Singular Behavior of Shear Flow Far from Equilibrium

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A rare exact solution to the nonlinear Boltzmann equation describing rheological phenomena far from equilibrium is reconsidered. We show the existence of a critical shear rate beyond which the solution fails to exist. The implication of this exact result is a singular transition from simple uniform shear flow to a more complex state, perhaps involving spatial ordering.

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From a theoretical point of view, complex nonlinear transport and rheological properties of fluids remain outside the scope of controlled analysis in general. The single exception is a low density gas described by the nonlinear Boltzmann equation for a state of uniform shear flow (i.e., flow in the x direction with constant velocity field gradient or shear rate, a, in the y direction). This is one of the few inhomogeneous states for which exact results can be obtained far from equilibrium, and therefore is of great significance in providing insight for the type of phenomena that can occur under extreme state conditions. Almost forty years ago Ikenberry and Truesdell [1] observed that the infinite hierarchy of coupled equations for velocity moments of the distribution function decouple for the special case of uniform shear flow and Maxwell molecules (r^{-4} potential). Moments of any degree are then determined by a finite set of coupled equations, providing a constructive procedure to characterize a solution to the Boltzmann equation. The components of the stress tensor determine the rheology for states far from equilibrium, and are characterized by three scalar functions: a shear rate dependent viscosity $\eta(a)$ and two viscometric functions $\psi_1(a)$ and $\psi_2(a)$ associated with normal stresses. The equations for these functions are solvable for all values of the shear rate [1,2], and provide a rare exact description of nonlinear transport outside the Navier-Stokes domain [3].

The fact that the second degree moments are well behaved at all shear rates has led to the implicit assumption that this is true for all moments. Indeed, this property extends to the third degree moments as well [4]. Our primary observation here is that a strict limitation on the shear rate appears in the *steady state* solution to the moment equations of fourth degree. We find solutions for $a < a_c$, no solutions for $a = a_c$, and unphysical solutions for $a > a_c$. We conclude that the Ikenberry-Truesdell solution describes a uniform shear flow steady state only if the shear rate is less than the critical value a_c. Above the critical shear rate, the assumed form of the distribution function for uniform shear flow [Eq. (2) below] no longer reaches a steady state. Instead a more complex steady state is expected, presumably involving new spatial structures. As this result is exact, there can be no doubt that a transition from uniform shear flow to *some* qualitatively different state occurs. What happens as this critical value is approached from below? We offer suggestions below for further theoretical analysis and stress feasibility of numerical studies using either molecular dynamics or Monte Carlo direct simulation methods for the Boltzmann equation.

The nonlinear Boltzmann equation has the general form

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\cdot\nabla_r\right)f+\nabla_v\cdot\left(m^{-1}\mathbf{F}_{\rm ext}f\right)=J[f,f]\;, \quad (1)$$

where $\mathbf{F}_{\rm ext}$ is an external force whose form is chosen below and $J[f,f]$ is the collision operator. The distribution function, $f(\mathbf{r},\mathbf{v},t)$, for uniform shear flow has the form

$$f(\mathbf{r},\mathbf{v},t)=f(\mathbf{v}-\mathbf{U}(\mathbf{r}),t)\equiv f(\boldsymbol{\xi},t)\;, \quad (2)$$

where $\boldsymbol{\xi}\equiv\mathbf{v}-\mathbf{U}(\mathbf{r})$ and $U_x(\mathbf{r})=ay$, $U_y(\mathbf{r})=U_z(\mathbf{r})=0$ are the components of the macroscopic velocity field representing flow in the x direction with constant gradient a along the y axis. For large t, it is expected that this distribution function approaches a quasistationary state, $f(\boldsymbol{\xi},t)\to f_s(\xi,T(t))$, where the temperature, $T(t)$, is a spatially homogeneous function of time determined from the average kinetic energy, $\frac{3}{2}k_BT(t)=\frac{1}{2}\langle m\xi^2\rangle$. This is an example of a "normal" solution, whose space and time dependence occurs entirely through the hydrodynamic fields. It is easily verified from normalization and the first velocity moments that this specific form of the distribution function describes an average velocity field $\mathbf{U}(\mathbf{r})$ and constant (in both space and time) density n. More generally, when defined in terms of the peculiar velocity $\boldsymbol{\xi}$, all moments are spatially uniform—hence the name "uniform" shear flow. In the absence of any external

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force there is viscous heating and $T(t)$ increases monotonically. To simplify the discussion here, we include a nonconservative external force (thermostat) chosen to remove this heating effect. The exact equivalence between the Ikenberry-Truesdell solutions with and without thermostat has been discussed elsewhere [5]. For Maxwell molecules, the quasistationary solution without thermostat corresponds to the stationary solution with thermostat. The external force is taken to be $\mathbf{F}_{\rm ext}/m = -\alpha\boldsymbol{\xi}$, and the constant α is determined as a function of the shear rate by the condition $T(t \to \infty)$ =const.

The Boltzmann equation for uniform shear flow becomes

$$\left(\frac{\partial}{\partial t} - \frac{\partial}{\partial \xi_i}(\alpha\delta_{ij} + a_{ij})\xi_j\right) f = J[f,f] \ , \qquad (3)$$

where $a_{ij} = a\delta_{ix}\delta_{jy}$. In this equation we have taken into account that initial conditions of the form $f(\boldsymbol{\xi},0)$ map into solutions $f(\boldsymbol{\xi},t)$; i.e., no additional space dependence is generated. Furthermore, we are interested in solutions that approach a stationary state, $f(\boldsymbol{\xi},t) \to f_s(\boldsymbol{\xi})$. To explore this possibility we study the corresponding properties for the moments of the distribution function. Equations for the moments of f are obtained by multiplying (3) by polynomials in $\{\xi_i\}$; the precise choice of polynomials is a matter of convenience. The primary observation of Ref. [1] is that the infinite hierarchy of velocity moments obtained from the nonlinear Boltzmann equation for Maxwell molecules is formally solvable in this case. The reason is twofold: First, for the special case of Maxwell molecules the contributions from the collision operator for a moment of given degree depend only on moments up to that degree; second, contributions from the convective terms of the Boltzmann equation also involve only moments of the given degree as a consequence of the uniformity of the distribution function. In this way a constructive procedure to determine all moments is identified. As no approximations are required, the moments obtained are exact consequences of Eq. (3). To illustrate, the moment equations of second degree, $M_{ij} \equiv \langle\xi_i\xi_j\rangle$, are found to be

$$\begin{aligned}\left(\frac{\partial}{\partial t} + 2\alpha\right) M_{ij}(t) + a_{ik}M_{kj}(t) + a_{jk}M_{ki}(t) &= \frac{1}{n}\int d\boldsymbol{\xi}\,\xi_i\xi_j J[f,f] \\ &= -\nu\left[M_{ij}(t) - \tfrac{1}{3}\delta_{ij}M_{kk}(t)\right] \ , \end{aligned} \qquad (4)$$

where the explicit form of $J[f,f]$ for Maxwell molecules has been used in the second equality and ν is a constant proportional to n and depending on the parameters of the potential. An equation for the temperature, $T(t) = mM_{kk}(t)/3k_B$, follows from (4),

$$\left(\frac{\partial}{\partial t} + 2\alpha\right) T(t) + \frac{2m}{3k_B} a_{ij}M_{ij}(t) = 0 \ . \qquad (5)$$

The thermostat parameter α is now determined by the condition that the temperature approach a constant,

$$\alpha(a) = -\frac{m}{3k_B T(\infty)} a_{ij}M_{ij}(\infty) \ . \qquad (6)$$

Equations (4) and (6), together with given initial conditions, completely determine the moments of degree 2. Their solution is straightforward and will not be given here, other than to note that $\alpha(a)$ is the real root of $\nu a^2 = 3\alpha(\nu + 2\alpha)^2$.

It is instructive to write Eq. (4) in a matrix form,

$$\frac{\partial}{\partial t}x_\sigma(t) + L_{\sigma\beta}x_\beta(t) = T(t)b_\sigma \ , \qquad (7)$$

$$x_\sigma \longleftrightarrow \{M_{xx}, M_{yy}, M_{zz}, M_{xy}, M_{xz}, M_{yz}\} \ , \qquad (8)$$

where $L_{\sigma\beta}$ and b_σ are constant matrices identified from (4). The formal solution to (7) is

$$x(t) = e^{-Lt}x(0) + \int_0^t d\tau e^{-L\tau}T(t-\tau)b \ . \qquad (9)$$

A necessary condition for the existence of the steady state is that the real parts of the eigenvalues of L be positive definite. There is a single sixfold degenerate eigenvalue, $\lambda(a) = 2\alpha(a) + \nu$. Since $\alpha(a)$ is positive for all values of the shear rate, the eigenvalue is positive as well. The steady state uniform shear flow second degree moments are then $x(\infty) = (L^{-1}b)T(\infty)$.

This approach extends to the analysis of moments of higher degree as well. There are ten independent moments of third degree that obey a linear first order equation similar to (7). It is found that the eigenvalues of the corresponding linear matrix are again positive for all values of the shear rate and the moments go to zero for long times [6]. In the following we focus on the analysis of the moments of degree 4. There are fifteen independent moments, which decouple into two subsets of six and nine moments, respectively. The moments in the first subset tend to zero in the long time limit [6]. The remaining nine moments define a column matrix

$$\begin{aligned} y_\sigma \longleftrightarrow \{&\langle\xi^4\rangle, \langle\xi^2(\xi_x^2 - \tfrac{1}{3}\xi^2)\rangle, \langle\xi^2(\xi_y^2 - \tfrac{1}{3}\xi^2)\rangle, \langle\xi_x^4 - \tfrac{6}{7}\xi^2(\xi_x^2 - \tfrac{1}{10}\xi^2)\rangle, \langle\xi_y^4 - \tfrac{6}{7}\xi^2(\xi_y^2 - \tfrac{1}{10}\xi^2)\rangle, \\ &\langle\xi_z^4 - \tfrac{6}{7}\xi^2(\xi_z^2 - \tfrac{1}{10}\xi^2)\rangle, \langle\xi^2\xi_x\xi_y\rangle, \langle\xi_x\xi_y(\xi_x^2 - \tfrac{3}{7}\xi^2)\rangle, \langle\xi_x\xi_y(\xi_y^2 - \tfrac{3}{7}\xi^2)\rangle\} \ . \end{aligned} \qquad (10)$$

The linear equation for $y(t)$ determined from (3) is

$$\left(\frac{\partial}{\partial t}+\mathcal{L}\right)y(t)=B(t)\ , \tag{11}$$

with the formal solution

$$y(t)=e^{-\mathcal{L}t}y(0)+\int_0^t d\tau e^{-\mathcal{L}\tau}B(t-\tau)\ . \tag{12}$$

Here $B_\sigma(t)$ is a matrix depending on moments of lower degree, now known, which approaches a constant value for large t. Define $c_1(a)\equiv 4\alpha(a)+\frac{2}{3}\nu$, $c_2(a)\equiv 4\alpha(a)+\frac{7}{6}\nu$, $c_3(a)\equiv 4\alpha(a)+2.097\nu$. Then the elements of the constant matrix $\mathcal{L}$ are given by

$$\mathcal{L}=\begin{pmatrix} c_1 & 0 & 0 & 0 & 0 & 0 & 4a & 0 & 0 \\ 0 & c_2 & 0 & 0 & 0 & 0 & \frac{32}{21}a & 2a & 0 \\ 0 & 0 & c_2 & 0 & 0 & 0 & -\frac{10}{21}a & 0 & 2a \\ 0 & 0 & 0 & c_3 & 0 & 0 & -\frac{96}{245}a & \frac{16}{7}a & 0 \\ 0 & 0 & 0 & 0 & c_3 & 0 & -\frac{96}{245}a & 0 & \frac{12}{7}a \\ 0 & 0 & 0 & 0 & 0 & c_3 & \frac{24}{245}a & \frac{12}{7}a & \frac{12}{7}a \\ \frac{7}{15}a & \frac{2}{7}a & \frac{9}{7}a & -a & -a & -a & c_2 & 0 & 0 \\ 0 & \frac{15}{49}a & -\frac{6}{49}a & -\frac{15}{14}a & -\frac{15}{14}a & \frac{15}{14}a & 0 & c_3 & 0 \\ 0 & -\frac{6}{49}a & \frac{15}{49}a & \frac{3}{7}a & \frac{10}{7}a & -\frac{3}{7}a & 0 & 0 & c_3 \end{pmatrix}\ . \tag{13}$$

There are nine different eigenvalues of $\mathcal{L}$, with three sets of complex conjugate pairs. Figure 1 shows the six real parts of these eigenvalues as a function of the shear rate. The eigenvalue denoted by $\ell_1(a)$ decreases to zero at the critical value $a_c\simeq 5.178\nu$. This implies that all moments approach finite steady state values as $t\to\infty$ for aa_c, $y(\infty)=\mathcal{L}^{-1}B(\infty)$. Conversely, all fourth degree moments grow without bound as $t\to\infty$ for $a\geq a_c$. As an illustration, Fig. 2 shows the steady state value of $\langle\xi^4\rangle$ relative to its Maxwell-Boltzmann value for $a<a_c$, indicating a divergence as a approaches a_c. For $a>a_c$ there are in principle two possibilities: (1) stationary solutions to Eq. (3) exist, but moments of degree 4 or higher diverge; (2) stationary solutions to (3) do not exist. In the first case we would expect to discover the divergent moments from the stationary solution $y(\infty)=\mathcal{L}^{-1}B(\infty)$. However, the elements of both $\mathcal{L}$ and $B(\infty)$ remain finite for $a>a_c$; instead of divergent moments the solutions to this equation are unphysical (e.g., $\langle\xi^4\rangle<0$). Thus we conclude that stationary uniform shear flow is limited to $a<a_c$.

What is the nature of the solution of the Boltzmann equation (1) for $a>a_c$? The most interesting possibility is that a stationary normal solution still exists, but whose space dependence is not as simple as that assumed for uniform shear flow. As a normal solution, this space dependence would be determined entirely by the hydrodynamic fields. The transition from uniform shear flow to this new solution would be associated with an instability of the shear flow hydrodynamics, as determined by small spatial perturbations, at a certain shear rate smaller than or equal to a_c. Above that shear rate a different stable hydrodynamic solution would exist, characterizing the new normal solution to the Boltzmann equation. The instability of the second degree moments to spatial perturbations is reflected in the moments of higher degree for the uniform state. A possible precurser of new spatial structure may be the increasing anisotropy of these fourth degree moments as $a\to a_c$. For instance, $\langle\xi_x^4\rangle$ is about 50 times larger than $\langle\xi_y^4\rangle$ and 80 times larger than $\langle\xi_z^4\rangle$. The value of a_c determined from the fourth degree moments is only an upper bound for this transition between normal solutions, since it is possible that solutions to higher degree moment equations have stronger restrictions.

Resolution of these questions from the theoretical side might be accomplished from a linear stability analysis of (3), extended to include small spatial variations rel-

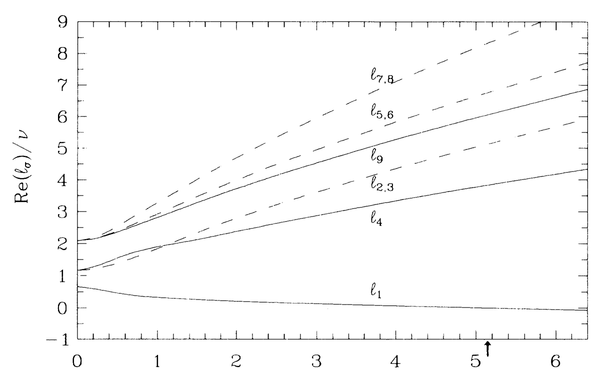

FIG. 1. Shear rate dependence of the real parts of the eigenvalues $\ell_\sigma(a)$ of the matrix $\mathcal{L}$. The solid (dashed) lines correspond to the real (complex) eigenvalues. The arrow indicates the location of the critical shear rate a_c.

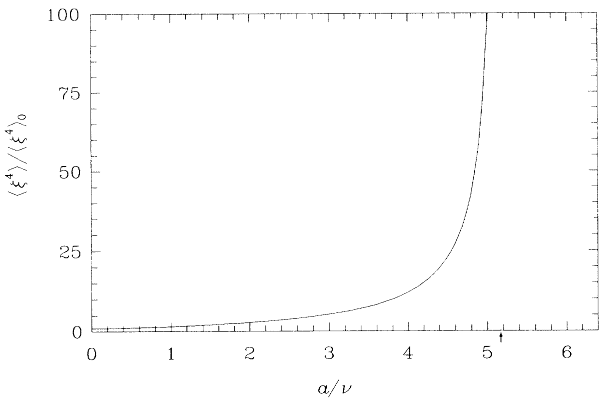

FIG. 2. Shear rate dependence of the steady state value of the moment $\langle \xi^4 \rangle$ relative to its Maxwell-Boltzmann value $\langle \xi^4 \rangle_0$. The arrow indicates the location of the critical shear rate a_c.

ative to uniform shear flow. This stability analysis can be transformed to a more practical form via the corresponding moment equations for spatially inhomogeneous solutions. A related analysis of a possible long wavelength hydrodynamics instability for uniform shear flow, based on approximate moment equations for the Boltzmann equation, has been given by Loose and Hess [7]. Using a rheological equation of state from simulations they indeed find an instability; however, use of the corresponding equation of state from the Boltzmann equation solution does not lead to an instability. A more accurate stability analysis based on the inhomogeneous Boltzmann equation is required before any firm correlation between the moment divergence and a hydrodynamic instability can be claimed.

Complementary numerical studies are both practical and potentially more illuminating. Molecular dynamics simulation at low density for comparison with Boltzmann equation predictions already has been applied to this problem, although at lower shear rates [8]. A more direct numerical construction of the solution to the Boltzmann equation is possible using the Monte Carlo simulation method of Bird [9]. This is a method suited to states far from equilibrium and whose accuracy is well established. It has been applied recently to shear flow for comparison with the shear rate dependence of the viscosity calculated from the Ikenberry-Truesdell solution, with good agreement [10]. Since the predicted value of the critical shear rate discussed here is precise, there should be no difficulty in locating the singularity by these numerical methods.

Finally, we note that a transition from uniform shear flow to an ordered state at large shear rates has been observed in molecular dynamics simulations for dense fluids [11]. The simulations for simple atomic potentials (hard spheres, Lennard-Jones) show an ordering of the particles into strings, hexagonally packed, and directed along the flow [7,12,13]. It has been suggested that this transition is also due to a hydrodynamic instability [14], although liquid structure plays an essential role in the origin of these instabilities at short wavelengths. The Boltzmann equation is limited to longer wavelengths and thus is not directly applicable, even if density effects could be scaled. However, it is possible that a remnant of this transition remains at low density although presumably at higher shear rates (this is the spirit of the low density calculation by Loose and Hess [7]).

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