$$

\n
$$
- \frac{g^6}{16} \sqrt{\pi} \left(\frac{kT}{m} \right)^{1/2} \left(\frac{4\pi}{7} \right) - \frac{2g^2}{3} \frac{3\sqrt{\pi}}{4} \left(\frac{kT}{m} \right)^{5/2} (4\pi)
$$

\n
$$
- \frac{g^4}{6} \frac{\sqrt{\pi}}{2} \left(\frac{kT}{m} \right)^{3/2} \left(\frac{4}{3\pi} \right) \right].
$$
 (B8)

Note that the third and sixth terms in Eq. $(B8)$ cancel out. For any natural number *n* it is easy to show that

 \int_0^{∞} $\int_{0}^{\infty} dgg^{3+n} \exp(-mg^{2}/4kT) = \frac{1}{2} \left(\frac{4kT}{m} \right)^{(n+4)/2} \Gamma\left(\frac{n+4}{2}\right),$ $(B9)$

where $\Gamma(x)$ is the gamma function [28]. Substitution of Eq. $(B9)$ into Eq. $(B8)$ leads to

$$
\Theta = 2\left(\frac{kT}{m}\right)^{3/2} \pi^{7/2} \Omega \left[\frac{1}{10} \left(\frac{kT}{m}\right) \frac{1}{2} \Gamma(4) \left(\frac{4kT}{m}\right)^{4} + \frac{1}{60} \frac{1}{2} \Gamma(5) \left(\frac{4kT}{m}\right)^{5} + \frac{1}{10} \left(\frac{kT}{m}\right) \frac{1}{2} \Gamma(4) \left(\frac{4kT}{m}\right)^{4} - \frac{1}{28} \frac{1}{2} \Gamma(5) \left(\frac{4kT}{m}\right)^{5} - \frac{1}{9} \left(\frac{kT}{m}\right) \frac{1}{2} \Gamma(4) \left(\frac{4kT}{m}\right)^{4} \right]
$$

=
$$
2\left(\frac{kT}{m}\right)^{5/2} \pi^{7/2} \left(\frac{4kT}{m}\right)^{4} \Gamma(4) \Omega
$$

$$
\times \left(\frac{1}{20} + \frac{16}{120} + \frac{1}{20} - \frac{16}{56} - \frac{1}{18}\right)
$$

=
$$
-\frac{34816}{105} \left(\frac{kT}{m}\right)^{13/2} \pi^{7/2} \Omega.
$$
 (B10)

Using the value of Ω given by Eq. (B4) in Eq. (B10) we conclude that

$$
\left\{ \mathbf{C}_{x}^{2}, \Delta^{\star}(\mathbf{C}_{x}^{2}, \mathbf{C}_{1x}^{2}) \right\} = -\frac{1088}{105} \left(\frac{kT}{m} \right)^{7/2} \sqrt{\pi} n^{2} \sigma^{2}.
$$
 (B11)

As mentioned previously the result from the computer algebra code gives the same value, for this particular case.

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