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Nonlinear Fluid Dynamics Description of non-Newtonian Fluids

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Nonlinear hydrodynamic equations for visco-elastic media are discussed. We start from the recently derived fully hydrodynamic nonlinear description of permanent elasticity that utilizes the (Eulerian) strain tensor. The reversible quadratic nonlinearities in the strain tensor dynamics are of the 'lower convected' type, unambiguously. Replacing the (often neglected) strain diffusion by a relaxation of the strain as a minimal ingredient, a generalized hydrodynamic description of viscoelasticity is obtained. This can be used to get a nonlinear dynamic equation for the stress tensor (sometimes called constitutive equation) in terms of a power series in the variables. The form of this equation and in particular the form of the nonlinear convective term is not universal but depends on various material parameters. A comparison with existing phenomenological models is given. In particular we discuss how these ad-hoc models fit into the hydrodynamic description and where the various non-Newtonian contributions are coming from.

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I. INTRODUCTION

Hydrodynamics is a well established field to describe macroscopically simple fluids by means of the Navier-Stokes-, continuity, and heat conduction equations. However, it applies also to more complex fluids that are fully characterized by conservation laws and broken symmetries. This more general hydrodynamic method has been established in the 60s [1–3] and applied e.g. to superfluids [4] and liquid crystals [5]. It is based on (the Gibbsian formulation of) thermodynamics [6, 7], symmetries and well-founded physical principles [8]. A detailed description of this method can be found in [5, 9]. Somewhat related approaches have been used for liquid crystals [10–13] and more generally in [14, 15].

On the other hand, non-Newtonian fluids are believed to show non-universal behavior and a host of different empirical models have been proposed [16–21] to cope with the flow rheology of such substances. Typically these models are formulated as generalizations of the linear, Newtonian relation between stress and deformational flow allowing for additional time derivatives and nonlinearities. They are tailored to accommodate empiric findings or are based on principles [20] that are ad-hoc and generally insufficient.

Quite recently we have derived a nonlinear hydrodynamic description of elastic media [22, 23] that is based on first principles, only, making use of thermostatics, linear irreversible thermodynamics, symmetries and broken symmetries, and invariance principles. It has been confirmed within the GENERIC formalism [24]. Allowing in this hydrodynamic description the strains to relax (and not only to diffuse) a generalized hydrodynamic description of nonlinear viscoelasticity is obtained in terms of a dynamic equation for the strain tensor [22, 23]. After a thorough exposition of this formulation (Sec.II) we transform it into a description in terms of a dynamic equation for the stress tensor (Sec.III). This can only be done approximately in the form of a power expansion in the variables. Up to second order a formulation is obtained that can directly be compared with many of the empirical models proposed to describe non-Newtonian rheology. The comparison (Sec.IV) reveals possible inconsistencies and connects the various ad-hoc additions of those models with physical relevant processes, like strain relaxation, elasticity and viscosity. In two appendices we sketch possible extensions of the hydrodynamic description of viscoelasticity.

A comparison with recent constitutive equations that refer to specific microscopic variables and processes, like convective constraint release [25–27], will not be done in the present paper. Here we rather concentrate on the simplest generalized hydrodynamic description of non-Newtonian rheology in terms of a relaxing strain field, while a detailed comparison with those theories requires the use of additional relaxing fields.

II. STRAIN TENSOR DESCRIPTION

In this section we review the hydrodynamic description of nonlinear elasticity and its generalization to viscoelasticity in terms of the Eulerian strain tensor U_{ij}. We start with a short reminder of its definition. In a stress-free elastic body, we consider a point at the (initial) coordinate $\mathbf{a}$. As the body is displaced, rotated, compressed and sheared, the given point is displaced to $\mathbf{r}$. Since all points of the body have a unique pair of $\mathbf{a}$ and $\mathbf{r}$, the function $\mathbf{r}(\mathbf{a})$ is unique and invertible, the result of which we denote as $\mathbf{a}(\mathbf{r})$. Describing physics in local terms, in particular, the state variables characterizing the elastic body are taken as functions of the real space coordinates $\mathbf{r}$ rather than of the reference space coordinates $\mathbf{a}$. Therefore we choose $\boldsymbol{a}(\boldsymbol{r})$ rather than $\boldsymbol{r}(\boldsymbol{a})$ as the starting point to construct the (Eulerian) strain tensor $U_{ij} = \frac{1}{2}[\delta_{ij} - (\nabla_j a_k)(\nabla_i a_k)]$, which can be written in the more familiar form $U_{ij} = \frac{1}{2}[\nabla_j u_i + \nabla_i u_j - (\nabla_i u_k)(\nabla_j u_k)]$ using the displacement field $u_i(\boldsymbol{r}) = r_i - a_i(\boldsymbol{r})$. In the Lagrangian description, where all fields are function of the initial coordinates rather than the local ones, the (Langrangian) strain tensor is $U^L_{ij} = \frac{1}{2}[(\partial r_k/\partial a_j)(\partial r_k/\partial a_i) - \delta_{ij}]$ or $U^L_{ij} = \frac{1}{2}[\partial u_i/\partial a_j + \partial u_j/\partial a_i + (\partial u_k/\partial a_i)\cdot(\partial u_k/\partial a_j)]$, with $u_i(\boldsymbol{a}) = r_i(\boldsymbol{a}) - a_i$. Since the dynamics for $\boldsymbol{a}$ is simply $d\boldsymbol{a}/dt = 0$ in the absence of phenomenological currents, the dynamics for the Eulerian strain tensor reads [22, 23]

$$\frac{d}{dt}U_{ij} - A_{ij} + U_{ki}\nabla_j v_k + U_{kj}\nabla_i v_k = X^{(ph)}_{ij} \tag{1}$$

with $d/dt \equiv \partial/\partial t + \boldsymbol{v}\cdot\boldsymbol{\nabla}$, where v_i is the velocity and $A_{ij} = \frac{1}{2}(\nabla_i v_j + \nabla_j v_i)$ its symmetrized gradient describing deformational flow. This equation contains, apart from the transport derivative $\sim \boldsymbol{v}\cdot\boldsymbol{\nabla}$ due to Galilean translational invariance, a linear and a nonlinear coupling to flow. The former ($\sim A_{ij}$) reflects the spontaneously broken translational symmetry in elastic media (in an equation for the displacement vector the equivalent term subtracts the solid body translations, which must not change the elastic state of the fluid). The nonlinear one is of the lower convected type and results from the freedom to choose the orientation of a material-fixed frame independently from that of the laboratory frame [22, 23]. For a Lagrangian strain tensor the upper convected derivative would be obtained. On the right hand side there is the phenomenological (quasi-)current $X^{(ph)}_{ij}$, which describes, in the case of permanent elasticity, purely diffusional processes. In that case Eq.(1), when combined with the dynamic equations for the other hydrodynamic variables, the density, ρ, the energy density, ϵ, and the density of linear momentum, g_i, gives rise to three additional truly hydrodynamic modes, i.e. the two doubly degenerated transverse sound modes (compared to the two vorticity diffusions of simple fluids) and vacancy diffusion [5]. For decaying strains in viscoelastic media $X^{(ph)}_{ij}$ contains relaxational processes (see below). The strain tensor U_{ij} is symmetric, in order to exclude solid body rotations, since the latter must not change the elastic state.

The second relevant dynamic equation is the momentum balance or generalized Navier-Stokes equation

$$\rho\frac{d}{dt}v_i + \nabla_i p + \nabla_j \sigma_{ij} = 0 \tag{2}$$

with the isotropic pressure p and the stress tensor σ_{ij}, which is of the form

$$\sigma_{ij} = -\Psi_{ij} + \Psi_{ki}U_{jk} + \Psi_{kj}U_{ik} + \sigma^{(ph)}_{ij} \tag{3}$$

Here Ψ_{ij} is the *elastic* stress tensor, the thermodynamic conjugate to the strain tensor. The linear and nonlinear Ψ_{ij} contributions are the counterterms to the linear and nonlinear flow contributions in Eq. (1) that are necessary to cancel any contribution to the entropy production [9], since all these terms are reversible. Thus, these parts of the stress tensor are completely fixed by general physical principles. The phenomenological part of the stress tensor is contained in $\sigma^{(ph)}_{ij}$ (see below) and describes in the simplest form Newtonian viscosity. The stress tensor is symmetric (or can be made so) in order to guarantee angular momentum conservation [5].

Throughout this paper we will assume incompressibility that reduces the continuity or mass conservation equation to div$\boldsymbol{v} = 0$. In addition we neglect the thermal degree of freedom and, thus, the heat conduction equation.

Phenomenological or material specific properties enter our equations on three different occasions, two are dynamic and one is static in nature. For the former we stay within the reach of well-founded linear irreversible thermodynamics [28], i.e. we assume a linear relation between the "forces" Ψ_{kl} and A_{kl} and the "fluxes" $X^{(ph)}_{ij}$ and $\sigma^{(ph)}_{ij}$

$$X^{(ph)}_{ij} = -\alpha_{ijkl}\Psi_{kl} \tag{4}$$

$$\sigma^{(ph)}_{ij} = -\nu_{ijkl}A_{kl} \tag{5}$$

describing strain relaxation and viscosity, respectively. By allowing nonlinear dependences of the 'fluxes" on the "forces", one would leave the solid grounds of well-established statistical physics, since not very much is known on the validity range of such theories. Nevertheless, the relations (4, 5) are nonlinear in the sense that the material tensors depend on the variables of the systems, in particular on temperature T, pressure and the strain tensor. The dependence on the scalar quantities is rather trivial and will not be shown explicitly, while the strain tensor dependence is more complicated. Starting from an equilibrium state $U_{ij} = 0$ we can expand the material tensors into powers of U_{ij}. Up to quadratic order in Eqs.(1,2), which is what we need for the comparison in Chapt. IV, we find for the general form of the rank 4 material tensors

$$\begin{aligned} \alpha_{ijkl} &= \frac{\alpha_1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + \frac{\alpha_2}{2}(U_{ik}\delta_{jl} + U_{jk}\delta_{il} + U_{il}\delta_{jk} + U_{jl}\delta_{ik}) + \alpha_3\delta_{ij}\delta_{kl} + \\ &\quad \alpha_4(\delta_{ij}U_{kl} + \delta_{kl}U_{ij}) + \alpha_5\delta_{ij}\delta_{kl}U_{pp} + \alpha_6(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})U_{pp}. \end{aligned} \tag{6}$$

Since the "fluxes" are derived from a potential (the entropy production) by partial derivation, the symmetries of the tensors A_{ij} and Ψ_{ij} are transferred to $\sigma_{ij}^{(ph)}$ and $X_{ij}^{(ph)}$, respectively and are reflected in the material tensors being symmetric under the interchange of $i-j$, $k-l$, and $ij-kl$. For the viscosity tensor ν_{ijkl} we can simplify the form (6), since $A_{kk} = 0$. Thus, all parts $\sim \delta_{ij}$ or δ_{kl} are projected out in the entropy production and only the coefficients $\nu_{1,2,6}$ appear in Eq.(5). Note that neither U_{ij}, nor Ψ_{ij}, nor the total stress tensor σ_{ij} have to be traceless, despite A_{ij} being so. If desired, the expansion (6) can easily be continued to arbitrary order. A more general form of Eqs.(4,5) is discussed in Appendix A. In the main part of this paper we will stick to the simplest form given above that constitutes the minimal generalization of hydrodynamics, but still captures viscoelasticity.

The static phenomenological properties are expressed by a relation between the strain and the elastic stress

$$\Psi_{ij} = K_{ijkl}U_{kl} \tag{7}$$

where the elastic tensor K_{ijkl} generally depends on all variables, in particular on the strain tensor itself. In second order its form is slightly different from that of the α tensor, since it is derived from an elastic energy, which is cubic in the strain tensor, resulting in the constraint $2K_6 = K_4$

$$\begin{aligned} K_{ijkl} &= \frac{K_1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + \frac{K_2}{2}(U_{ik}\delta_{jl} + U_{jk}\delta_{il} + U_{il}\delta_{jk} + U_{jl}\delta_{ik}) + K_3\delta_{ij}\delta_{kl} + \\ &\quad K_4(\delta_{ij}U_{kl} + \delta_{kl}U_{ij} + \frac{1}{2}[\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}]U_{pp}) + K_5\delta_{ij}\delta_{kl}U_{pp}. \end{aligned} \tag{8}$$

Thus we are left with two linear elastic moduli $K_{1,3}$ and three quadratic ones $K_{2,4,5}$. To ensure positivity of the elastic energy, from which (7) is derived, third order terms with large and positive elastic moduli are necessary (see Appendix A).

III. STRESS TENSOR DESCRIPTION

In the preceding section viscoelastic hydrodynamics was expressed by the strain tensor and its derivatives. We will now rewrite these equations with the stress tensor as variable by replacing the strain tensor (and its derivatives) by the stress tensor (and its derivatives). This can only be done in an approximate way, since the equations are nonlinear. We will set up a power series expansion up to second order in the (old and new) variables. Of course, the resulting equations are less general than the starting ones and only applicable, if quadratic nonlinearities are sufficient for the problem at hand. It is then also sufficient to restrict the phenomenological expansions (4,5,7) to quadratic order. In addition, we will neglect in this section also those phenomenological terms that are connected with the traces of the variables (U_{kk}, Ψ_{kk}, $X_{kk}^{(ph)}$), i.e. there are only six static and dynamic material parameters left ($\alpha_{1,2}$, $\nu_{1,2}$, and $K_{1,2}$). A more general treatment including the trace-related second order phenomenological constants will be given in Appendix B. With this proviso the dynamic strain equation reads

$$\frac{d}{dt}U_{ij} - A_{ij} + U_{ki}\nabla_j v_k + U_{kj}\nabla_i v_k = -\frac{1}{\tau_1}U_{ij} - \frac{1}{\tau_2}U_{ik}U_{jk} \tag{9}$$

with the abbreviations $\tau_1^{-1} = \alpha_1 K_1$ and $\tau_2^{-1} = 2\alpha_1 K_2 + 2\alpha_2 K_1$. The stress tensor that enters the Navier-Stokes equation (2) is written as

$$\sigma_{ij} = -K_1 U_{ij} + K_2' U_{ik}U_{jk} - \nu_1 A_{ij} - \nu_2(U_{ik}A_{jk} + U_{jk}A_{ik}) \tag{10}$$

with $K_2' = 2K_1 - 2K_2$. Linearizing in the phenomenological expansions (4,5,7), i.e. $K_2 = \nu_2 = \tau_2^{-1} = 0$, does not linearize the stress tensor, because of the convective nonlinearities in the strain relaxation equation (9). Only the condition $K_2' = 0$ (and $\nu_2 = 0$) linearizes the stress tensor, which is however a rather artificial condition, since it requires the linear and quadratic elastic moduli to be equal exactly.

Taking the derivative d/dt of σ_{ij} in (10) and replacing dU_{ij}/dt according to Eq.(9) we get

$$\frac{d}{dt}\sigma_{ij} = -f[U_{ij}, A_{ij}, \frac{d}{dt}A_{ij}, \Omega_{ij}] \tag{11}$$

in terms of the strain tensor, the deformational flow and its derivative, and the vorticity $\Omega_{ij} = \frac{1}{2}(\nabla_j v_i - \nabla_i v_j)$ describing rotational flow. The latter enters due to the convective nonlinearities in (9). To convert this into the desired dynamic equation for the stress tensor we have to invert $\sigma_{ij} = \sigma_{ij}[U_{ij}, A_{ij}]$, Eq.(10), into $U_{ij} = U_{ij}[\sigma_{ij}, A_{ij}]$. This is done approximately by the power expansion $U_{ij} = U_{ij}^{(lin)} + U_{ij}^{(quad)} + \ldots$, where $U_{ij}^{(lin)}$ and $U_{ij}^{(quad)}$ contain expressions linear and quadratic in the variables, respectively. In particular we find

$$K_1 U_{ij}^{(lin)} = -\sigma_{ij} - \nu_1 A_{ij} \tag{12}$$

$$K_1^3 U_{ij}^{(quad)} = K_2'\sigma_{ik}\sigma_{jk} + (K_2'\nu_1 + K_1\nu_2)(\sigma_{ik}A_{jk} + \sigma_{jk}A_{ik}) + \nu_1(K_2'\nu_1 + 2K_1\nu_2)A_{ik}A_{jk} \tag{13}$$

Using these expressions the dynamic equation for the stress tensor takes the final form

$$\begin{aligned} \tau_1 \frac{D_a}{Dt}\sigma_{ij} + \sigma_{ij} &= -\nu_\infty A_{ij} - \nu_1\tau_1 \frac{D_b}{Dt}A_{ij} + \frac{r}{K_1}\sigma_{ik}\sigma_{jk} \\ &\quad + \frac{\tau_1\nu_2}{K_1}\left([\sigma_{jk} + \nu_1 A_{jk}]\frac{\partial}{\partial t}A_{ik} + [\sigma_{ik} + \nu_1 A_{ik}]\frac{\partial}{\partial t}A_{jk}\right) + O(3) \end{aligned} \tag{14}$$

where $\nu_\infty = \nu_1 + \tau_1 K_1$ and $r = \tau_1/\tau_2 - K_2'/K_1$ and

$$\frac{D_a}{Dt}T_{ij} \equiv \frac{d}{dt}T_{ij} - a(T_{ik}A_{jk} + T_{jk}A_{ik}) - (T_{ik}\Omega_{jk} + T_{jk}\Omega_{ik}) \tag{15}$$

for any tensor T_{ij} and number a. For $a = -1$ ($a = +1$) D_a/Dt is the lower (upper) convected derivative, for $a = 0$ the Jaumann or corotational derivative, while for a general a a linear combination of those is invoked. In our case the numbers a and b are

$$a = -1 + \frac{\nu_1}{K_1\tau_2} - \frac{K_2'}{K_1^2}\frac{\nu_\infty}{\tau_1} \tag{16}$$

$$b = -1 + \frac{\nu_1}{2K_1\tau_2} - \frac{K_2'}{2K_1^2}\frac{\nu_\infty}{\tau_1} - \frac{K_2'}{2K_1} - \frac{\nu_2}{\nu_1} \tag{17}$$

In the very special 'linear' case discussed above ($K_2' = \tau_2^{-1} = \nu_2 = 0$) we get $a = b = -1$ and both time derivatives in Eq.(14) are of the lower convected type (as was the case for U_{ij} without any approximation). However, this case is rather artificial and, generally, the phenomenological terms push these time derivatives away from the lower convected type and make them material dependent.

Splitting Eq.(14) into the trace and the traceless part, it is easy to see that σ_{kk} and $(d/dt)\sigma_{kk}$ are of O(2), since $A_{kk} = 0$, and do not influence the dynamics of the traceless part σ_{ij}^0 up to second order. This statement remains true, even if the trace-related second order phenomenological constants are taken into account (Appendix B).

IV. COMPARISON WITH CONSTITUTIVE MODELS

Eq.(14) is in a form that many empirical constitutive models have and a direct comparison is possible. First there are the models of the form

$$\tau_1 \frac{D_a}{Dt}\sigma_{ij} + \sigma_{ij} = -\nu_\infty A_{ij} \tag{18}$$

that lack the time derivative of A_{ij} and the terms $\sim r$ and $\sim \nu_2$. For $a = -1, 0, 1$ these are the Maxwell models [20], while for $-1 < a < 1$ it is the Johnson-Segalman model [29]. Apparently in these models the Newtonian viscosity ν_1 has been neglected. For the actual viscosity ν_∞ that means it is given by $\tau_1 K_1$ alone. This seems to be a good approximation for polymer solutions, where ν_1 is interpreted as the viscosity of the solvent and $\tau_1 K_1$ as due to the polymers. Of course, if $\nu_1 = 0$, it only makes sense to also put $\nu_2 = 0$. In addition, $r = 0$ generally implies

$\tau_2^{-1} = 0 = K_2'$ (the relation $\tau_1/\tau_2 = K_2'/K_1$ that also leads to $r = 0$ is rather singular) meaning that these models are all within the linear case described above. However, this in turn has the consequence that $a = -1$, which makes the Johnson-Segalman model inconsistent. The Maxwell model with $a = 1$ is applicable in a Lagrange description, while $a = 0$ is not appropriate for a stress tensor like quantity (but rather for an orientational order parameter [30]).

The next group of models take into account the time derivative of A_{ij} [16]

$$\tau_1 \frac{D_a}{Dt}\sigma_{ij} + \sigma_{ij} \;=\; -\nu_\infty A_{ij} - \nu_\infty \lambda_r \frac{D_a}{Dt} A_{ij} \tag{19}$$

but neglect the terms $\sim r$ and $\sim \nu_2$. Thus, they again are in the linear class, a fact which is compatible with $a = b$ that is inherent to these models. Again $a = -1$ (Oldroyd A) is required for the Eulerian case (and $a = 1$, or Oldroyd B, for the Lagrangian description) while $a = 0$ (Jeffries) is inappropriate.

The first model that goes beyond the linear case is the Giesekus model [19]

$$\tau_1 \frac{D_1}{Dt}\sigma_{ij} + \sigma_{ij} \;=\; -\nu_\infty A_{ij} - \frac{2c\tau_1}{\nu_\infty}\sigma_{ik}\sigma_{jk} \tag{20}$$

with the phenomenological number c with $0 < c < 1$. This model neglects ν_1 and consistently also ν_2. Since c is positive, comparison with (14) shows that $r < 0$ or $K_2'/K_1 > \tau_1/\tau_2$ is assumed. However, with $K_2' \neq 0$ one immediately concludes from (16) that a has to deviate from the pure value -1 (or 1). Thus, a model with nonlinearities in the stress tensor like the Giesekus model must not be of the pure lower (or upper) convected type, but has to show a material dependent deviation from that. For $\nu_1 = 0 = \nu_2$ this deviation from $a = -1$ is $-2c/K_1$.

All the models above are restricted to nonlinearities quadratic in the variables and do not show any term we have hidden in $O(3)$ in Eq.(14). This is different in the PTT and PT models [31, 32] that have a stress tensor nonlinearity of the form $c\sigma_{ij}\sigma_{kk} + O(4)$, which for $A_{kk} = 0$ is of third order. These models have consistently a stress convection with a different from -1. However, there are several other 3rd order terms (cf. Eq.(B.7)) and even 2nd order ones, not considered in these models.

Another popular rheological model is the 'second order fluid' [20] that contains a contribution

$$\sigma_{ij}^{(2)} = -c_1 \frac{D_{\pm 1}}{Dt} A_{ij} - c_2 A_{ik} A_{jk} \tag{21}$$

in the stress tensor ($\pm$ for the Lagrangian and Eulerian picture, respectively). Comparison with Eq.(10) reveals that it is contained there with $c_1 = \nu_1\tau_1$ and $c_2 = \nu_1\tau_1(b+1)$ (for the Eulerian case), where b is defined in Eq.(17). In the linear case defined above $b = -1$ and therefore $c_2 = 0$. Thus, the quadratic flow contributions to the stress tensor are due to the strain dependence of elasticity, viscosity and strain relaxation, when the strain field is taken into account as a relaxing variable and then eliminated.

Even when no convective derivatives are considered, very often nonlinear phenomenological stress/strain relations $\underline{\sigma}^{(ph)} = F(\underline{A})$ are used. As an example, in a power law fluid the Newtonian shear viscosity ν_1 is replaced by $\nu = \sum_{n=1}^{N} \nu_n (A_{ik}A_{ik})^{(n-1)/2}$. Much more complicated forms are used [33]. The problem with all these models is the compliance with thermodynamics. The expansion (6) for the viscosity tensor in terms of U_{ij} avoids this problem and can be carried on to any order desired. However, similar contributions to the stress tensor originate from the expansions of α_{ijkl} and K_{ijkl}. In addition, these expansions also change quite considerably the structure of the dynamic equation for the stress tensor (14) rendering inconsistent any model that uses a power law description of the shear viscosity, only.

V. SUMMARY

In this manuscript we have shown that the hydrodynamically derived model for non-Newtonian fluids in terms of the Eulerian strain tensor contains most of the standard rheological models as special cases and discards a few of them. Of course, the former is more general, as it contains powers of the relevant fields of arbitrary order when written in terms of the stress tensor. The hydrodynamic method allows to discriminate those parts of the dynamics that are due to general principles from the unavoidable phenomenological part. The latter is given here in the form of truncated power series in the strain tensor that can systematically be generalized when necessary. For the phenomenological part we stick to the well-established 'linear irreversible thermodynamics', which, being linear in the generalized forces, nevertheless leads to equations highly nonlinear in the variables like the strain tensor.

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APPENDIX A: Generalization of the phenomenological equations

In Eqs.(4,5) we have omitted possible reversible crosscouplings between flow and strain dynamics

$$X_{ij}^{(ph)} = -\alpha_{ijkl}\Psi_{kl} - \beta_{ijkl}A_{kl} \tag{A.1}$$

$$\sigma_{ij}^{(ph)} = -\nu_{ijkl}A_{kl} + \beta_{klij}\Psi_{kl} \tag{A.2}$$

characterized by the tensor β_{ijkl}, which is of a slightly more complicated form than α_{ijkl} in Eq.(6). Being reversible it is not derived from a potential and lacks the $ij-kl$ symmetry. This results in two different components $\beta_{4a}\delta_{ij}U_{kl} + \beta_{4b}\delta_{kl}U_{ij}$, where, however, the latter (as well as $\beta_{3,5}$) vanish due to incompressibility. Thus we are left with 4 parameters $\beta_{1,2,4a,6}$. Such terms, coming with probably small reactive transport parameters, have to compete with the parameter free, symmetry required terms already being part of Eqs.(1,2). These reversible crosscouplings are possible in the viscoelastic case only, and are absent for permanent elasticity. In the latter case only diffusion $X_{ij}^{(ph)} = D\nabla_k(\nabla_i\Psi_{jk} + \nabla_j\Psi_{ik})$ is present.

We also list here the 5 third order static elastic contributions that follow from an elastic energy quartic in the strain tensor

$$\begin{aligned} K_{ijkl}^{(3)} = {} & K_6\delta_{ij}\delta_{kl}U_{pp}U_{qq} + K_7([\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}]U_{pp}U_{qq} + \delta_{ij}\delta_{kl}U_{pq}U_{pq}) \\ & + K_8(\delta_{ij}U_{kp}U_{lp} + \delta_{kl}U_{ip}U_{jp} + \tfrac{1}{4}[\delta_{ik}U_{jl} + \delta_{jk}U_{il} + \delta_{il}U_{jk} + \delta_{jl}U_{ik}]U_{pp}) \\ & + K_9(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})U_{pq}U_{pq} + K_{10}(\delta_{ik}U_{jp}U_{lp} + \delta_{jk}U_{ip}U_{lp} + \delta_{il}U_{jp}U_{kp} + \delta_{jl}U_{ip}U_{kp}) \end{aligned} \tag{A.3}$$

and that are a generalization of the first and second order terms kept in Sec.II. A necessary stability condition involving the second order modulus K_2 requires $27K_1(K_9 + K_{10}) > 2K_2^2$.

APPENDIX B: Influence of the tensor traces

Here we discuss the influence of those phenomenological parameters in Eqs.(6,7) that are connected with the traces of the tensors involved, $\alpha_{3,4,5,6}$, ν_6, and $K_{3,4,5}$, as well as $\beta_{1,2,4a,6}$ (A.1,A.2) neglected in the main text. The phenomenological parts of the dynamic and static equations then read

$$\begin{aligned} X_{ij}^{(ph)} = {} & -\alpha_1\Psi_{ij} - \alpha_2(U_{ik}\Psi_{jk} + U_{jk}\Psi_{ik}) - \alpha_3\delta_{ij}\Psi_{kk} - \alpha_4(\delta_{ij}U_{kl}\Psi_{kl} + U_{ij}\Psi_{kk}) - \alpha_5\delta_{ij}U_{kk}\Psi_{ll} \\ & -2\alpha_6U_{kk}\Psi_{ij} - \beta_1A_{ij} - \beta_2(U_{ik}A_{jk} + U_{jk}A_{ik}) - \beta_{4a}\delta_{ij}U_{kl}A_{kl} - 2\beta_6U_{kk}A_{ij} \end{aligned} \tag{B.1}$$

$$\begin{aligned} \sigma_{ij}^{(ph)} = {} & -\nu_1A_{ij} - \nu_2(U_{ik}A_{jk} + U_{jk}A_{ik}) - 2\nu_6A_{ij}U_{kk} + \beta_1\Psi_{ij} + \beta_2(\Psi_{ik}U_{jk} + \Psi_{jk}U_{ik}) \\ & + \beta_{4a}\Psi_{kk}U_{ij} + 2\beta_6\Psi_{ij}U_{ll} \end{aligned} \tag{B.2}$$

$$\Psi_{ij} = K_1U_{ij} + 2K_2U_{ik}U_{jk} + K_3\delta_{ij}U_{kk} + K_4(\delta_{ij}U_{kl}U_{kl} + 2U_{ij}U_{kk}) + K_5\delta_{ij}U_{kk}U_{ll} \tag{B.3}$$

The strain relaxation and the stress take the form

$$\begin{aligned} \frac{D_{-1}}{Dt}U_{ij} - A_{ij} = {} & -\tau_1^{-1}U_{ij} - \tau_2^{-1}U_{ik}U_{jk} - \tau_3^{-1}\delta_{ij}U_{kk} - \tau_4^{-1}\delta_{ij}U_{kl}U_{kl} - \tau_5^{-1}\delta_{ij}U_{kk}U_{ll} \\ & -\tau_6^{-1}U_{ij}U_{kk} - \beta_1A_{ij} - \beta_2(U_{ik}A_{jk} + U_{jk}A_{ik}) - \beta_{4a}\delta_{ij}U_{kl}A_{kl} - 2\beta_6U_{kk}A_{ij} \end{aligned} \tag{B.4}$$

$$\begin{aligned} \sigma_{ij} = {} & -\tilde{K}_1U_{ij} + \tilde{K}_2'U_{ik}U_{jk} - \tilde{K}_3\delta_{ij}U_{kk} + \tilde{K}_3'U_{ij}U_{kk} - \tilde{K}_4\delta_{ij}U_{kl}U_{kl} \\ & -\tilde{K}_5'\delta_{ij}U_{kk}U_{ll} - \nu_1A_{ij} - \nu_2(U_{ik}A_{jk} + U_{jk}A_{ik}) - 2\nu_6A_{ij}U_{kk} \end{aligned} \tag{B.5}$$

with the new parameters $\tau_3^{-1} = \alpha_1K_3 + \alpha_3K_1 + 3\alpha_3K_3$, $\tau_4^{-1} = \alpha_1K_4 + 2\alpha_3K_2 + 3\alpha_3K_4 + \alpha_4K_1$, $\tau_5^{-1} = 2\alpha_3K_4 + \alpha_4K_3 + \alpha_5K_1 + \alpha_1K_5 + 3\alpha_3K_5 + 3\alpha_5K_3 + 2\alpha_6K_3$, $\tau_6^{-1} = 2\alpha_2K_3 + \alpha_4K_1 + 2\alpha_1K_4 + 3\alpha_4K_3 + 2\alpha_6K_1$, $\tilde{K}_1 = K_1(1-\beta_1)$, $\tilde{K}_3 = K_3(1-\beta_1)$, $\tilde{K}_4 = K_4(1-\beta_1)$, $\tilde{K}_2' = K_2' + 2\beta_2K_1 + 2\beta_1K_2$, $\tilde{K}_3' = 2\tilde{K}_3(1 + \beta_2 + \frac{3}{2}\beta_{4a}) - 2\tilde{K}_4 + K_1(\beta_{4a} + 2\beta_6)$, and $\tilde{K}_5' = K_5(1-\beta_1) - 2K_3\beta_6$.

The procedure to switch from the strain relaxation equation to an effective dynamic equation for the stress tensor is the same as described in Sec. III. Due to the many new terms, it is more involved, and it is complicated additionally by the fact that the traces and the traceless parts of σ_{ij} and U_{ij} are related by different parameters (even in the linear case) as can be seen from the modified Eq.(12)

$$\tilde{K}_1 U_{ij}^{(lin)} = -\sigma_{ij} - \nu_1 A_{ij} + \frac{\tilde{K}_3}{3\tilde{K}_3 + \tilde{K}_1}\delta_{ij}\sigma_{kk} \tag{B.6}$$

We refrain from writing down more details, but discuss the structure of the final result

$$\begin{aligned} \tau_1 \frac{D_{\tilde{a}}}{Dt}\sigma_{ij} \; &+ \; \sigma_{ij} + B_1 \delta_{ij}\sigma_{kk} = -\tilde{\nu}_\infty A_{ij} - \nu_1 \tau_1 \frac{D_{\tilde{b}}}{Dt} A_{ij} + \frac{\tilde{r}}{\tilde{K}_1}\sigma_{ik}\sigma_{jk} + B_2 \sigma_{kk}\frac{\partial}{\partial t}A_{ij} \\ &+ \; \frac{\tau_1 \nu_2}{\tilde{K}_1}\left([\sigma_{jk} + B_3 \delta_{jk}\sigma_{pp} + \nu_1 A_{jk}]\frac{\partial}{\partial t}A_{ik} + [\sigma_{ik} + B_3\delta_{ik}\sigma_{qq} + \nu_1 A_{ik}]\frac{\partial}{\partial t}A_{jk}\right) \\ &+ \; \sigma_{kk}(B_4\sigma_{ij} + B_5 A_{ij}) + \delta_{ij}(B_6 \sigma_{kl}\sigma_{kl} + B_7 \sigma_{kl}A_{kl} + B_8 A_{kl}A_{kl} + B_9 \sigma_{kk}\sigma_{ll}) + O(3) \end{aligned} \tag{B.7}$$

with $\tilde{\nu}_\infty = \nu_1 + K_1\tau_1(1-\beta_1)^2$ and the tilted numbers $\tilde{a}$, $\tilde{b}$, and $\tilde{r}$ being much more complicated than the untilted ones (16,17). There are structurally new terms related to σ_{kk}, a linear one (with $B_1 = \tilde{K}_3/(3\tilde{K}_3 + \tilde{K}_1) + \tau_1/\tau_3$), and eight quadratic ones characterized by coefficients $B_{2,\ldots,9}$. Nevertheless, the trace of the stress tensor and its time derivative are of second order and do not influence the dynamics of the traceless part in $O(2)$. Thus, for the deviator, $\sigma_{ij}^0 = \sigma_{ij} - (1/3)\delta_{ij}\sigma_{kk}$, the dynamic equation has exactly the form (14), but with the tilted numbers and parameters instead of the untilted ones. For the trace we find to second order

$$\begin{aligned} \tau_1 \frac{d}{dt}\sigma_{kk} + (1+3B_1)\sigma_{kk} \;&=\; 2\tilde{b}\nu_1\tau_1 A_{kl}A_{kl} + \frac{\tilde{r}}{\tilde{K}_1}\sigma_{kl}^0\sigma_{kl}^0 + \frac{2\tau_1\nu_2}{\tilde{K}_1}\left(\sigma_{kl}^0 + \nu_1 A_{kl}\right)\frac{\partial}{\partial t}A_{kl} \\ &+\; 3(B_6\sigma_{kl}^0\sigma_{kl}^0 + [B_7 + \frac{2}{3}\tau_1\tilde{a}]\,\sigma_{kl}^0 A_{kl} + B_8 A_{kl}A_{kl}) + O(3) \end{aligned} \tag{B.8}$$

indicating that its relaxational dynamics is completely given by the flow A_{ij} and the traceless part σ_{ij}^0.

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