

# Reversible and dissipative macroscopic contributions to the stress tensor: Active or passive?

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**Abstract.** The issue of dynamic contributions to the macroscopic stress tensor has been of high interest in the field of bio-inspired active systems over the last few years. Of particular interest is a direct coupling (“active term”) of the stress tensor with the order parameter, the latter describing orientational order induced by active processes. Here we analyze more generally possible reversible and irreversible dynamic contributions to the stress tensor for various passive and active macroscopic systems. This includes systems with tetrahedral/octupolar order, polar and non-polar (chiral) nematic and smectic liquid crystals, as well as active fluids with a dynamic preferred (polar or non-polar) direction. We show that it cannot *a priori* be seen, neither from the symmetry properties of the macroscopic variables involved, nor from the structure of the cross-coupling contributions to the stress tensor, whether the system studied is active or passive. Rather, that depends on whether the variables that give rise to those cross-couplings in the stress tensor are driven or not. We demonstrate that several simplified descriptions of active systems in the literature that neglect the necessary counter term to the active term violate linear irreversible thermodynamics and lead to an unphysical contribution to the entropy production.

## 1 Introduction

The study of the collective behavior in active media covers a large range of length and time scales. Systems of interest include, listed with increasing characteristic length scale: biological motors [1–5], pattern formation in bacterial growth phenomena [6–15] as well as schools of fish and flocks of birds [16–19]. We will use the term active to characterize systems for which the unit cells (motors, bacteria, fish, locusts and birds) are driven out of equilibrium internally or individually, typically by chemical reactions. This behavior must be contrasted to systems which are driven out of equilibrium by externally applied fields such as a temperature gradient in thermal convection or the pump field in a laser. However, for the purpose of this paper, there is no fundamental difference between driven systems and active ones, since what matters here is that both are driven systems in contrast to passive systems.

Throughout the present study we focus on the structure of the macroscopic stress tensor and its reversible dynamic as well as dissipative dynamic coupling terms to other collective and macroscopic variables. The stress ten-

sor is often the quantity where active processes show up, in particular, the “active term” as it is called in the literature of active systems. We will show that this term and other couplings to the stress tensor exist in driven/active systems as well as in passive systems.

In a simple fluid the macroscopic stress tensor has three contributions: the isotropic enthalpy,  $(p + f)\delta_{ij}$ , involving the pressure,  $p$ , and the energy density,  $f$ , the momentum density transport,  $\rho v_i v_j$ , with  $\rho$  the density and  $v_i$  the velocity, and the viscous stress [20–22]. The former two are reversible and have no phenomenological parameters associated with it, while the viscous part of the stress tensor in a simple fluid is proportional to the symmetrized velocity gradients,  $\sim A_{ij} = \frac{1}{2}(\nabla_i v_j + \nabla_j v_i)$ , and is characterized by two material parameters, shear and bulk viscosity.

Here we want to study reversible and irreversible contributions to the stress tensor as the symmetry is reduced compared to that of a simple fluid. In particular we study the coupling terms of the stress tensor to macroscopic variables associated with broken continuous symmetries [21–23].

For reversible contributions to the stress tensor the existence of one (or more) preferred direction(s) emerges as a necessary condition. Complex fluids and condensed

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systems of that nature include (passive) nematic, smectic and columnar liquid crystals as well as gels and conventional solids of sufficiently low symmetry. Systems that are driven by external fields (*e.g.* electric, magnetic, temperature and concentration gradients) also show preferred directions. Active systems often have a preferred (polar or non-polar) direction induced by the active processes. All of them show stress tensor couplings with scalar variables of the kind that is called “active term” in the literature.

To obtain reversible contributions to the stress tensor that couple to gradient fields (*e.g.* of temperature, concentration, or electric potential) in the framework of linear irreversible thermodynamics, broken parity is identified as an additional key ingredient. This includes chiral systems and those with tetrahedral (octupolar) order. Another type of vector coupling to the stress tensor is provided by relative rotations in anisotropic gels and elastomers.

Symmetric second-rank order parameter tensors also couple reversibly to the stress tensor and include the appropriate couplings of the scalar variables that denote the degree of order. There are somewhat more exotic stress tensor couplings to non-symmetric tensor fields (in superfluid  $^3\text{He-A}$  and  $^3\text{He-A}_1$ ) and a kind of self-coupling between stress and flow (in axial systems lacking time reversal symmetry) resembling the form of Newtonian viscosity, but being reversible. Here we also mention the standard cross-coupling to elastic stresses and its counterpart in the dynamic displacement equation, which are (both) fixed by the broken translational symmetry and do not carry a phenomenological material parameter.

As necessary condition for the occurrence of additional dissipative contributions we identify the existence of (at least) one preferred direction, which is odd under time reversal. This includes superfluid  $^3\text{He-A}$  and  $^3\text{He-A}_1$ , as well as uniaxial magnetic gels, where in both cases the breaking of time reversal invariance is intrinsic. Systems that are driven by an external magnetic field can also show such dissipative contributions. Active systems, with a polar or axial dynamic preferred direction (due to the motion of active entities), also belong to this class of broken time reversal symmetry. Active systems that are described by a nematic director or by a polarization do not have such dissipative contributions.

The paper is organized as follows. In sect. 2 we first discuss general properties of reversible dynamic contributions to the stress tensor using the well-known flow alignment feature in nematic liquid crystals as an example. In particular, we discuss how the zero-entropy production requirement for reversible processes is implemented and that any entropy production has to be provided by dissipative couplings, in passive as well as in driven/active systems. Then, we give many examples for reversible stress tensor couplings to scalar, vector, and tensor variables. We show that such couplings occur, irrespective of whether the systems are passive or driven/active. In sect. 3 we analyze critically some popular simplified approaches that use dynamic contributions to the stress tensor, but neglect the counter terms necessary within the framework of linear ir-

reversible thermodynamics. We show that this leads to the unphysical situation that reversible effects change the entropy, either increasing or decreasing it, depending on the nature of the active entities involved. In sect. 4 we outline a bridge to systems driven very far from equilibrium. We conclude with a brief summary. Dissipative contributions to the stress tensor are summarized in appendix A.

## 2 Reversible dynamic contributions to the stress tensor

### 2.1 General aspects of reversible dynamic contributions

General symmetry considerations already give some restrictions on the structure of reversible dynamic contributions to the stress tensor  $\sigma_{ij}$ . Since the velocity  $\mathbf{v}$  and the density of linear momentum  $\mathbf{g}$  are odd under parity and time reversal, reversible dynamic contributions to the stress tensor  $\sigma_{ij}$  must be even under parity and even under time reversal, because of the momentum density conservation  $(\partial/\partial t)g_i + \nabla_j\sigma_{ij} = 0$ . Obviously, dissipative contributions to the stress tensor (cf. appendix A) have to be odd under time reversal.

A second important restriction is the condition of zero-entropy production for reversible contributions [21–23]. Thus, the phenomenological reversible currents (super-script  $R$ ) are most easily derived by writing down all symmetry-allowed contributions to the various currents and then make sure that the entropy production is zero. As a well-known example we present this condition for a uniaxial nematic liquid crystal with the director field  $n_i$  and the scalar order parameter (degree of ordering)  $S$

$$R = -j_i^{\sigma,R}\nabla_i T - j_i^{e,R}E_i - \sigma_{ij}^R\nabla_j v_i + Y_i^R h_i + Z^R W = 0. \quad (1)$$

where we have used the notation of ref. [23]. Here  $j_i^{\sigma,R}$  and  $j_i^{e,R}$  are the reversible parts of the heat and electric current density, respectively, while  $Y_i^R$  and  $Z^R$  are the reversible parts of the quasi-currents in the balance equations  $(\partial/\partial t)n_i + Y_i = 0$  and  $(\partial/\partial t)S + Z = 0$  for the director field and the order parameter.  $h_i$  is the molecular field of the director and  $W \equiv (\partial/\partial S)f$  the thermodynamic conjugate to the order parameter with  $f$  the energy density. Reversible currents have the same time reversal behavior as the time derivative of the appropriate variable. They are either even under spatial inversion,  $\sigma_{ij}^R, Z^R$ , or polar vectors,  $j_i^{e,R}, j_i^{\sigma,R}$ . The quasi-current  $Y_i^R$  is transverse to  $\mathbf{n}$  and even under parity. Typically, zero-entropy production is achieved by the cancellation of two mutual cross-coupling terms.

For the stress tensor there is a reversible contribution [21, 22, 24–26] that takes the form

$$\sigma_{ij}^R = -\frac{1}{2}\lambda_{kji}h_k, \quad (2)$$

with the counter term

$$Y_i^R = -\frac{1}{2}\lambda_{ijk}\nabla_j v_k, \quad (3)$$

that ensures zero-entropy production. To guarantee the proper transformation behavior under rigid rotations and to satisfy the  $n_i \rightarrow -n_i$  equivalence of nematic liquid crystals, along with  $n_i^2 = 1$ ,  $\lambda_{ijk}$  has the form

$$\lambda_{ijk} = (\lambda - 1)\delta_{ij}^\perp n_k + (\lambda + 1)\delta_{ik}^\perp n_j, \quad (4)$$

with  $\delta_{ij}^\perp \equiv \delta_{ij} - n_i n_j$ . It contains a phenomenological material parameter  $\lambda$  that determines the flow alignment angle when a uniaxial nematic is subjected to a shear flow [21,25,27]. The contributions with the fixed prefactor “1” reflect the rotational behavior of vectors and tensor. Similarly, the advective part of the time derivatives (not shown in eqs. (2) and (3)), due to Galilean invariance, does not carry a phenomenological parameter.

Reversible flow alignment contributions occur in all systems with broken rotational symmetry, like *e.g.* smectic C [21], biaxial nematic liquid crystals [28,29] and nematic liquid crystalline elastomers [30] as well as superfluid  $^3\text{He-A}$  [31] and superfluid  $^3\text{He-A}_1$  [32]. This coupling also exists for polar nematic liquid crystals [33], with a polarization vector (instead of the director), and for active polar fluids [34], where the preferred direction is a velocity. Naturally, for systems with lower symmetry there are more flow alignment parameters, *e.g.* three for orthorhombic biaxial nematics.

It is obvious that the reversible contributions in eqs. (2) and (3) cannot be derived from a phenomenological potential, since they are nilpotent in the entropy production and not at all related to the free energy. They exist for passive (ordinary) nematics (where  $h_i = 0$  and  $\nabla_j v_i = 0$  in equilibrium), as well as in nematics driven, *e.g.* by an external simple shear flow ( $\nabla_j v_i^{\text{ext}} = \Xi_0 \delta_{ix} \delta_{jy}$ ). In the latter case the stationary non-equilibrium state has a non-zero, constant  $\nabla_y v_x \sim \Xi_0$ . In both cases the entropy production comes from the dissipative parts of the dynamics,  $2R = (1/\gamma_1)h_i h_i + \nu_{ijkl}(\nabla_j v_i)(\nabla_l v_k)$ , where  $\gamma_1$  is the rotational viscosity and  $\nu_{ijkl}$  is the viscosity tensor. In the passive case  $R = 0$  in equilibrium, while in the driven case  $R \sim \Xi_0^2$  is constant in the stationary state, in accordance with basic thermodynamic principles.

This flow alignment example clearly shows that from the structure of the cross-coupling terms between flow and director rotations one cannot *a priori* discriminate passive from active/driven systems. This is, however, not only true for the specific variables used here, but also for other examples involving quite different types of variables, as will be shown below.

## 2.2 Reversible dynamic contributions to the stress tensor coupling to scalar quantities

Close to phase transitions the scalar order parameter, which describes the degree of ordering, is typically a macroscopic variable relaxing on a long but finite time scale [23]. This concept has been introduced by Khalatnikov in the context of the  $\lambda$ -phase transition from normal fluid to superfluid in  $^4\text{He}$  [35] and is also the basis of any time-dependent Ginzburg-Landau description. There

is a reversible dynamic coupling between the scalar order parameter,  $S$ , and flow that reads using the notation of ref. [23]

$$\sigma_{ij}^R = (\beta_{\parallel} n_i n_j + \beta_{\perp} \delta_{ij}^\perp) W \quad (5)$$

and

$$Z^R = (\beta_{\parallel} n_i n_j + \beta_{\perp} \delta_{ij}^\perp) A_{ij} \quad (6)$$

with the thermodynamic conjugate  $W$  and the quasi-current  $Z$  defined after eq. (1), and the symmetrized velocity gradient  $A_{ij}$ . This term was suggested first for the isotropic-superfluid  $^3\text{He-A}$  phase transition [36] and later discussed for several phase transitions involving uniaxial liquid crystalline phases, like the nematic-smectic A [37] and the nematic-columnar [38] phase transitions. In addition this coupling was used to characterize the macroscopic behavior of nematic liquid crystalline elastomers [30], polar nematics [33] as well as active polar fluids [34]. These couplings describe *e.g.* flow-induced order.

Near the  $\lambda$  phase transition in  $^4\text{He}$  the appropriate coupling is isotropic, and can be absorbed into the definition of the pressure term, although the counter term coupling to compressional flow,  $\text{div } \mathbf{v}$ , is still there. On the other hand, it is obvious that for all anisotropic systems the type of coupling of eqs. (5) and (6) exists for any relaxing scalar variables. Typical examples are reaction-diffusion systems [39–41], where one has usually several relaxing scalar variables. In the long wavelength limit the diffusive terms are irrelevant and one is left with  $n$  relaxation equations for the  $n$  concentrations of the reactants. Therefore, in a uniaxially anisotropic fluid environment there are  $2n$  reversible dynamic coupling terms relating flow with concentrations.

This type of coupling can also be found in active systems, *e.g.* active polar gels driven by concentration dynamics using a nematic-like description (eqs. (23) and (26) of ref. [42])

$$\sigma_{\alpha\beta}^R = -(\zeta p_\alpha p_\beta + \bar{\zeta} \delta_{\alpha\beta} + \zeta' p_\gamma p_\gamma \delta_{\alpha\beta}) \Delta\mu \quad (7)$$

and

$$r^R = (\zeta p_\alpha p_\beta + \bar{\zeta} \delta_{\alpha\beta} + \zeta' p_\gamma p_\gamma \delta_{\alpha\beta}) A_{\alpha\beta}, \quad (8)$$

where  $\Delta\mu$  is the chemical potential difference associated with ATP and where  $p_\alpha$  denotes the polarization field. Since the polarization is not normalized to one (in contrast to the director) there are additional independent contributions  $\sim \zeta'$ . Apart from that (and a different sign convention of the stress tensor) eqs. (7) and (8) are completely equivalent to eqs. (5) and (6) discussed above.

Thus, we can again draw the conclusion that the “active term”, eq. (7), occurs in passive systems as well. The discrimination between active or passive systems depends on whether  $W$  (or  $\Delta\mu$ ) is related to passive relaxational or to driven processes. In the latter case the driving can be provided by an external field or by the coupling to other, internally driven variables. The structure of the reversible coupling of the stress tensor to scalar macroscopic variables is the same in all cases.

### 2.3 Reversible dynamic contributions to the stress tensor coupling to vector quantities

There are two types of vectors that can couple reversibly to the stress tensor in certain systems. First, we discuss relative rotations in anisotropic gels/elastomers/polymers and, second, gradients of temperature, concentration, electric potential etc. in systems either with tetrahedral (octupolar) order or with chirality.

Relative rotations as macroscopic variables are characteristic of complex fluids with two subsystems, one of which is typically a gel or an elastomer (or a polymer described by a relaxing elasticity), the other shows a preferred direction. Their use has been pioneered for the static description of nematic elastomers by de Gennes [43]. This concept was generalized into the nonlinear domain and applied also to the dynamics of nematic liquid crystalline elastomers [30], uniaxial magnetic gels [44], and active gels with an axial dynamic preferred direction [45].

The reversible contributions to the stress tensor take the form (here we use the notation introduced for magnetic gels in ref. [44])

$$\sigma_{ij}^R = \xi_{ijk}^R W_k, \quad (9)$$

with the counter term

$$Z_i^R = -\xi_{kli}^R A_{kl}, \quad (10)$$

where  $Z_i$  is the quasi-current, and  $W_k$  is the thermodynamic conjugate, connected to relative rotations. The material tensor is of the form

$$\xi_{kli}^R = \xi^R (m_k \delta_{li}^\perp + m_l \delta_{ki}^\perp), \quad (11)$$

with  $m_i$  denoting the preferred direction. In the case of uniaxial magnetic gels this is the spontaneous magnetization (for driven isotropic magnetic gels it is induced by an external field), for nematic liquid crystal elastomers it is the director, while for axial active gels it is the axial dynamic preferred direction. The latter is a vorticity-like quantity [45] that does not vanish in the stationary state, thus signaling a non-equilibrium situation. These preferred directions have quite different symmetry properties for the different systems, nevertheless the form of eqs. (9)-(11) is the same, since the variables involved also have different symmetry properties, suitable to those of the preferred directions. Again, passive and active systems show the same type of coupling. These couplings describe changes in the relative orientation of preferred direction and the gel matrix due to flow and, vice versa, induced stresses, due to changes in the relative orientation.

The appearance of tetrahedral (or octupolar) order leads to a number of rather specific cross-coupling terms of the reversible part of the stress tensor to gradients of temperature, concentration, or electric scalar potential  $\phi$  (with the electric field  $E_i = -\nabla_i \phi$ ) [46–48]. In the optically isotropic  $T_d$  phase this reads

$$\sigma_{jk}^R = -\Gamma_1 T_{ijk} E_i - \Gamma_2 T_{ijk} \nabla_i T - \Gamma_3 T_{ijk} \nabla_i c, \quad (12)$$

with  $T_{ijk}$ , the tetrahedral order parameter containing four equivalent unit vectors of tetrahedral orientation. This is the key quantity promoting these coupling terms, since it is odd under parity (and completely symmetric in all indices) [49]. The counter terms in the electric, heat, and concentration current are, respectively,

$$j_i^{e,R} = \Gamma_1 T_{ijk} A_{jk}, \quad (13)$$

$$j_i^{\sigma,R} = \Gamma_2 T_{ijk} A_{jk}, \quad (14)$$

$$j_i^{c,R} = \Gamma_3 T_{ijk} A_{jk}. \quad (15)$$

They describe *e.g.* that a shear flow leads to electric, heat and concentration currents perpendicular to the shear plane [46], and, vice versa, gradients of temperature, concentration and electric potential lead to stresses in the plane perpendicular to the gradients.

There are tetrahedral phases of lower symmetry that show an additional nematic order. Depending on the orientation of the director with respect to the tetrahedral axes, they can be of D2d [47], D2 and S4 [48] symmetry. In all these cases there are reversible coupling terms of the same type as eqs. (12)-(15). However, there are twice (D2d and S4) or three times (D2) as many independent  $\Gamma$  coefficients as in the Td phase [47, 48]. Due to the low symmetry of these phases the geometry of induced stresses and currents is rather complicated, *e.g.* it involves hyperbolic flows and stresses, as well as oblique currents.

Similar reversible cross-couplings between flow and gradients of the scalar variables as for tetrahedral phases are present in a number of chiral systems including cholesteric and chiral smectic liquid crystals, and active chiral systems with an axial or polar dynamic preferred direction. Using the notation of [45], where these terms have been presented for the first time, we get

$$j_i^{\sigma R} = q_0 \chi_{ijk}^{\sigma} A_{jk}, \quad (16)$$

$$j_i^{eR} = q_0 \chi_{ijk}^e A_{jk}, \quad (17)$$

$$j_i^{\alpha R} = q_0 \chi_{ijk}^{\alpha} A_{jk}, \quad (18)$$

$$\sigma_{jk}^R = -q_0 (\chi_{ijk}^{\alpha} \nabla_i \mu_{\alpha} + \chi_{ijk}^e E_i + \chi_{ijk}^{\sigma} \nabla_i T), \quad (19)$$

with the third-rank material tensors

$$\chi_{ijk}^{\xi} = \chi^{\xi} (\epsilon_{ikm} w_j w_m + \epsilon_{ijm} w_k w_m). \quad (20)$$

Here,  $w_i$  is the axial dynamic preferred axis (vorticity type), but it works the same way, if it is taken as the polar dynamic preferred axis (velocity type), since there is always an even number of  $w_i$  factors. For the same reason, eqs. (16)-(19) are applicable to cholesteric and chiral tilted smectic liquid crystals, where  $w_i$  stands for the director. Considering thin films of the latter systems, where the helix is (almost) unwound and along the  $z$ -axis, an external shear flow, whose shear plane (the plane spanned by the velocity and the gradient direction) contains the  $z$ -direction, induces (electric, heat and concentration) currents perpendicular to the shear plane. Vice versa, external currents perpendicular to the  $z$ -axis that is, in the  $x$ - $y$  plane, induce stresses in the plane perpendicular to the currents. Again, we notice that these reversible cross-couplings exist for passive as well as active systems.



## 2.4 Reversible dynamic contributions to the stress tensor coupling to tensor quantities

Instead of using the scalar degree of ordering to describe phase transitions (as in sect. 2.2) one can use the full tensor (or vector) order parameter structure. For the uniaxial-biaxial nematic phase transition this has been done by Jacobsen and Swift [50] and further analyzed in ref. [51]. For the isotropic-uniaxial phase transition the analogous analysis has been given in ref. [52]. In the context of the uniaxial-biaxial phase transition one denotes by  $n_i$  the director of the uniaxial phase, while the directors  $l_i$  and  $m_i$ , which are used to complete the orthonormal triad, are used to characterize biaxial order. Using the two-dimensional biaxial order parameter  $\xi_{ij} = b(m_i m_j - l_i l_j)$ , its conjugate  $S_{kl}$ , and the quasi-current  $\Phi_{kl}$ , which are all symmetric and traceless, there is, in the uniaxial phase, a reversible cross-coupling of the stress tensor with biaxial orientational fluctuations of the form

$$\sigma_{ij}^R = \tau_{ijkl} S_{kl}, \quad (21)$$

$$\Phi_{kl}^R = \tau_{ijkl} A_{ij}, \quad (22)$$

where  $\tau_{ijkl}$  contains only one parameter

$$\tau_{ijkl} = \frac{\tau}{2} (\delta_{il}^{\perp} \delta_{jl}^{\perp} + \delta_{il}^{\perp} \delta_{jk}^{\perp} - \delta_{ij}^{\perp} \delta_{lk}^{\perp}), \quad (23)$$

and where  $\Phi_{kl}^R$  is the reversible part of the quasi-current associated with the biaxial order parameter  $\xi_{ij}$ .

In ref. [53] the dynamics of the uniaxial order parameter tensor  $Q_{ij} = (S/2)(3n_i n_j - \delta_{ij})$  has been given and compared to the dynamics of the director  $n_i$  and the ordering strength,  $S$ , on both sides of the isotropic-uniaxial phase transition. In the isotropic phase orientational fluctuations couple reversibly to the stress tensor

$$\sigma_{ij}^R = -2\lambda_1 \psi_{ij}, \quad (24)$$

$$\Phi_{ij}^R = -2\lambda_1 A_{ij}^{\text{tr}}, \quad (25)$$

where  $\psi_{ij} = (\partial f / \partial Q_{ij})$  (with  $f$  a Landau type free energy density [54]) is the thermodynamic conjugate to  $Q_{ij}$  and  $\Phi_{ij}^R$  is the reversible part of the quasi-current associated with the uniaxial order parameter  $Q_{ij}$ .  $A_{ij}^{\text{tr}}$  is the traceless part of  $A_{ij}$ , and  $\lambda_1$  a generalized flow alignment coefficient that describes possible shear flow-induced nematic order in the isotropic phase.

In the nematic phase one obtains in linear order [53]

$$\sigma_{ij}^R = -\frac{3}{2} (c_{ijkl}^{\lambda} + c_{ijkl}^{\beta}) \psi_{kl}, \quad (26)$$

$$\Phi_{ij}^R = -\frac{3}{2} (c_{klij}^{\lambda} + c_{klij}^{\beta}) A_{kl}, \quad (27)$$

with

$$c_{ijkl}^{\lambda} = \lambda S_{\text{eq}}^2 n_k (n_i \delta_{lj}^{\perp} + n_j \delta_{li}^{\perp}), \quad (28)$$

$$c_{ijkl}^{\beta} = (\beta_{\perp} \delta_{ij}^{\perp} + \beta_{\parallel} n_i n_j) \left( n_k n_l - \frac{1}{3} \delta_{kl} \right). \quad (29)$$

In terms of the director and the degree of ordering, eqs. (26) and (27) read

$$\sigma_{ij}^R = -\frac{1}{2} \lambda S_{\text{eq}} (n_i h_j^{\perp} + n_j h_i^{\perp}) - (\beta_{\perp} \delta_{ij}^{\perp} + \beta_{\parallel} n_i n_j) W, \quad (30)$$

$$Y_i^R = -\frac{1}{2} \lambda S_{\text{eq}} (n_k \delta_{ij}^{\perp} + n_j \delta_{ik}^{\perp}) A_{jk}, \quad (31)$$

$$Z^R = -(\beta_{\perp} \delta_{ij}^{\perp} + \beta_{\parallel} n_i n_j) A_{ij}, \quad (32)$$

reflecting both the flow alignment type coupling, eq. (2), and the degree of order coupling, eq. (5) with a slightly different definition of the material parameters  $\beta$  and  $\lambda$ .

The contributions to the stress tensor in eqs. (24) and (30) are of the type of the ‘‘active term’’, where reversible stresses are induced by an order parameter (or its fluctuations). However, in the present context of a thermodynamic isotropic-uniaxial phase transition, these terms describe passive, equilibrium processes. They only become active or non-equilibrium, if the system (the phase transition) is driven. This can happen by applying an external field, *e.g.* an externally imposed shear flow, which results in a (flow-induced) nematic order even above the thermodynamic phase transition [27]. In that case the stationary state shows  $\psi_{ij} \neq 0$  and a finite stress is obtained. Another way of making these terms active would be the coupling of the degree of ordering to a driven reaction-diffusion system, leading to finite stresses, because of  $W \neq 0$  (cf. eq. (30)). The latter case might be difficult to realize experimentally. The message, however, is that, from the form of eq. (24) or (26), one cannot decide whether the processes involved are passive or active.

It must be mentioned that the dynamics of the isotropic-nematic phase transition using the full order parameter  $Q_{ij}$  has been studied first by de Gennes [54] (compare also the discussion in his monograph [27]). However, in ref. [54] the coupling between the order parameter and the stress tensor was considered to be a dissipative one, since the time derivative  $(\partial/\partial t)Q_{ij}$  was used as a force. In refs. [27, 54] an entropy source was introduced via  $T\dot{S} = \dot{Q}_{\alpha\beta} \Phi_{\alpha\beta} + \sigma'_{\alpha\beta} A_{\alpha\beta}$  with the viscous stress  $\sigma'_{\alpha\beta}$  and with  $\Phi_{\alpha\beta} = -\frac{\partial F}{\partial Q_{\alpha\beta}}$ , where  $F$  is the Landau energy associated with the isotropic-nematic phase transition [54]. Correspondingly one obtains for the viscous stress  $\sigma'_{\alpha\beta} = 2\eta A_{\alpha\beta} + 2\mu \dot{Q}_{\alpha\beta}$  with viscous coefficients  $\eta$  and  $\mu$  [27, 54].

There is also an example for a reversible cross-coupling of the stress tensor involving non-symmetric tensors. In uniaxial superfluids, *e.g.* superfluid  $^3\text{He-A}$  and  $-A_1$ , the spatial angular momentum is not conserved due to a (small) spin-orbit coupling allowing for non-symmetric stresses [32, 55]

$$\sigma_{ij}^R = -\lambda l_k \epsilon_{kij} \text{div } \boldsymbol{\lambda}^{(s)} \quad (33)$$

and

$$I_{\varphi} = \lambda l_k \epsilon_{kij} \nabla_i v_j, \quad (34)$$

with  $\lambda_i^{(s)}$  the conjugate to the superfluid velocity  $v_i^{(s)} \sim \nabla_i \varphi$ , and  $I_{\varphi}$  the quasi-current of the superfluid phase

$((\partial/\partial t)\varphi + I_\varphi = 0)$ . The preferred direction, usually called  $l$ , is odd under time reversal. For isotropic superfluids the latter does not exist and no such coupling is possible.

For active axial systems with a dynamic preferred direction,  $w_i$ , that is odd under time reversal but even under spatial inversion there is a reversible self-coupling of flow and stress of the form [45]

$$\sigma_{ij}^R = -\nu_{ijkl}^R A_{kl}, \quad (35)$$

with

$$\begin{aligned} \nu_{ijkl}^R &= \nu_1^R w_p (\epsilon_{ikp} \delta_{jl}^\perp + \epsilon_{jlp} \delta_{ik}^\perp + \epsilon_{ilp} \delta_{jk}^\perp + \epsilon_{jkp} \delta_{il}^\perp) \\ &+ \nu_2^R w_p (\epsilon_{ikp} w_j w_l + \epsilon_{jlp} w_i w_k \\ &+ \epsilon_{ilp} w_j w_k + \epsilon_{jkp} w_i w_l), \end{aligned} \quad (36)$$

that does not need any counter term, since the entropy production  $\sim \nu_{ijkl}^R A_{ij} A_{kl}$  vanishes identically. This type of contribution to the stress tensor was pioneered by M. Liu [55] for superfluid  $^3\text{He-A}$  and also exists for superfluid  $^3\text{He-A}_1$  [32], both with  $l_i$  instead of  $w_i$ . It also is present in uniaxial magnetic systems [44] (including gels and elastomers) with the magnetization as the preferred direction, and in chiral active polar ones, when  $w_i$  is replaced by  $q_0 f_i$  [34] with  $q_0$  the chiral pseudoscalar and  $f_i$  the relative velocity.

Reversible contributions to the stress tensor that are not related to phenomenological material-dependent processes, but are dictated by symmetry, occur in systems with spontaneously broken translational symmetry [21]. Therefore, they do not have a phenomenological coefficient. Denoting the displacement vector by  $u_i$ , its thermodynamic conjugate force by  $\phi_{ij}$  and its quasi-current by  $X_i$  we find, *e.g.* for biaxial discotic columnar phases [28]

$$\sigma_{ij}^R = -(n_i n_k + m_i m_k) \phi_{kj} \quad (37)$$

and

$$X_i^R = -(n_i n_j + m_i m_j) v_j. \quad (38)$$

These contributions reflect the 2-dimensional broken translational symmetry along the directions  $n_i$  and  $m_i$ . They describe the correct Galilei transformation, requiring, in the absence of any dissipative process, the time derivative of a homogeneous displacement vector to be the velocity of the system. Corresponding expressions exist for layered fluid smectic phases and crystalline phases [21]. In the latter case, the  $\phi_{ij}$  in eq. (37) is the elastic stress that follows from the energy by derivation with respect to the strain tensor. Nevertheless, the cross-couplings, eqs. (37) and (38), cannot be derived from a potential, but are required to give zero-entropy production. These couplings are responsible for transverse sound-like propagating excitations in such systems.

### 3 Problems with common simplified truncated approaches

In the previous section we have discussed selected examples of reversible coupling terms between the stress

tensor and other variables. We have seen that for every such cross-coupling term there is a counter term  $\sim A_{ij}$  in the current or quasi-current associated with the thermodynamic force the stress tensor is coupling to. This is simply a consequence of linear irreversible thermodynamics [21–23, 56].

It has become popular recently to describe active hydrodynamic systems by a simplified procedure: Taking the passive hydrodynamic equations (*e.g.* nematic or polar nematic) an “active term” is added to the stress tensor that drives the system into non-equilibrium. However, these are typically reversible terms of the kind described in the preceding section that require counter terms. Often, however, these counter terms are neglected, thus violating thermodynamic principles. In addition, we will show that this also leads to unphysical results.

In eq. (10) of ref. [57] there is an “active term”, a contribution to the stress tensor of the form

$$\sigma_{ij}^{\text{active}} = -\zeta Q_{ij}, \quad (39)$$

where  $Q_{ij}$  is the nematic order parameter tensor and  $\zeta > 0$  ( $\zeta < 0$ ) for extensile (contractile) active rods. Given the behavior under time reversal this contribution must be reversible and not dissipative. Therefore one would expect a counter term in the dynamic equation for  $Q_{ij}$ . Inspecting, however, eqs. (5) and (6) of ref. [57] we notice that there is no such counter term, thus violating linear irreversible thermodynamics.

In addition, this term (without its counter term) leads to an entropy production, eq. (1), of the form  $R = \zeta Q_{ij} A_{ij}$  that does not have a definite sign. As a result, entropy might increase for extensile rods and therefore decrease for contractile ones (or vice versa), which does not seem to be physically reasonable. Using the full set of hydrodynamic equations [42] for active nematics, it is clear that the dissipative driving into non-equilibrium does not come from the “active term”, but from the coupling to a chemical energy-consuming subsystem of molecular motors or equivalent biological entities, typically described by driven reaction-diffusion equations.

A very similar reasoning applies to some truncated descriptions of active polar nematic systems. Considering eq. (9a) and eq. (4) of refs. [58] and [59], respectively, one finds active stresses

$$\sigma_{ij}^\alpha = \alpha c \left( P_i P_j + \frac{1}{2} P^2 \delta_{ij} \right) \quad (40)$$

and

$$\sigma_{ij}^a = W c \left( p_i p_j - \frac{1}{2} p^2 \delta_{ij} \right), \quad (41)$$

respectively, where  $c$  is the concentration of active particles and  $P_i$  (or  $p_i$ ) is the polar nematic vector (the two expressions differ by the definition of the pressure); the prefactors ( $\alpha$  and  $W$ ) can have both signs depending on the type of active entities considered.

Again, these active contributions are reversible and must not produce entropy. The necessary counter terms cannot be found, neither in eqs. (2) to (4) of ref. [58], nor

in eqs. (1) or (2) of ref. [59]. Thus, we arrive at the same conclusions as above, namely that these simplified truncated approaches violate linear irreversible thermodynamics and lead to unphysical entropy production expressions that lack a definite sign. A hydrodynamic theory of active polar nematics, using the velocity of the active entities as order parameter [34] and including the necessary counter terms, shows that dissipative driving ( $R \sim \mathbf{F}^2$ ) is not due to active terms of the form of eqs. (40) or (41), but due to the existence of a finite relative velocity,  $\mathbf{F}$ , between the active and passive parts of the system. The latter is maintained by the metabolism/chemical energy conversion of the active entities that lead to the polar ordering.

We close this section by pointing out that neglecting reversible counter terms even in passive systems leads to unphysical results. As an example we mention shear flow-induced birefringence and shear wave attenuation close to the (isotropic to nematic or uniaxial to biaxial nematic) phase transitions [50, 51, 54] that are described by reversible pairs of flow/orientation couplings of similar structure as the active terms discussed above. Neglecting there the necessary counter terms would remove these experimentally well-known physical effects.

#### 4 Linear irreversible thermodynamics versus systems driven far from equilibrium

Up to this point we have used the framework of linear irreversible thermodynamics. In this section we would like to comment briefly on systems driven far from equilibrium so that linear irreversible thermodynamics might no longer be applicable. Linear irreversible thermodynamics is a well-established field of statistical physics with applications to many static and dynamic phenomena [21–23, 56]. Key features of linear irreversible thermodynamics include the existence of a generalized thermodynamic potential and of a dissipation function, which is positive semi-definite and controls all dissipative (heat-generating) processes of the system as well as Onsager symmetry for the dissipative transport coefficients.

This situation changes qualitatively as one goes to systems driven far from equilibrium. In general, a generalized thermodynamic potential in the spirit of a Lyapunov functional dictating the dynamic properties of a given system is no longer known. As has already been pointed out a few decades ago, however, there are special situations like the onset of laser action [60–62] or the onset of thermal convection in a simple fluid in the high Prandtl number limit [63], for which one can derive a purely dissipative generalized potential. In addition, it is also not clear how to generalize the symmetry principle of Onsager to all possible non-equilibrium situations.

For almost all macroscopic systems dissipation plays an important role when controlling its dynamics as well as its stationary or oscillatory properties. In order to maintain a stationary state in situations far from equilibrium one typically has a driving force such as an electric field, or gradients in temperature and/or concentration of suf-

ficient strengths, which are balanced by dissipative processes. Thus dissipative processes are of key importance in maintaining driven systems in states far from equilibrium and can therefore never be discarded.

All these various aspects have been demonstrated for stable spatially localized solutions in dissipative driven systems (compare, for example, ref. [64] for a recent review). The most studied prototype equation in this connection is the complex cubic-quintic Ginzburg-Landau equation [65, 66] as it arises as an envelope equation near the onset of a weakly inverted bifurcation to traveling waves. For this equation various types of stable spatially localized solutions have been found [67–70]. To emphasize that both, driving forces and dissipation, must maintain a balance to render the spatially localized solutions in systems driven far from equilibrium to be stable, the term dissipative solitons has been introduced [71] and is frequently used today. The direct biological impact of these concepts has been demonstrated recently [72], where it has been shown that a coupled system of equations combining the complex cubic-quintic Ginzburg-Landau equation with a simple reaction-diffusion system can lead to locally stable solutions with intrinsic flows of matter and energy thus paving the way to the elementary features of living systems.

Nevertheless, we think that the use of near-equilibrium tools, like linear irreversible thermodynamics and dissipative potentials, is justified in the derivation of basic macroscopic dynamic equations for active systems. First, active systems often are not really far from equilibrium, and second, even for systems manifestly far from equilibrium showing *e.g.* chaos, turbulence etc., the basic equations to describe them (*e.g.* the Navier-Stokes equations) are derived close to equilibrium. Only if one switches to a more coarse-grained description, like envelope and amplitude equations, the near-equilibrium tools cannot be used to derive such equations.

#### 5 Concluding remarks

We have investigated the dynamic coupling terms of the macroscopic stress tensor to variables associated with broken continuous symmetries. We have shown that such terms are characteristic of a large variety of complex fluids being passive or driven/active. In particular, the “active term”, as it is called in the literature of active systems, can be present in many passive systems as well. We emphasize that, apart from the structure of the coupling terms to the stress tensor, one cannot see whether the macroscopic system described is passive or active. We have also compared the results for the dynamic contributions to the stress tensor, which are obtained within the approach of dynamic preferred directions with those obtained from other macroscopic models for active systems. We also remark that macroscopic models that neglect the necessary counter term to the “active term” violate linear irreversible thermodynamics and lead to unphysical contributions to the dissipation.

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## Appendix A. Dissipative dynamic contributions to the stress tensor

As discussed in sect. 2, reversible contributions to the stress tensor  $\sigma_{ij}$  have to be even under time reversal, while dissipative ones are odd. Both parts must be even under parity. This observation leads to key input for the symmetry requirements for a specific system to allow dissipative cross-coupling terms between the density of linear momentum and specific other macroscopic variables.

We start this analysis with coupling terms to gradients of scalar variables that are even under parity and time reversal, such as temperature  $T$ , concentration fields  $c_\alpha$  or electric scalar potential  $\phi$  (with the electric field  $E_i = -\nabla_i\phi$ ). In terms of the thermodynamic forces there are coupling terms of the type

$$\sigma_{ij}^D = \mu_{ijk}^\sigma \nabla_k T + \mu_{ijk}^\alpha \nabla_k \mu_\alpha + \mu_{ijk}^e E_k, \quad (\text{A.1})$$

with the relative chemical potential  $\mu_\alpha$  associated with species  $\alpha$ . Such coupling terms are only possible for systems that are odd under time reversal as well as under parity.

The first systems studied macroscopically with these properties were cholesteric liquid crystals in external magnetic fields [73]. We note that one expects such effects to be small in this type of system, since the magnetic field is only applied externally and the broken time reversal symmetry is not an intrinsic property of the system itself. Meanwhile, this type of effects has been studied for two active systems that are intrinsically odd under time reversal and parity, namely for systems that have an axial dynamic preferred direction (and are chiral, in addition) [45] and for those with a polar dynamic preferred direction [34].

Using the notation of ref. [45], the entropy production due to eq. (A.1) is

$$R = q_0 A_{ij} (c_{ijk}^\sigma \nabla_k T + \zeta_{ijk}^e E_k + \zeta_{ijk}^\alpha \nabla_k \mu_\alpha), \quad (\text{A.2})$$

where the pseudoscalar  $q_0$  reflects chirality. Thermodynamics requires  $R > 0$  for dissipative processes, which translates into various positivity requirements for (combinations of) material parameters. The third-rank material tensors take the form

$$\zeta_{ijk} = \zeta_1 \delta_{ij}^\perp w_k + \zeta_2 w_i w_j w_k + \zeta_3 (w_i \delta_{jk}^\perp + w_j \delta_{ik}^\perp), \quad (\text{A.3})$$

with the axial dynamic preferred direction  $w_i$  [45].

These dissipative couplings between stress and temperature, concentration and electrical potential not only lead to the stress tensor eq. (A.1) with  $\mu_{ijk} = q_0 \zeta_{ijk}$ , but also to flow contributions to the heat, concentration, and electrical currents. For example, in a simple shear flow the dissipative heat current  $j_i^{\sigma,D}$

$$j_k^{\sigma,D} = \frac{\partial R}{\partial \nabla_k T} = q_0 \zeta_{ijk}^\sigma A_{ij} \quad (\text{A.4})$$

acquires a non-vanishing contribution in the direction perpendicular to the shear plane.

For a system with a polar dynamic preferred direction  $f_i$ , which transforms like a velocity (odd under parity and time reversal) [34] the analysis is similar to the one just given above. In the polar case, however, no pseudoscalar quantity  $q_0$  is needed to obtain the proper transformation behavior for  $R$  and  $\sigma_{ij}^D$ . It should be noted that in the description of active systems that use a polarization (instead of the dynamic preferred direction  $f_i$ ) no such dissipative stress contribution occurs, since the polarization is even under time reversal.

For macroscopic systems that are intrinsically odd under time reversal and that are characterized by an axial vector another class of coupling terms to the dissipative part of the stress tensor exists. For example, for uniaxial magnetic gels, with  $m_i$  the preferred direction connected to the intrinsic magnetization, one obtains [44]

$$R = \xi_{ijk}^\sigma A_{ij} W_k + c_{ijk}^J A_{ij} h_k^M, \quad (\text{A.5})$$

where  $h_k^M$  is the molecular field associated with the magnetization density  $M_i$  and  $W_k$  is the thermodynamic conjugate of relative rotations between the gel network and the magnetization density. The material tensors  $\xi_{ijk}^\sigma$  and  $c_{ijk}^J$  take the form

$$a_{ijk} = a (m_i \epsilon_{jkl} + m_j \epsilon_{ikl}) m_l. \quad (\text{A.6})$$

To obtain the dissipative parts of the currents and quasi-currents associated with eq. (A.5) we take the partial derivatives with respect to the appropriate thermodynamic forces

$$\sigma_{ij}^D = - \left( \frac{\partial R}{\partial (\nabla_j v_i)} \right) \dots = -\xi_{ijk}^\sigma W_k - c_{ijk}^J h_k^M, \quad (\text{A.7})$$

$$Z_i^D = \left( \frac{\partial R}{\partial W_i} \right) \dots = \xi_{kli}^\sigma A_{kl}, \quad (\text{A.8})$$

$$X_i^D = \left( \frac{\partial R}{\partial h_i^M} \right) \dots = c_{kli}^J A_{kl}. \quad (\text{A.9})$$

There are other macroscopic systems that show such types of dissipative cross-couplings. In achiral systems with an axial dynamic preferred direction these contributions arise as well [45]. The type of coupling provided by  $\sim c_{ijk}^J$  has been elucidated first for superfluid  $^3\text{He-A}$  [31] and superfluid  $^3\text{He-A}_1$  [32] about forty years ago. In this case this coupling is possible because there also exists a vector (usually called  $\hat{l}_i$ ), which is odd under time



reversal and characterizes the spontaneously arising preferred direction in space.

To conclude the section on dissipative cross-coupling contributions to the stress tensor, we discuss, for superfluid  $^3\text{He-A}$  and superfluid  $^3\text{He-A}_1$ , another type of coupling, between normal flow and superflow, of the form

$$\sigma_{ij}^D = -\xi_{ij}\nabla_k\lambda_k^{(s)} \quad (\text{A.10})$$

and

$$J_\varphi^D = -\xi_{ij}\nabla_j v_i^{(n)}, \quad (\text{A.11})$$

where  $\lambda_i^{(s)}$  is the thermodynamic conjugate of the superfluid velocity  $v_i^{(s)} = (\hbar/2m)\nabla_i\varphi$  and  $J_\varphi$  denotes the quasi-current of the phase  $\varphi$ . The tensor  $\xi_{ij}$  is of the uniaxial form

$$\xi_{ij} = \xi_\perp(\delta_{ij} - \hat{l}_i\hat{l}_j) + \xi_\parallel\hat{l}_i\hat{l}_j \quad (\text{A.12})$$

generalizing the second viscosity of an isotropic superfluid [35, 74, 75] to a uniaxial superfluid [31, 32, 76].

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