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published in
Physical Review E
2002

document version
Publisher's PDF, also known as Version of record

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citation for published version (APA)
Ajdari, A., Cates, M. E., & Head, D. A. (2002). Rheological chaos in a scalar shear-thickening model. *Physical Review E*, *66*(2), 025202.

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PHYSICAL REVIEW E **66**, 025202(R) (2002)

Rheological chaos in a scalar shear-thickening model

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(Received 7 April 2002; published 27 August 2002)

We study a simple scalar constitutive equation for a shear-thickening material at zero Reynolds number, in which the shear stress σ is driven at a constant shear rate $\dot{\gamma}$ and relaxes by two parallel decay processes: a nonlinear decay at a nonmonotonic rate $R(\sigma_1)$ and a linear decay at rate $\lambda\sigma_2$. Here $\sigma_{1,2}(t) = \tau_{1,2}^{-1}\int_0^t \sigma(t')\exp[-(t-t')/\tau_{1,2}]dt'$ are two retarded stresses. For suitable parameters, the steady state flow curve is monotonic but unstable; this arises when $\tau_2 > \tau_1$ and $0 > R'(\sigma) > -\lambda$ so that monotonicity is restored only through the strongly retarded term (which might model a slow evolution of the material structure under stress). Within the unstable region we find a period-doubling sequence leading to chaos. Instability, but not chaos, persists even for the case $\tau_1 \to 0$. A similar generic mechanism might also arise in shear thinning systems and in some banded flows.

DOI: 10.1103/PhysRevE.66.025202 PACS number(s): 05.45.Ac, 83.60.Rs, 83.10.Gr

Rheochaos can be defined as the occurrence of macroscopic chaos [1] in a viscoelastic material at a negligible Reynolds number. With the neglect of inertia that this implies, the nonlinearity must come not from the advection of momentum (as in the Navier-Stokes turbulence) but from the constitutive behavior of the material, which may include strong memory effects. Likewise, for the chaos to be macroscopically observable (for example in time series data on the stress measured at a fixed strain rate, or vice versa, in a bulk sample) a mechanism must be present that goes beyond the microscale chaos known to be present in, e.g., colloidal Stokes flow [2].

Strong candidates for rheochaos include micellar materials [3], dense lamellar phases [4], and also dense suspensions where erratic stress response at fixed strain rate (or vice versa) is widespread but poorly documented (see, e.g., Ref. [5]). It is not yet clear whether spatial as well as temporal inhomogeneity is present for all instances of rheochaos, and if so to what extent. This could range from a shear-banded flow in which the interface between the bands of the fast and slow flowing materials is unsteady in time (as suspected in micelles [3,6]) through to fully developed ''elastic turbulence'' as recently reported in polymer solutions near the overlap threshold [7]. Spatial inhomogeneities are also known to occur in shear-thickening colloid solutions [5,8]. However, the closely related phenomenon of director chaos in sheared nematics has been studied theoretically and does not seem to require spatial inhomogeneity [9]. In the present state of understanding, a theoretical search for temporal rheochaos in spatially homogenous models remains justified.

Recent work by the authors has studied the onset of temporal instability in spatially homogeneous mesoscopic models of the shear-thickening type [10]. One interesting prediction was that such instability could arise in a system where the steady state flow curve $\sigma(\dot{\gamma})$ is monotonic [10]. This contrasts with the conventional instability to spatial inhomogeneity in the form of shear bands: this is always associated with regions of negative slope on the flow curve [11–13]. The mesoscopic models of [10] are not fully tensorial but work with a single (spatially uniform) component of each of the stress and strain rate tensors (σ and $\dot{\gamma}$); nonetheless they contain an infinite number of degrees of freedom, corresponding to the distribution of local strain variables for different mesoscopic elements. This makes them complex to analyze.

In this paper we propose closely related but much simpler models in which there is only one degree of freedom (the shear stress σ) whose time evolution at constant strain rate $\dot{\gamma}$ is governed by a simple constitutive equation with retarded and nonlinear features. The simplest such model combines a nonlinear instantaneous relaxation rate for stress (chosen nonmonotonic) with a linear but retarded relaxation. For a single exponential retardation kernel, its dynamics can be completely understood: it shows spontaneous oscillation in a region of the flow curve with a positive slope, but no chaos. This is qualitatively like the mesoscopic model of Ref. [10] (although that model exhibits oscillations at a constant imposed stress rather than strain rate). In particular, the instability is associated with a negative slope on the ''bare'' flow curve (before the retarded term is added). A second, similar model, in which the nonlinear relaxation is itself delayed, shows chaos.

We first examine the simplest model alluded to above. This is defined by the equation

$$\dot{\sigma}(t) = \dot{\gamma} - R(\sigma) - \lambda\sigma_2, \tag{1}$$

where $\sigma_2(t) = \int_{-\infty}^{t} M_2(t-t')\sigma(t')\,dt'$ is a retarded stress and $M_2(t)$ is a memory kernel whose integral is unity. The first term on the right-hand side of this equation means that, in the absence of relaxation, stress increases linearly with straining (the elastic constant is set to unity)—a Hookean solid. The second term describes the instantaneous decay of stress at rate $R(\sigma)$, for example, through ''hops'' or plastic rearrangement of mesoscopic elements (returning these to an unstrained state) with jump rate R/σ. Unlike in the mesoscopic models of Ref. [10], no attempt is made to track the dynamics of individual elements. The third term is also a decay term, but describes retarded relaxation. This could represent ''delayed jumps'' which, perhaps because they involve

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a cooperative motion of many elements, take a distribution of finite times to accomplish (governed by the kernel M_2). More generally, a retarded term could represent some other slow structural reorganization of the material in response to stress.

For example, one could have a model of instantaneous jumps but with a "fluidity" or jump rate that itself adapts slowly to stress [14]. In this context it might be more natural to have a nonlinear retarded term such as

$$\dot{\sigma}=\dot{\gamma}-R(\sigma)-\lambda\sigma_2\sigma. \tag{2}$$

However, this gives qualitatively the same instability as described below for Eq. (1) [15]; we retain the linear version, for simplicity, below.

Solving Eq. (1) in the steady state gives immediately the flow curve, or rather its inverse,

$$\dot{\gamma}=R(\sigma)+\lambda\sigma. \tag{3}$$

The interesting case is when $R(\sigma)$ is nonmonotonic but $R(\sigma)+\lambda\sigma$ is monotonic. Then the flow curve is monotonic, but only because of the retarded contribution to the jump rate. One might suspect that a sufficiently sluggish retarded contribution might fail to correct the underlying instability in the region where $R'(\sigma)$ is negative: over short timescales the system appears to be unstable with respect to shear banding but at long time scales it is not. Here, the time scales are measured relative to the strain rate at which $R(\sigma)$ in Eq. (3) first becomes nonmonotonic; we choose units so that this is $O(1)$.

We analyze the case of a single exponential kernel, $M_2=\tau_2^{-1}\exp[-(t-t')/\tau_2]$. As is easily checked, for this kernel Eq. (1) can be replaced by a differential equation of second order. Differentiating Eq. (1) with respect to t, and noting that $\dot{\sigma}_2=(\sigma-\sigma_2)/\tau_2$, we obtain immediately

$$\ddot{\sigma}=-(\partial V/\partial\sigma)-\xi(\sigma)\dot{\sigma}, \tag{4}$$

which effectively describes a particle of unit mass in a one-dimensional potential V with damping constant ξ. Here

$$\tau_2 V(\sigma)=\int_0^{\sigma}R(\sigma')d\sigma'+\lambda\sigma^2/2-\dot{\gamma}\sigma, \tag{5}$$

$$\xi(\sigma)=R'(\sigma)+1/\tau_2. \tag{6}$$

As $\dot{\gamma}$ is varied, the steady state flow curve $\sigma(\dot{\gamma})$, as given by Eq. (3), is recovered as the solution of $V'(\sigma=0)$. The stability of the steady state solution requires that two further conditions are satisfied. The first is $V''(\sigma)>0$ (so that the effective potential has a minimum not a maximum). This is equivalent to $d\sigma/d\dot{\gamma}>0$ which is the usual criterion to avoid shear banding. However, the stability also requires that $\xi(\sigma)$ is positive at the minimum of V. When $R'(\sigma)$ in Eq. (6) is negative, this is only satisfied if the retardation time τ_2 is sufficiently short. When not satisfied, one has antidamping at the minimum of V so that small velocity fluctuations are amplified; this is reminiscent of a van der Pol oscillator [16]. Velocity fluctuations will grow until a limit cycle is reached

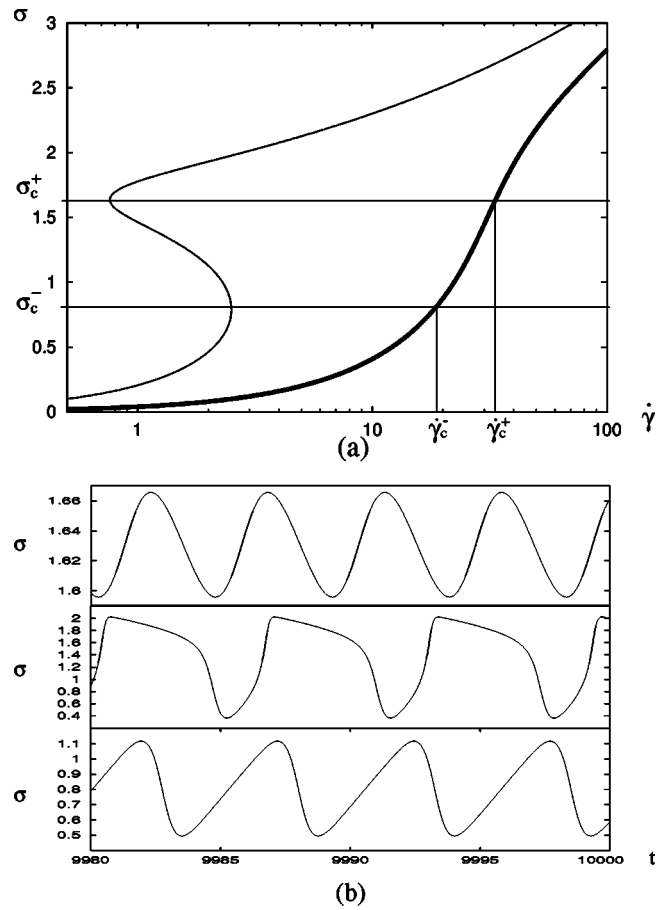

FIG. 1. (a) The bare flow curve $\dot{\gamma}=R(\sigma)$ (light line) and the final flow curve (heavy line) in the model of Eq. (1). Parameters are $\lambda=20$, $\tau_2=10$, and $R(\sigma)=0.6\sigma^5-3.3\sigma^3+5\sigma$. The region of instability $\sigma_c^-<\sigma<\sigma_c^+$ is shown, where $\sigma_c^-\approx0.799$ and $\sigma_c^+\approx1.631$, corresponding to $\dot{\gamma}_c^-\approx18.487$ and $\dot{\gamma}_c^+\approx33.382$. Note that for our choice of parameters, $\sigma_c^{\pm}$ almost coincide with the turning points of R. (b) Stress time series (same parameter values) at (from bottom to top) $\dot{\gamma}=18.49$, $\dot{\gamma}=30$, and $\dot{\gamma}=33.38$.

in which the positive damping at large amplitudes balances the antidamping near the minimum.

Examples of the "bare" flow curve, the final flow curve, and the region of the instability are shown in Fig. 1(a). Figure 1(b) shows a typical time series of the stress just inside, and well within, the unstable region. The limits of this region, $\sigma_c^{\pm}$, are Hopf bifurcation points where there is an onset of finite frequency sinusoidal oscillations with an amplitude varying as $|\dot{\gamma}-\dot{\gamma}_c|^{1/2}$.

Our choice of an exponential kernel is nongeneric: most integral kernels are not equivalent to any finite-order differential equation [16]. However, the above argument gives a generic mechanism of instability. If the flow curve is monotonic only because of a retarded term $[-\lambda<R'(\sigma)<0]$, then temporal instability survives if the retardation time is too long. Its presence does not depend on details of the kernel, but what it leads to might do so: in particular, chaos is impossible in a second-order system [16] such as Eq. (4). However, our finding of spontaneous oscillation but not chaos appears to be structurally stable: we were unable to

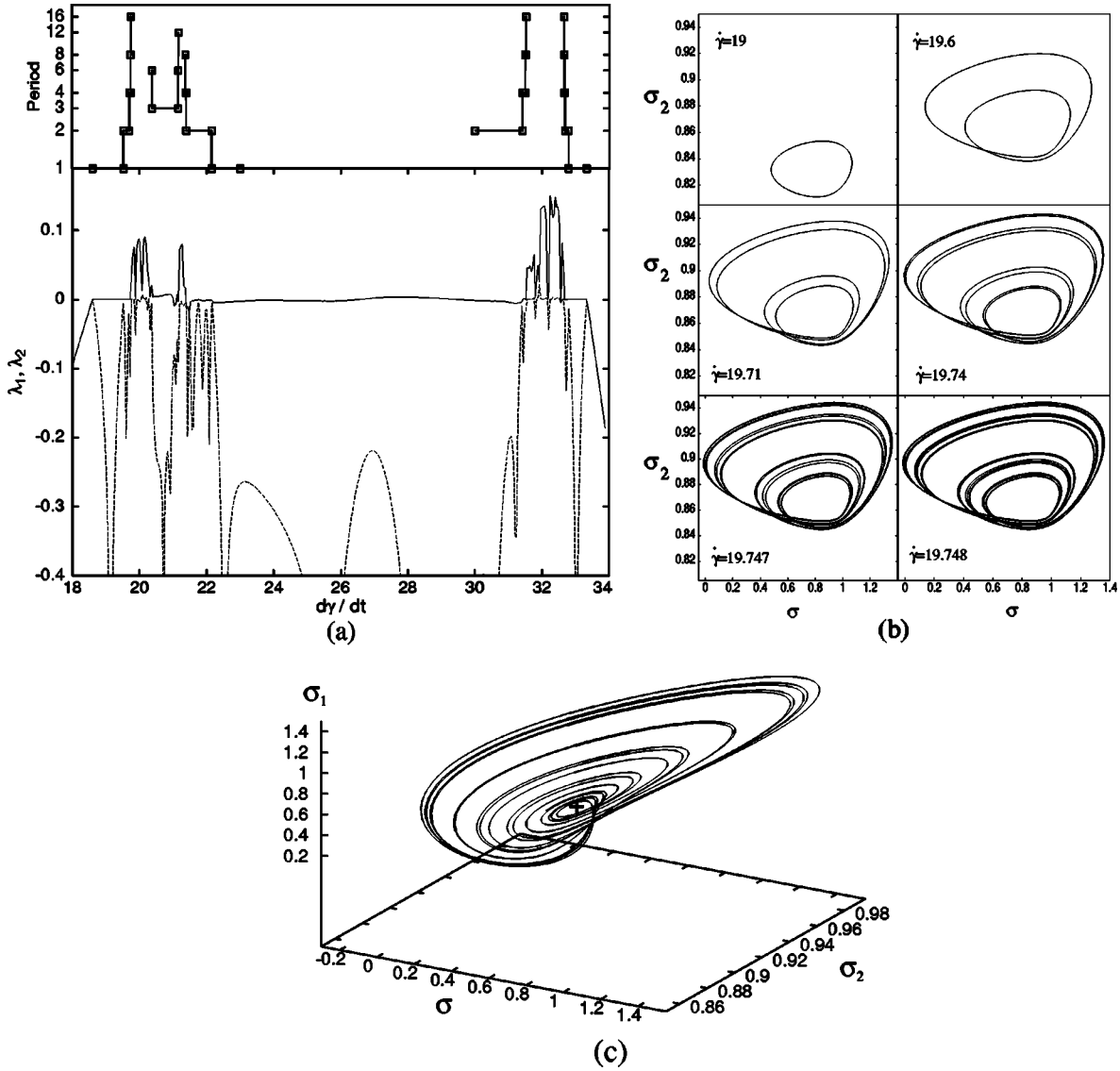

FIG. 2. (a) Upper plot: the period of stable orbits as a function of strain rate $\dot\gamma$ around the unstable region of the flow curve of Fig. 1, for the model of Eq. (7) with $\tau_1=0.5$ and the other parameters as in Fig. 1. Lower plot: Lyapunov exponents for trajectories, showing $\lambda_1>0=\lambda_2$ in the chaotic regions. (b) Orbits projected onto the (σ_2,σ) plane for various $\dot\gamma$ showing the period-doubling cascade with periods 1, 2, 4, 8, 16, and 32. (c) The strange attractor in $(\sigma_1,\sigma_2,\sigma)$ space for $\dot\gamma=20$ over a time period $5\times10^2<t<10^3$ (arb. units).

find chaos with M_2 taken as the sum of two exponentials (which gives a third-order dynamical system for which chaos is allowed).

In that case, what needs to be added to the model of Eq. (1) to give temporal chaos rather than just spontaneous oscillation? So far, the simplest variant we have found that definitely shows chaos is the following:

$$\dot\sigma(t)=\dot\gamma-R(\sigma_1)-\lambda\sigma_2, \tag{7}$$

where the stress in the nonlinear term, σ_1, is now also retarded. The steady state flow curve is the same as that for Eq. (1). For simplicity, we choose a single exponential kernel here too: $\sigma_1(t)=\int_0^t\sigma(t')\tau_1^{-1}\exp[-(t-t')/\tau_1]\,dt'$. To maintain continuity of interpretation with the simpler version of the model, we choose $\tau_1\lesssim1\ll\tau_2$. We study the situation where the monotonicity of the flow curve [still given by Eq. (3)] is restored only via the more retarded one of the two relaxation terms. While there is no longer a simple interpretation in terms of an effective potential or a damping function, the generic instability of the previous model remains. But now, within the unstable region, we find a period doubling cascade leading to chaos. Figure 2(a) shows, for a specified set of model parameters, the period and Lyapunov exponents $\lambda_1\geqslant\lambda_2\geqslant\lambda_3$ as a function of $\dot\gamma$ ($\lambda_1>0$ means that nearby trajectories exponentially separate [17]); Fig. 2(b) shows a series of period-doubling orbits in the (σ_2,σ) plane and Fig. 2(c) shows the strange attractor in $(\sigma_1,\sigma_2,\sigma)$ space. Its Lyapunov dimension $D_{\rm Lyap}=2+\lambda_1/|\lambda_3|$ varies with the parameters but is slightly greater than 2, typically $2.0<D_{\rm Lyap}<2.1$.

Physically it is not clear to us yet why the retardation of the nonlinear term (as well as the linear one) seems necessary to get chaos out of Eq. (7); presumably, however, this adds something which is missing even from the mesoscopic model of Ref. [10] (where chaos remained absent despite the infinite order of the system). Attempts to associate the retarded stresses in this model with, say, higher moments of the distribution of local strains in the model of Ref. [10] (where the first moment is the instantaneous stress) have so far proved unconvincing. A more detailed study is left for future work.

We conclude with a broader discussion. The key idea is that of a flow curve (for spatially homogeneous states) whose monotonicity is rescued only by a retarded contribution; if too much retarded, this does not restore temporal stability because the system continues to amplify perturbations over short time scales. Although the equations involved will look rather different, very similar physics could arise in materials of the shear-thinning type where shear banding is present [6,12,18] or narrowly avoided [19]. It might be very interesting to look more closely in shear-thinning micellar systems where, by varying density and temperature, one can arrange a material whose flow curve is only just monotonic [18]. Similar studies in colloids close to the transition from continuous to discontinuous shear thickening [5] would also be valuable, although this field is a lot less developed experimentally.

Quite similar equations, but with different variables and interpretation, might describe a preexisting shear-banded flow, whose stability remains unclear in many cases [20]. The simplest scenario would ascribe a single coordinate to describe the bands (e.g., the position of the interface between them, assumed flat) and seek to develop equations for its time evolution. Chaotic behavior of such an interface, rather than of a spatially homogeneous stress, might be the explanation of rheochaos seen in various micellar systems [3]. In the case where one of the bands is a static gel, empirical models such as those proposed in Refs. [21] have met with some success at explaining the observed (though not entirely steady [22]) dependence of stress on the strain rate when averaged across such a banded flow. Such models involve equations such as $\dot{h}=f(h)-1/\sigma$ where h is the width of a shear band, f is a nonlinear term arising from the difference in concentrations of the two bands, and σ is the stress [21]. Under controlled strain rate conditions (say) $1/\sigma$ is linear in h and the equation is not dissimilar to Eq. (1) without retardation. If a slow process can be identified (possibly concentration equilibration), then a retarded version of this type of equation could share the generic instability of the models discussed above.

M.E.C. and D.A.H. acknowledge the hospitality of the Institute for Theoretical Physics, UCSB, where part of this work was completed with the support of NSF Grant No. PHY99–07949. A.A. acknowledges the hospitality and support of the University of Edinburgh, where this work was initiated.

[1] Chaos is used here to mean bounded unsteady motion in a deterministic system that is neither periodic nor quasiperiodic, with trajectories that separate locally. We make no assumption about whether the chaos is low or high dimensional.

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