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Symmetry aspects in the macroscopic dynamics of magnetorheological gels and general liquid crystalline magnetic elastomers

Abstract: We investigate theoretically the macroscopic dynamics of various types of ordered magnetic fluid, gel, and elastomeric phases. We take a symmetry point of view and emphasize its importance for a macroscopic description. The interactions and couplings among the relevant variables are based on their individual symmetry behavior, irrespective of the detailed nature of the microscopic interactions involved. Concerning the variables we discriminate between conserved variables related to a local conservation law, symmetry variables describing a (spontaneously) broken continuous symmetry (e.g., due to a preferred direction) and slowly relaxing ones that arise from special conditions of the system are considered. Among the relevant symmetries, we consider the behavior under spatial rotations (e.g., discriminating scalars, vectors or tensors), under spatial inversion (discriminating e.g., polar and axial vectors), and under time reversal symmetry (discriminating e.g., velocities from polarizations, or electric fields from magnetic ones). Those symmetries are crucial not only to find the possible cross-couplings correctly but also to get a description of the macroscopic dynamics that is compatible with thermodynamics. In particular, time reversal symmetry is decisive to get the second law of thermodynamics right. We discuss (conventional quadrupolar) nematic order, polar order, active polar order, as well as ferromagnetic order and tetrahedral (octupolar) order. In a second step, we show some of the consequences of the symmetry properties for the various systems that we have worked on within the SPP1681, including magnetic nematic (and cholesteric) elastomers, ferromagnetic nematics (also with tetrahedral order), ferromagnetic elastomers with tetrahedral order, gels and elastomers with polar or active polar order, and finally magnetorheological fluids and gels in a one- and two-fluid description.

Keywords: dissipation function, ferromagnetic nematics, hydrodynamics, linear irreversible thermodynamics, magnetorheological fluids, reversible dynamics

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1 General aspects of macroscopic dynamics

1.1 Macroscopic variables

In a microscopic dynamic description, all (or a very large number of) microscopic degrees of freedom are taken into account. Macroscopic dynamics makes use of only those rather few dynamic variables that are not relaxed to their equilibrium values on long, macroscopic time scales. They come in three classes: (A) *conserved quantities* that can only be transported and (B) *symmetry variables*, describing a spontaneously broken continuous symmetry, e.g., a preferred direction that breaks orientational symmetry, but whose orientation is arbitrary, cannot relax at all in the homogeneous limit. Class A and B are the true hydrodynamic variables since for any excitation with frequency ω and wave vector k , there is $\omega \rightarrow 0$ for $k \rightarrow 0$. Class (C) *macroscopic variables* comprise all quantities that only slowly relax and therefore interact with the hydrodynamic variables on macroscopic time scales (where the many microscopic degrees of freedom have already relaxed to their equilibrium values). There are no general criteria to identify such macroscopic variables, and their existence depends on the nature of the individual system.

Examples for conserved quantities are the mass density ρ (or individual mass densities in a mixture without chemical reactions), the momentum density g_i , and the energy density ε . Among the symmetry variables, we consider the nematic director n_i , the vector p_i describing the orientation of a polar preferred direction, and m_i denoting the direction of a spontaneous magnetization. In active systems with polar order, e.g., moving bird flocks or fish schools, the preferred direction is given by the orientation f_i of an active nonzero velocity. In solid (elastic) media, translational symmetry is broken and characterized by the displacement vector u_i . Macroscopic variables are the magnetization M_i in a magnetizable medium, relative velocities w_i in a two-fluid system (different constituents moving with two different velocities), and relative rotations Ω_i among different preferred directions or between a preferred direction and rotations of an elastic body. In the framework of SPP1681 we have worked on many of these systems, which will be discussed in detail, in the following [1–15].

Throughout this chapter, we use the notation m_i, g_i, n_i etc., to characterize vectorial quantities, with indices i running from 1 to 3 in three dimensions. This is a standard notation used in physical hydrodynamics [16].

The variables described above are space-time fields, e.g., $\varepsilon(\mathbf{r}, t)$, that live on macroscopic time and length scales. In the Eulerian description, they are volume densities (of the total quantities of the whole system). The basic assumption to derive general equations for those variables is the applicability of thermodynamics. We start with the local formulation of the *first law of thermodynamics*, which can be interpreted as energy balance of the system. It relates changes of all the variables discussed above to changes of the entropy density $d\sigma$

$$d\varepsilon = Td\sigma + \mu_A dA + \psi_k^B d\nabla_k B + \mu_C dC \quad (1)$$

with T the temperature. The prefactors of the differentials are called (thermodynamic) conjugates.

The energy density is assumed to be a scalar, meaning it does not change under rotations. Thus, all the terms added up in Eq. (1) must be scalars. This is trivially fulfilled for $Td\sigma$ and for $\mu d\rho$, where $\mu_A dA \rightarrow \mu d\rho$ with μ the chemical potential. If A is a vector, like the momentum density, also μ_A has to be a vector connected by the scalar product, $\mu_A dA \rightarrow v_i dg_i$ with v_i the velocity. The B variables must not change the energy, so their contribution in Eq. (1) already contains a gradient. As a consequence also their conjugates must be vectors. Generally the B variables are vector-like quantities by themselves, e.g., $B \rightarrow n_i$, with $\psi_k^B d\nabla_k B \rightarrow \psi_{ik}^n d\nabla_k n_i$. In the case of the nematic director, external (electric or magnetic) fields are important and an additional contribution is added, $h_i^n dn_i$, where h_i^n is due to the dielectric or diamagnetic anisotropy of the nematic phase. Often the two contributions are combined into $h_i^n dn_i$, with $h_i^n = h_i^{nl} - \nabla_k \psi_{ik}^n$ and n_i belonging to class C .

Similarly, when the gradient of the displacement vector $\nabla_k u_i$ is replaced by a symmetric second-rank strain tensor U_{ij} , eliminating solid body rotations that must not contribute to the energy density, U_{ij} belongs to class C with $\mu_C dC \rightarrow \Phi_{ij} dU_{ij}$, where Φ_{ij} is the elastic stress tensor. For a thorough discussion of the relation of u_i with U_{ij} in the nonlinear case, we refer to the study by Pleiner et al. [17].

For the magnetization M_i in a magnetizable system, we have $\mu_C dC \rightarrow h_i^M dM_i$ with h_i^M the internal magnetic field. In a ferromagnetic system, $M_i = M m_i$ can be split into the unit vector m_i , denoting the direction of the spontaneous magnetization, which is a class B variable with $\mu_B dB \rightarrow h_i^m dm_i$, and into the strength of magnetic order M , a class C variable with $\mu_C dC \rightarrow h^M dM$. For active polar order, $F_i = F f_i$, the direction f_i is a class B variable with $\mu_B dB \rightarrow h_i^f df_i$, while F relaxes to its constant stationary value provided by the active entities of the system and is a class C variable. [18]. Relative rotations are rotations of, e.g., the ferromagnetic direction δm_i relative to rotations of the elastic media, $2\Omega_{ij} \equiv \nabla_i u_j - \nabla_j u_i$. They are linearly defined by $\Omega_i = m_j \Omega_{ij} - \delta m_i$ and are class C variables with $\mu_C dC \rightarrow L_i^\Omega d\Omega_i$. For a nonlinear definition of relative rotations, refer the study by Menzel et al. [19].

The *statics of a system* is then given by the phenomenological relation between the thermodynamic quantities and the variables. These relations involve static susceptibilities, e.g., compressibility, specific heat, thermal expansion for the scalar variables, Frank-type rank-4 tensors with Frank coefficients for B variables, and the elastic tensor containing elastic moduli or a rank-2 rotational tensor for the C variables. An efficient way of setting up these static relations is to use a phenomenological *energy functional* of all variables, which is given as follows:

$$E = \int dV \varepsilon(\{\sigma, A, \nabla_k B, C\}) \quad (2)$$

from which the conjugates follow by variational derivation according to Eq. (1). The crucial point is that the functional is restricted by symmetries, in particular space inversion and time reversal symmetry.

Before we discuss these symmetries in the following, we give here the general form of the *dynamic equations* for examples of the different classes of variables:

$$\mathbf{A} \quad (\partial/\partial t)\rho + \nabla_k j_k^{(p)} = 0 \quad (3)$$

$$\mathbf{A} \quad (\partial/\partial t)g_i + \nabla_j \sigma_{ij} = 0 \quad (4)$$

$$\mathbf{B} \quad (\partial/\partial t)n_i + Y_i^n = 0 \quad (5)$$

$$\mathbf{C} \quad (\partial/\partial t)U_{ij} + X_{ij}^{(el)} = 0 \quad (6)$$

$$\mathbf{C} \quad (\partial/\partial t)\Omega_i + X_i^\Omega = 0 \quad (7)$$

$$\mathbf{C} \quad (\partial/\partial t)M_i + Y_i^M = 0 \quad (8)$$

defining the mass current density $j_k^{(p)}$ and the stress tensor σ_{ij} for *A* variables and the (quasi-) currents Y_i^n and $X_{ij}^{(el)}$, X_i^Ω , and Y_i^M for the others. The *A* variables show a divergence in the dynamics, while the *B* variables come with a gradient in the statics. As an effect, both types of variables give rise to the truly hydrodynamic behavior, $\omega(k \rightarrow 0) \rightarrow 0$, while the *C* variables do not.

1.2 Spatial inversion symmetry: statics

In three-dimensional space, not only rotations but also spatial inversion, S_I where $\mathbf{r} \rightarrow -\mathbf{r}$, is an important symmetry operation. Physical quantities behave differently under S_I , either they are invariant (“symmetric”, or “even”) with the signature $\epsilon_S = +1$ or they change sign (they are “antisymmetric” or “odd”) with $\epsilon_S = -1$. The signatures $\epsilon_S = \pm$ are the only possibilities since when inversion is applied twice, the original state is regained, $S_I^2 = +1$.

(True) scalar quantities have $\epsilon_S = +1$, e.g., the variables ρ , σ , and ε , and also the conjugates T and μ . (True) vectors are odd with $\epsilon_S = -1$, in particular, the polarization and the polar preferred direction, electric fields, and the gradient ∇_i , as well as the velocity-type quantities like g_i , v_i , but also a relative velocity w_i . There is a different kind of vectors, called *axial vectors*, that are even under S_I with $\epsilon_S = +1$. Among them, there is the vorticity $2\omega_i = \epsilon_{ijk} \nabla_j v_k$, the magnetization M_i , and any magnetic field H_i . Relative rotations Ω_i are also axial vectors. Slightly more complicated is the case of nematic order. Usually the director n_i is treated as a vector with the additional constraint that all equations involved are invariant under the replacement $n_i \rightarrow -n_i$. Obviously, the director cannot be a polar vector and has to be treated as an axial one, as a necessary condition. We note that the $n_i \rightarrow -n_i$ invariance is a stronger constraint.

Since the energy density in Eq. (1) has $\epsilon_S = +1$, the conjugates of A and C class variables must have equal signatures, while for B class variables, the S_I behavior of the conjugates is opposite to that of the variables due to the gradient involved. For the director, it means ψ_{ij}^n has $\epsilon_S = -1$ but h_i^n has $\epsilon_S = +1$. Regarding the dynamics, Eqs. (3)–(8), the time t is S_I symmetric, and therefore, the quasi-currents of B and C class variables have the same spatial signature as the appropriate variables. For A class variables, this is opposite due to the gradient involved.

Depending on the symmetry of a given system, it can be S_I symmetric (“centrosymmetric”) or not. Prominent examples for the latter are *chiral phases*, like cholesteric liquid crystals. Such phases show optical activity (rotation of the plane of polarization of light) that is described by a pseudoscalar quantity q_0 with $\epsilon_S = -1$. The use of q_0 in the macroscopic dynamic equations exactly characterizes those contributions and couplings that are specific for the chiral system and that would be forbidden in an achiral phase without a q_0 . Well-known examples are the linear twist term in the Frank energy in cholesterics that give rise to the helical ground state and the static couplings between director rotations and, e.g., the thermal degree of freedom leading to the static Lehmann effect [20], absent in the achiral nematic phase. The origin of the existence of a q_0 can be microscopic, when chiral molecules are present, or it is a result of a complicated internal structure like in the smectic C_{B2} phase [21]. In the latter case, q_0 and $-q_0$ lead to two chiral structures with opposite handedness but with the same energy for the ground state: ambidextrous chirality [22].

The existence of a polar vector p_0 also leads to a noncentrosymmetric phase, like polar nematics [23, 24]. Again, the occurrence of p_0 denotes those contributions that would not be allowed in a centrosymmetric phase. Such a phase is not chiral.

A third possibility to break S_I symmetry in a phase is due to the existence of *tetrahedral (or octupolar) order* described by a third-rank tensor order parameter $T_{ij} = \sum_{\alpha=1}^4 n_i^{(\alpha)} n_j^{(\alpha)} n_k^{(\alpha)}$, where the $n_i^{(\alpha)}$ are the position vectors of the corners of a tetrahedron [25]. T_{ijk} has $\epsilon_S = -1$ since the $n_i^{(\alpha)}$ are true vectors. The hydrodynamic variables related to tetrahedral order are the 3-D rigid rotations $d\Gamma_i \sim \epsilon_{ipq} T_{pkl} dT_{qkl}$ of the T_{ijk} structure. This is a class B variable with $\psi_k^\beta d\nabla_k B \rightarrow \psi_{ik}^\Gamma d\nabla_k \Gamma_i$ and

$$(\partial/\partial t)\Gamma_i + Y_i^\Gamma = 0 \quad (9)$$

The variable Γ_i is S_I symmetric as is the quasi-current Y_i^Γ , while ψ_{ik}^Γ is odd. Similar to the nematic case, one can introduce a h_i^Γ that contains $\nabla_j \psi_{ij}^\Gamma$ and is even under S_I . There are subtleties in the nonlinear regime due to the non-Abelian nature of three-dimensional rotations, for details refer the study by Brand and Pleiner [26]. A phase that only contains tetrahedral order is neither chiral nor polar. If there is additionally nematic order (along a 2-fold symmetry axis of the tetrahedron), a D_{2d} phase occurs that is still not chiral (and not polar) but shows *ambidextrous helicity* [27]. Only when the tetrahedral structure is suitably combined with a biaxial nematic order, a chiral (but still

nonpolar) phase D_2 can occur, where the q_0 is due to the different preferred directions involved [28]. Again, this is an example for *ambidextrous chirality*.

1.3 Time reversal symmetry

Under time reversal T_R , where $t \rightarrow -t$, all macroscopic quantities are either even (T_R symmetric) with $\epsilon_T = +1$ or they are odd (T_R antisymmetric) with $\epsilon_T = -1$ since $T_R^2 = +1$. There is no need to discriminate among the A, B, C case variables because the gradient is T_R symmetric, $\epsilon_T(\nabla_i) = +1$. The energy density is T_R symmetric, and therefore, the conjugates have the same T_R signature as the variables, $\epsilon_T(e) = \epsilon_T(C_e)$ for any variable e with conjugate C_e . Examples for T_R symmetric variables are $\rho, \sigma, n_i, p_i, U_{ij}$, and Ω_i , as well as their conjugates $\mu, T, h_i^n, h_i^p, \Phi_{ij}$, and L_i^Ω . Odd quantities with $\epsilon_T = -1$ are g_i and all types of velocities, including relative velocities and vorticity, M_i , and h_i^M , as well as magnetic fields.

T_R is closely related to the *second law of thermodynamics*. The latter states that reversible, time reversal symmetric processes with $\epsilon_T = +1$ must not change the entropy – they are nondissipative, while irreversible processes with $\epsilon_T = -1$ must increase the entropy – they are dissipative. Therefore, one can uniquely split up any current or quasi-current into a reversible (superscript R) and an irreversible part (superscript D), e.g., for a variable e , the following equation can be written:

$$(\partial e / \partial t) + Y_e^R + Y_e^D = 0. \tag{10}$$

If time is reversed, Eq. (10) reads $(\partial e / \partial t) + Y_e^R - Y_e^D = 0$. independent of the signature $\epsilon_T(e)$.

Thus, $\epsilon_T(Y_e^R) = \epsilon_T(e)$ and $\epsilon_T(Y_e^D) = -\epsilon_T(e)$. In particular,

$$\epsilon_T(Y_e^R) = \epsilon_T(\partial e / \partial t) = \begin{cases} -1 & \text{for } T_R - \text{even } e \\ +1 & \text{for } T_R - \text{odd } e \end{cases} \tag{11}$$

$$\epsilon_T(Y_e^D) = -\epsilon_T(\partial e / \partial t) = \begin{cases} +1 & \text{for } T_R - \text{even } e \\ -1 & \text{for } T_R - \text{odd } e \end{cases} \tag{12}$$

Among the reversible currents, σ_{ij}^R, Y_i^{MR} are T_R even with $\epsilon_T = +1$, while $j_i^{\rho R}, Y_i^{nR}$, and $X_{ij}^{(el)R}$ are odd, with $\epsilon_T = -1$. On the other hand, the dissipative currents σ_{ij}^D, Y_i^{MD} have $\epsilon_T = -1$, while $j_i^{\rho D}, Y_i^{nD}$, and $X_{ij}^{(el)D}$ are even, $\epsilon_T = +1$.

We add the entropy balance equation (which is a conservation law in the reversible case) and the energy density conservation law to obtain the following equations:

$$\partial \sigma / \partial t + j_i^\sigma = 2R/T \tag{13}$$

$$\partial \mathcal{E} / \partial t + j_i^\mathcal{E} = 0 \tag{14}$$

with the dissipation function R as the entropy source term. Putting all macroscopic dynamic equations, including Eqs. (3)–(8), into the Gibbs relation, Eq. (1), one gets a representation of the dissipation function in terms of the dissipative currents and thermodynamic forces

$$2R = -j_i^{\sigma D} \nabla_i T - \sigma_{ij}^D A_{ij} + Y_i^{nD} h_i^n + Y_i^{MD} h_i^M + X_{ij}^{(el)D} \Phi_{ij} + X_i^{\Omega D} L_i^\Omega + Y_i^{\Gamma D} h_i^\Gamma > 0 \quad (15)$$

Note that for case C variables, the thermodynamic forces are just the conjugates, while for case A variables (the conserved variables), the forces are the gradients of the conjugates $2A_{ij} = \nabla_j v_i + \nabla_i v_j$. Case B variables (symmetry variables) have been written in the form of class C variables, e.g., with h_i^n , the conjugate to dn_i , as force, rather than using ψ_{ij}^n , the conjugate to $d\nabla_j n_i$. There is no dissipative contribution $\sim j_i^{\rho D}$ since the mass density current is the momentum density $\rho v_i = g_i$, which is reversible.

In Eq. (15), pure divergence terms have been omitted since they do not contribute to the total entropy production $\sim \int R dV$ in the bulk. R is even under T_R since in the opposite case, one would get $R < 0$ for time reversed dissipative processes in contradiction to thermodynamics.

The dissipation function R can be used as a potential for dissipative currents. Within *linear irreversible thermodynamics*, with a linear relationship between currents and forces, one can set up phenomenologically R as a harmonic function of the thermodynamic forces. The dissipative currents then follow by partial derivation of R with respect to the forces. Examples are $j_i^{\sigma D} = -\partial R / \partial \nabla_i T$, $Y_i^{nD} = \partial R / \partial h_i^n$, and $X_i^{(el)D} = \partial R / \partial \Phi_{ij}$. If R contains nonvanishing Φ_{ij} contributions, e.g., $\sim \Phi_{ij} \Phi_{kl}$, the resulting $X_{ij}^{(el)D}$ describes relaxation of the strain tensor. This is the hydrodynamic description of *viscoelasticity* of non-Newtonian fluids or relaxing gels. In solids or chemically bonded elastomers, only gradients of the elastic stresses act as forces, e.g., $R \sim (\nabla_j \Phi_{ij})(\nabla_l \Phi_{kl})$, and the strains behave as conserved quantities (“permanent elasticity”) showing diffusion rather than relaxation.

Generally, the dissipative currents are related to the forces by linear relations

$$Y_a^D = \zeta_{ab}^D F_b \quad (16)$$

where a and b denote the variables involved. By the very existence of the potential R , ζ_{ab}^D is symmetric, $\zeta_{ab}^D = \zeta_{ba}^D$. Of course, such relations have to fulfill the T_R symmetry properties discussed above, in particular, $\epsilon_T(\zeta_{ab}^D) = \epsilon_T(ab)$. If the (dissipative) transport parameter ζ_{ab}^D is constant or only depends on structural properties that are T_R symmetric, like the nematic director, only variables of equal T_R signature can couple, $\epsilon_T(a) = \epsilon_T(b)$. On the other hand, if there is a T_R -odd quantity present, like the magnetization or a magnetic field, an odd power of them in ζ_{ab}^D leads to $\epsilon_T(\zeta_{ab}^D) = -1$, and the variables a and b must behave oppositely $\epsilon_T(a) = -\epsilon_T(b)$.

For the *reversible dynamics*, there are somewhat different rules. Since the reversible currents must not contribute to R , they have to fulfill the following equation:

$$\begin{aligned} 0 = & -j_i^{\rho R} \nabla_i \mu - j_i^{\sigma R} \nabla_i T - \sigma_{ij}^R A_{ij} + Y_i^{nR} h_i^n \\ & + Y_i^{MR} h_i^M + X_{ij}^{(el)R} \Phi_{ij} + X_i^{\Omega R} L_i^\Omega \end{aligned} \quad (17)$$

replacing Eq. (15) for the dissipative currents. Since the bilinear products of thermodynamic forces and reversible currents add up to zero, there is no potential, from which the latter can be derived.

The reversible currents come in two parts, $Y_a^R = Y_a^0 + \tilde{Y}_a^R$, where Y_a^0 are the non-phenomenological, material-independent, symmetry-given contributions, and \tilde{Y}_a^R are the phenomenological parts characterized by (reversible) transport coefficients. The former comprise advection, convection, and rotational covariant derivatives, in addition to, e.g., the isotropic pressure p and the Ericksen stress in the stress tensor, and read as follows [2, 16, 29]:

$$j_i^{\rho 0} = \rho v_i \quad (18)$$

$$j_i^{\sigma 0} = \sigma v_i \quad (19)$$

$$j_i^{\varepsilon 0} = (\varepsilon + p) v_i \quad (20)$$

$$\begin{aligned} \sigma_{ij}^0 = & g_i v_j + \delta_{ij} p - \Phi_{ij} + \Phi_{jk} U_{ik} + \Phi_{ik} U_{jk} \\ & + \frac{1}{2} (\psi_{ki}^n \nabla_j n_k + \psi_{kj}^n \nabla_i n_k) \end{aligned} \quad (21)$$

$$Y_i^{m0} = v_k \nabla_k n_i + \epsilon_{ijk} n_j \omega_k \quad (22)$$

$$Y_i^{M0} = v_k \nabla_k M_i + \epsilon_{ijk} M_j \omega_k \quad (23)$$

$$X_{ij}^{(el)0} = v_k \nabla_k U_{ij} - A_{ij} + U_{kj} \nabla_i v_k + U_{ki} \nabla_j v_k. \quad (24)$$

with $p = -\varepsilon + \mu\rho + T\sigma + v_i g_i + h_i^M M_i$ containing all extensive variables and their conjugates. The stress tensor has been symmetrized in order to guarantee angular momentum conservation using the condition that Eq. (1) is invariant under rotations and that a divergence of an antisymmetric term is irrelevant [16]. All nonphenomenological contributions together fulfill Eq. (17).

Concerning the phenomenological parts, we first note that $\tilde{j}_i^{\rho R} = 0$ because $(\partial\rho)/(\partial t) + \nabla_i g_i = 0$. For the other currents, one has to set up the linear current-force relations explicitly

$$\tilde{Y}_a^R = \zeta_{ab}^R F_b \quad (25)$$

In accordance with Eq. (17), there must be a counter term $\tilde{Y}_a^R = \zeta_{ab}^R F_a$ such that $\zeta_{ab}^R + \zeta_{ba}^R = 0$ indicating antisymmetry with respect to a and b . Of course, the relation Eq. (25) has to be invariant under T_R with the result $\epsilon_T(\zeta_{ab}^R) = \epsilon_T(\tilde{Y}_a^R F_b)$ or $\epsilon_T(\zeta_{ab}^R) = -\epsilon_T(ab)$ due to $\epsilon_T(F_b) = \epsilon_T(C_b) = \epsilon_T(b)$. If ζ_{ab}^R is constant or only depends on structural properties that are T_R symmetric, like the nematic director, only variables of opposite T_R signature can couple, $\epsilon_T(a) = -\epsilon_T(b)$. On the other hand, if there is a T_R -odd quantity present, like the magnetization or a magnetic field, an odd power in ζ_{ab}^R leads to $\epsilon_T(\zeta_{ab}^R) = -1$, and variables a and b of the same T_R signature can couple. In such systems, it is possible to have dissipative as well as reversible self-couplings (or cross-couplings between two given variables), when the dissipative and reversible transport parameters have different T_R signatures. An example is (isotropic) heat conduction in ferromagnetic systems, where $j_i^{\sigma R} = -\kappa \nabla_i T$ and $j_i^{\sigma R} = -\kappa^T \epsilon_{ijkMk} \nabla_j T$ [12]; we note that the latter contribution also exists when linearized since in a ferromagnetic system $\langle M_i \rangle \neq 0$.

Time reversal symmetry is often overlooked and sometimes mistreated as shown in the study by Brand et al. [1, 11].

1.4 Spatial inversion symmetry: dynamics

Of course, spatial inversion S_I is relevant for the dynamics as well. From Eqs. (3)–(8), we deduce that the physical currents of class A variables (e.g., j_i^{ρ} and σ_{ij}) have an S_I signature opposite to that of the variables, $\epsilon_S(j_a) = -\epsilon_S(a)$, while for all other variables, the quasi-currents have $\epsilon_S(Y_a) = +\epsilon_S(a)$. This applies to the reversible parts, as well as well as to the irreversible ones. Writing the second law of thermodynamics in the form of Eqs. (15) and (17), the forces for class A variables are gradients of the conjugates (defined in Eq. (1)) with the result $\epsilon_S(F_a) = -\epsilon_S(C_a) = -\epsilon_S(a)$, while for the other variables, the forces are just the conjugates resulting in $\epsilon_S(F_a) = \epsilon_S(C_a) = \epsilon_S(a)$.

For the dissipative and reversible transport parameters $\zeta_{ab}^{(D,R)}$ defined in Eqs. (16) and (25), this implies $\epsilon_S(\zeta_{ab}) = +\epsilon_S(ab)$ for all types of variables. Thus, if ζ_{ab} is a constant or only depends on structural properties that are S_I symmetric, like the nematic director, only variables of equal S_I signature can couple, $\epsilon_S(a) = +\epsilon_S(b)$. On the other hand, if there is a S_I -odd quantity present, like the pseudoscalar q_0 or the tetrahedral order T_{ijk} , an odd power of those in ζ_{ab} leads to $\epsilon_S(\zeta_{ab}) = -1$, and the variables a and b must behave oppositely $\epsilon_S(a) = -\epsilon_S(b)$. Examples are the dissipative dynamic Lehmann effect in chiral systems, where, e.g., temperature gradients (polar vectors) can couple to director rotations (axial vectors) [11, 30] and flow-induced reversible (heat) fluxes in tetrahedral systems, where the nonpolar A_{ij} couples to the polar vector $j_i^{\sigma R}$ [31].

2 Nematic order and magnetism

2.1 Isotropic and uniaxial magnetic elastomers and gels

Isotropic ferrogels are systems without a preferred direction combining the properties of a gel and of a conventional magnetic liquid. The hydrodynamic description of isotropic ferrogels has been presented in the study by Jarkova et al. [32]. Quite recently, a continuum model for ferrogels from an engineering perspective has been developed [33]. In parallel microscopic studies of the field-controlled change of shape and elasticity of magnetic gels using particle-based simulations have been advanced; these developments have been elucidated in the study by Weeber et al. [34]. A large effort has been dedicated to the mesoscopic characterization of magnetic hybrid materials such as magnetic gels and elastomers over the last few years, compare, for example, the study by Menzel [35] for a recent review. Important progress along these lines has been achieved for the tunable dynamic moduli of magnetic elastomers on the mesoscale by combining experimental data from X-ray tomography with coarse-grained dipole – spring modeling [36]. From an applied point of view, there has been a special emphasis on the magnetic field–controlled mechanical behavior of magnetosensitive elastomers in applications for actuator and sensor systems [37].

Uniaxial magnetic gels with a permanent magnetic moment have been synthesized by performing the cross-linking process in an external magnetic field [38] and afterward characterized with respect to their optical, magnetic, and mechanical properties [38]. For the hydrodynamic description of uniaxial magnetic gels and elastomers, we refer to the study by Bohlius et al. [39]. In the study by Menzel [40], it has been demonstrated how one can bridge the scales from particles to macroscopic length scales in a uniaxial magnetic gel. Very recently, the structure and the magneto-optical response of anisotropic fibrillous organoferrogels with mobile magnetic nanoparticles have been investigated [41]. While in this study no magnetic hysteresis has been found but an optical hysteresis is detected, revealing a complex interplay between the gel and the mobile magnetic particles [41].

2.2 Magnetic nematic elastomers and gels

Nematic elastomers with a magnetic degree of freedom are described by the director, the strain field, relative rotations between director and elastic rotations, and by a magnetization variable that is zero in equilibrium [2]. We concentrate here on the magnetic effects. There are reversible dynamic couplings between director rotations and the magnetization dynamics, $(\partial n_i / \partial t) \sim h_j^M$ and $(\partial M_i / \partial t) \sim h_j^n$, and similarly between relative rotations and the magnetization, $(\partial \Omega_i / \partial t) \sim h_j^M$ and $(\partial M_i / \partial t) \sim L_j^\Omega$, all with reversible material tensors of the form $\sim \epsilon_{ijk} n_k$. The symbol \sim in the relations above

indicates that the vectorial quantities on the left and on the right hand side are connected by second-rank tensors. It is the different T_R signature of the variables involved and the antisymmetry of the material tensors that allow these couplings. Applying an external magnetic field, rotations of the director and relative rotations are induced (through h_i^M). Through similar reversible couplings, external shear flow induces relative rotations.

In ordinary nematics flow alignment of the director is a prominent feature: shear flow rotates the director at a finite angle w.r.t. the flow direction. This angle is independent of the shear rate Σ and depends on a reversible flow parameter. In the present case, shear flow also induces relative rotations, which induce a magnetization, and finally, all three variables, n_i , Ω_i , and M_i are oriented, independent of the shear rate. Only the magnitude of the magnetization and the relative rotations are proportional to Σ . Since the system is elastic, the shear rate has to be oscillatory, $\Sigma \cos(\omega_s t)$. Since there is static coupling between relative rotations and strains, the relative rotations show a phase shift compared to the shear rate indicating that in this complicated system, flow alignment is no longer a pure reversible feature.

If a chiral agent is added or a chiral nematogenic molecule is used, a chiral magnetic nematic elastomer or gel is obtained [3]. The only new structural element compared to the achiral case is the pseudoscalar q_0 that is odd under S_I . A well-known consequence is the occurrence of a linear (nematic) twist term in the energy density functional, giving rise to a helical ground state, as well as bilinear coupling terms between twist and temperature and density changes and strains. The latter describe the temperature, density, and strain dependence of the helical pitch. They also give rise to the static part of the Lehmann effect. Note that there are no static couplings to changes of the magnetization due to the odd T_R signature of the latter.

In the dynamics, the presence of q_0 allows couplings between polar currents and axial forces (and vice versa) like, e.g., $j_i^{\sigma R} \sim q_0 h_j^M$ and $Y_i^{MR} \sim q_0 \nabla_j T$. Applying a temperature gradient along the helical axis, $\nabla_z T$, the growth of M_z is induced that saturates into a finite stationary magnetization, $M_z^{(stat)} \sim q_0 \nabla_z T$. Instead of the temperature gradient, also an external electrical field can be used, thus creating an electric field-induced magnetization [3]. The elastic degree of freedom is irrelevant for these effects.

2.3 Ferromagnetic nematics

Ferromagnetic nematic liquid crystals are fluids with two different kinds of internal order, a nematic one due to the ordering of standard nematogenic molecules and a ferromagnetic one due to the ordering of magnetic nanoplatelets made of complicated iron oxide compounds. Such suspensions have been realized by Mertelj et al. [42, 43]. In equilibrium, the two preferred directions n_i and m_i are parallel. Thus, there is an energetic penalty, $A_2[(n \cdot m)^2 - 1]$, for deviations from the ground state. In addition, there

are various (reversible and irreversible) dynamic couplings of these two variables. The full hydrodynamic description for ferromagnetic nematics has been given in the study by Potisk et al. [9], which is a generalization of an earlier work [44] on “ferronematics”, i.e., magnetic nematic liquid crystals without permanent magnetization. From a microscopic point of view, the study of the structure and rheology of hybrid mixtures of magnetic nanoparticles in liquid crystals using particle resolved simulations has started a few years ago. Quite recently, the focus in this area has been on the transport properties in liquid crystal-magnetic colloid mixtures [45].

A particularly interesting cross-coupling is the (simplified) dissipative dynamics [8, 44]

$$Y_i^{nD} = \frac{1}{\gamma_1} h_i^n + \chi_{ij}^D h_j^m \tag{26}$$

$$Y_i^{mD} = b_{\perp}^D h_i^m + \chi_{ji}^D h_j^n \tag{27}$$

with the symmetric material tensor $\chi_{ij}^D = \chi_2^D m_k n_k (\delta_{ij} - n_i n_j)$. The crucial point for its existence is that χ_{ij}^D is odd under T_R , as well as under the replacement $n_i \rightarrow -n_i$. For the experiment considered below, the force h_i^n is due to the Frank rotational elastic energy and a surface anchoring energy, while h_i^m describes the orienting force of an external field on a ferromagnet; of course, both forces contain contributions from the A_2 energy given above.

In the studies by Potisk et al. [8, 9], magneto-optic response experiments are described. On a sample of homogeneous structure $n||m$, a field is applied perpendicularly that rotates the magnetization toward the field direction. Due to the coupling between m and n , also the director starts to rotate, which is hindered by the surface anchoring and the subsequent deformation of the director field. Finally, a stationary state is reached that depends on the field strength. Sending light through the sample (along the field direction), using a polarizer and analyzer, the measured phase shift between the ordinary and extraordinary beams allows to monitor the final director orientation, as well as the switching dynamics. It turns out that the director relaxation (γ_1) and the magnetization relaxation (b_{\perp}^D) alone are not sufficient to describe the results, but a nonzero χ_2^D is mandatory. Indeed, χ_2^D turns out to be rather large of the order of γ_1 . This is a clear indication that, in such complicated systems, dynamic cross-couplings are important.

There is a reversible counterpart, $\chi_{ij}^R = \chi^R \epsilon_{ijk} n_k$, which is even under T_R and leads to a vanishing entropy production due to the antisymmetry of ϵ_{ijk} . The effect of adding those terms to Eqs. (26) and (27), not yet detected experimentally, would be an out-of-plane rotation of the director [9].

Another area to detect the complexity of ferromagnetic nematics is flow. In ordinary nematics, there are basically two flow effects, viscosity and flow alignment due to a reversible coupling between (symmetric) shear flow and director rotations. In

ferromagnetic nematics, there are additional cross-couplings possible due to the odd T_R signature of the magnetization. In particular, there are dissipative couplings between flow and the nematic and magnetic forces, h_i^n and h_i^m , and a reversible one involving h_i^m and a reversible version of the viscous coupling [10]. As a result, the velocity profile in a (simple) shear experiment generally deviates from linearity, when a field is present, and effective viscosities become field dependent increasing by a factor up to two in accordance with experiments [46]. In ordinary nematics, there are 3 Miesowicz viscosities according to the three possibilities to fix the director relative to the shear geometry (along the velocity field, along the velocity gradient, and perpendicular to both). In ferromagnetic nematics, there are nine different ways to fix independently (by different external fields) the orientation of n and m relative to shear flow. Due to the various couplings among the variables, these effective viscosities are rather complicated functions of the reversible and irreversible transport parameters involved [10].

2.4 Ferromagnetic nematics with tetrahedral order

When tetrahedral order is added to ferromagnetic nematics, three preferred structures exist – the nematic director n_i , the magnetization m_i , and the tetrahedral structure T_{ijk} (for the latter, refer the study by Fel [25] and the discussion around Eq. (9)). The interesting point in this situation is that m_i and T_{ijk} have opposite symmetry properties, $\epsilon_S = +1$ $\epsilon_T = -1$, and $\epsilon_S = -1$, and $\epsilon_T = +1$, respectively. We will only consider the case where the director is fixed to be parallel to one of the 2-fold symmetry axes of T_{ijk} . This is the structure of a D_{2d} phase [26]. The hydrodynamic variables are the rotations of the director and the rotation of the tetrahedral structure about the director. Rotations of the magnetization are independent degrees of freedom, but due to an energetic coupling between n_i and m_i , the two directions are parallel in equilibrium, and the phase is uniaxial. For the full hydrodynamics of such a phase, refer the study by Potisk et al. [12].

In the statics, the most prominent feature in the D_{2d} phase is the linear gradient term in the energy density $\sim T_{ijk} n_i \nabla_j n_k$. It favors helical structures of n_i and T_{ijk} (about a second, perpendicular 2-fold tetrahedral symmetry axis). For the magnetization, there is an analogous linear gradient term, $\sim T_{ijk} m_i \nabla_j m_k$. The sum of the two linear terms favors a combined helical structure of n_i and m_i (and T_{ijk}), thereby preserving n_i and m_i remaining parallel. Since the phase is achiral (there is no pseudoscalar quantity present), this is ambidextrous helicity.

In the reversible dynamics, there are very special couplings that only exist since all three ordered structures are involved. An example is the coupling between the thermal degree of freedom and director rotations, $j_i^{OR} = \xi_{ij}^{Tn} h_j^n$ and $Y_i^{nR} = \xi_{ji}^{Tn} \nabla_j T$. Here, ξ_{ji}^{Tn} contains linearly n_k , m_b , and T_{pqr} , thereby accommodating the $n_i \rightarrow -n_i$ invariance, ensuring the reversibility of the currents (due to m_i), and compensating for the polarity

of the heat current and the temperature gradient (due to T_{pqr}). Since ξ_{ji}^{Tn} is symmetric, the two contributions to Eq. (17) cancel as required.

In the dissipative dynamics, the magnetization couples, e.g., to the thermal degree of freedom, $J_i^{\sigma D} = \psi_{ji}^{TD} h_j^m$ and $Y_i^{mD} = -\psi_{ji}^{TD} \nabla_j T$. Here, ψ_{ji}^{TD} contains linearly m_l (ensuring the irreversibility of the currents) and T_{pqr} (compensating for the polarity of the heat current and the temperature gradient). Since ψ_{ji}^{TD} is symmetric, the two contributions to Eq. (15) are equal and add up as required. They describe that rotations of the magnetization create heat currents, while temperature gradients drive the dynamics of the magnetization.

3 Elasticity with nonnematic order

In this section, we discuss various elastic systems that show either a polar preferred direction or an ordered tetrahedral structure – finally together with a permanent magnetization. Differences can be traced back to the different symmetry signatures of the order parameters involved.

3.1 Polar and active polar gels

If in a nematic liquid crystal the director n_i is replaced by a polar vector p_i , the direction of a permanent polarization, a polar (nematic) phase results. Due to the different symmetry properties, $\epsilon_S(n_i) = +1$ and $\epsilon_S(p_i) = -1$, the polar phase behaves differently compared to the nematic one. In particular, polar phases tend to form splay textures [23] due to a linear splay term $\nabla_i p_i$ in the energy functional. Neither in nematics nor in ferromagnetic systems linear splay ($\nabla_i n_i$ or $\nabla_i m_i$) is possible.

In the study by Brand et al. [5], the hydrodynamics of polar gels is given. It turns out that the reversible dynamics is isomorphic to that of nematic gels [47] and need not to be repeated here. In the statics, there is the standard piezoelectric coupling between polarization and strain. Here, in the uniaxial case, it contains three static susceptibilities. There is another static coupling between polar textures and elasticity, $\psi_{ij}^p \sim U_{kl}$ and $\Phi_{ij}^{(el)} \sim \nabla_k p_l$, with $\psi_{ij}^p \equiv (\partial \epsilon) / (\partial \nabla_i p_j)$. Relative rotations between p_i and the network, $\Omega_i \equiv \delta p_j \Omega_{ij} - \delta p_i$, couple to bend distortions of the polarization, $L_i^\Omega \sim \delta_{ik}^\perp p_j \nabla_j p_k$ and $\psi_{ij}^p \sim \delta_{ik}^\perp p_j \Omega_k$. This effect is not possible in nematic gels (because of the $n_i \rightarrow -n_i$ invariance) and neither in ferrogels (due to $\epsilon_T(m_i) = -1$).

Variants of polar ordered systems are *active polar* ones, where biological entities (e.g., fish schools, bird flocks, locust swarms, bacteria, etc.) move coherently in a specific direction, without a head to tail symmetry. The preferred direction is given by the velocity, $F_i = F f_i$, of the active entities. If the movement stops, also the order

vanishes. To maintain the active velocity, the entities have to provide energy. This energetic intake is dissipated in the system and defines a nonequilibrium state with $F = F_0$. Nevertheless, it can be described in the usual hydrodynamic way just adding the driving force [18].

In the study by Pleiner et al. [4], we investigate an active polar, viscoelastic system with a relaxing elasticity, as, e.g., occurs in movements of bacteria colonies in a gel background. The additional variables are the relaxing strain tensor U_{ij} , rotations of the preferred direction, δf_i with $f_i \delta f_i = 0$, relaxation of F (toward F_0), and relative rotations between f_i and the network, $\Omega_i = f_j \Omega_{ij} - \delta f_i$. Note that Ω_i has the same symmetry signatures as f_i . Apart from the active velocity, there is also a passive one, $v_i^{(1)}$, describing movements of the passive background. In such a two-fluid system, the question arises, with which velocity a given variable is convected or transported (Eqs. 18–24). It turned out that there is no general principle to answer this question, and generally, those transport or convection velocities are material dependent, containing passive and active parts [48, 49]. In a linearized passive dynamics, the transport terms drop out, but in the active case, with a constant active velocity in the stationary state, even the linearized theory contains transport etc. due to the active velocity, and this active transport is an important part of the dynamics. We mention that sometimes a polarization vector is used to describe active transport [50], but due to the different T_R signature of the polarization compared to a velocity, the transport becomes "irreversible" violating thermodynamics and opening up an unphysical dissipation channel [4].

Results involving the active polar order and the elasticity are coupled relaxations of compressional strains, U_{zz} , with F and shear strains U_{xz} with relative rotations Ω_x , where F relaxes to F_0 and the other quantities to zero. The sound spectrum of a dynamical system is another important aspect of the physics involved. In simple fluids, one has (ordinary) sound with $\omega^2 = c_1^2 k^2$, with ω and k being the frequency and wave number of the linear excitation, respectively. It is purely reversible, and dissipation only enters at higher k -orders. If in addition the active velocity is taken into account, a second sound excitation arises that is coupled to the first one. However, the full sound spectrum is generally no longer invariant under $\omega \rightarrow -\omega$, indicating a nonequilibrium situation [18]: In the stationary state, wave propagation parallel or antiparallel to the active velocity is different. The elastic degree of freedom gives, by coupling to flow, another sound-like excitation (shear elastic wave) that couples to the other sound-like excitations. However, in the present case, elasticity is relaxing, and dissipation does not come at higher k powers but is already present to order k^0 , which makes the final sound spectrum for this material very complicated [4].

3.2 Tetrahedral and ferromagnetic tetrahedral gels

In the study by Brand and Pleiner [6], we consider tetrahedral gels and elastomers, i.e. elastic media with a tetrahedral ordered structure described by the rank-3 tensor T_{ijk}

(already discussed above). A liquid phase that only contains tetrahedral order, T_d , is optically isotropic since the rank-2 dielectric tensor cannot couple to T_{ijk} . When combined with (isotropic) elasticity, a ϵT_d phase arises. Although the time reversal and space inversion properties of polarization and tetrahedral order are the same, the properties of ϵT_d are quite different from the polar (nematic) phase. One reason is that $T_{iik} = 0$ and ϵT_d is apolar. On the other hand, the rank-3 properties of T_{ijk} allow for specific couplings to the elastic tensors, in particular, $J_i^{\sigma D} \sim T_{ijk} \Phi_{jk}^{(el)}$ and $X_{ij}^{(el)D} \sim T_{ijk} \nabla_k T$ describe a dissipative coupling between elasticity and the thermal degree of freedom. The specific aspect of this coupling lies in the geometry – elastic shear stresses induce a heat current perpendicular to the shear plane, and a temperature gradient induces growth of perpendicular elastic shear strains. For a relaxing elasticity, this results finally in a stationary induced strain. Out-of-plane phenomena are characteristic for tetrahedral order.

If chirality is present (due to a pseudoscalar q_0), a T phase occurs in the liquid case [28] and a ϵT in the elastic case. The combination $q_0 T_{ijk}$ gives a S_T -positive rank-3 tensor that allows, particularly, couplings of the elastic degree of freedom with rotations of the tetrahedral structure, $\delta \Gamma_i$, and with relative rotations, Ω_i , (in the statics and in the dissipative dynamics) and with flow (in the reversible dynamics). Here, $\Omega_i = \epsilon_{ijk} \Omega_{jk} - \delta \Gamma_i$ means rotations of the tetrahedral orientation with respect to the elastic medium.

If a tetrahedral gel or elastomer is in addition ferromagnetic, the different time reversal and space inversion signatures of the tetrahedral structure and the permanent magnetization allow for a very rich hydrodynamic theory [13]. We assume the magnetization to be rigidly connected to one of the 2-fold symmetry axes of the tetrahedral structure. This preferred direction is taken as the z -direction. Relative rotations here mean combined rotations of m_i and T_{ijk} relative to the elastic network.

We concentrate on the interplay of elasticity and magnetization in the presence of tetrahedral order. First, shear strains create (statically) magnetization patterns $\nabla_z m_x \sim U_{yz}$ and $\nabla_z m_y \sim U_{xz}$ with the magnetization perpendicular to the shear plane. Compressional strains lead to magnetization patterns in the perpendicular plane $(\nabla_x m_y + \nabla_y m_x) \sim U_{zz}$. This should not be mixed up with the standard magnetostriction, where compressional strains U_{zz} or $U_{xx} + U_{yy}$ result in changes of the magnitude of the magnetization, $\delta M/M_0$. In addition, there is an indirect coupling of temperature gradients to elastic shear stresses, mediated by relative rotations, such that $\nabla_x T$ and $\nabla_y T$ create $\Phi_{yz}^{(el)}$ and $\Phi_{xz}^{(el)}$, respectively.

In the case of a transient network, a temperature gradient along the preferred axis triggers temporal changes of the strains, which finally result in a stationary elastic shear stress, $\Phi_{xy}^{(el)}$, in the perpendicular plane. This is due to a reversible as well as an irreversible coupling. In addition, there are also planar compressional stationary strains ($\Phi_{yy}^{(el)} = -\Phi_{xx}^{(el)}$) due to reversible couplings.

The transverse directions x and y introduced in the previous paragraphs are not determined without a suitable external force. The system is uniaxial and transversely

isotropic, optically. Of course, the tetrahedral structure has a fourfold (improper) symmetry, only in optical measurements this cannot be detected. However, if one applies the external temperature gradient perpendicularly (e.g., $\nabla_x T$), the transverse isotropy is broken, which can be detected optically. Measuring the ratio $\Phi_{yz}^{(el)}/\Phi_{xz}^{(el)}$ of the induced stationary elastic strains, one gets the orientation of the transverse tetrahedral structure relative to the externally defined x -direction [13].

4 Magnetorheological fluids

4.1 A minimal hydrodynamic model

In the study by Potisk et al. [14], we discuss a simple, minimal model to describe the static and dynamic properties of magnetorheological fluids (MRFs). The main ingredient is transient elasticity (relaxing strain variable) that is induced by an external magnetic field. We model this by assuming the elastic moduli and the relaxation times to be proportional to M_i^2 , where M_i is the magnetization, which is zero in equilibrium and in the absence of an external field. Obviously, this description can only be applied to small and intermediate fields, since for high fields there are saturation effects and the elastic moduli will not grow indefinitely. Important is also magnetostriction that provides a static coupling between elasticity and magnetization. In order to make this magnetorheological model as simple as possible, thermal and concentration effects have been neglected, e.g., disregarding temperature gradients and sedimentation. Isotropy of the material properties is assumed, thereby also eliminating relative rotations as variables.

From the symmetry point of view, this model is like a magnetic, viscoelastic suspension. The most important aspects are not due to symmetry reasons but come from internal material properties, like the tendency of the magnetic particles to form chains (and induce elasticity) in the presence of external fields. Another example for a special internal material property is the field dependence of the viscosity in some magnetic fluids [51]. The influence of this property on the threshold behavior in thermal instabilities has been investigated theoretically in the study by Pérez et al. [7].

The material is assumed to be within two parallel plates with the magnetic field $H_i = \delta_{iz} H_0$ perpendicular to them. First, we have applied this magnetorheological model to external *static shear deformations* (strains) $S_{ij} = S \delta_{iz} \delta_{jx}$. The two forces applied lead to three coupled nonlinear equations for the elastic shear stress, Ψ_{xz} , the magnetization parallel to the field, M_z , and the component M_x perpendicular to the external field (compare Eqs. 29–31 in ref.[14]). Analysis of these equations shows that all three quantities are nonvanishing as a consequence of the magnetostrictive coupling (in this geometry and with two external forces) of M_x , M_z , and Ψ_{xz} in the energy, as well as in the resulting minimizing equations. The resulting elastic shear stress, Ψ_{xz} , is proportional to H_0^2 and initially grows linearly as a function of S , then reaches a maximum (the

“static yield stress”) at a field-independent yield strain, and diminishes beyond. This is the unstable region, where elasticity breaks down and gives way to a liquid-like response. The tilt of the magnetization, characterized by a nonzero M_x , increases with S . If a normal pressure is applied additionally, the system becomes more rigid, and the static yield stress (as well as the appropriate yield strain) increases considerably. These effects are qualitatively in accordance with experiments.

Under *external shear flow*, $v_x = \dot{\gamma}z$ with shear rate $\dot{\gamma}$, one has to consider the full dynamic equations for the magnetization, the elasticity, and flow. The most important cross-coupling is between the magnetization and flow. It comes in two parts, the first is the convection term $\epsilon_{ijk}M_j\omega_k$ for \dot{M}_i in Eq. (23) and the second is a phenomenological contribution to the magnetization current $Y_i^{MR} \sim c_{ijk}^R A_{jk}$. Both contributions are reversible since c_{ijk}^R contains an odd number of M_i factors. The shear part of the latter, c_2^R , is the analog to the flow alignment parameter in nematic liquid crystals and has been given in the context of ferrofluids in the studies by Müller and Liu [52] and Müller et al. [53]. The counter terms, necessary to guarantee zero entropy production, then enter the full stress tensor, σ_{ij} , in addition to the viscous stress due to the external flow and the elastic stress.

For stationary shear flow, where $\dot{\gamma} = \dot{\gamma}_0$ is a constant, the full stress tensor shows, as a function of $\dot{\gamma}_0$, a steep increase up to, what is called the dynamic yield shear stress, beyond which the increase is much slower. The dynamic yield shear stress increases quadratically with the external field H_0 . The behavior at higher $\dot{\gamma}_0$ shows shear thinning and therefore fits better to a Casson or Herschel-Bulkley model [54], rather than to the standard Bingham model.

For oscillating shear flow, $\dot{\gamma} = \dot{\gamma}_0 \cos\omega t$, the linear response of the system is described by a complex shear modulus, $G = G' + iG''$, whose real and imaginary parts describe the reactive (reversible) and dissipative response, respectively. As a function of the frequency, the storage modulus G' increases quadratically until it reaches a constant plateau value. The loss modulus G'' increases linearly up to maximum and decreases until a minimum is reached and finally grows linearly again according to a simple viscous behavior. These features correspond fairly well to experimental findings [55, 56]. The shear modulus $|G|$ can exhibit two plateaus, at low and at intermediate frequencies, related to the two relaxation processes involved, strain relaxation and magnetic relaxation. The Cox-Merz rule, an empirical law relating nonlinear stationary to linear high frequency properties [57, 58], is often fulfilled in polymer dynamics but not in our model of MRFs. The difference is probably due to the fact that the columnar structure is destroyed under large steady shear but not when small amplitude oscillatory shear is applied.

4.2 A two-fluid description

In the minimal model of MRFs, we have used only one velocity field to describe the dynamics of all variables. Similar to the case of suspensions of large particles or in the

dynamics of phase separation, we allow in a two-fluid description [15] macroscopic movements of the magnetic particles relative to the background fluid. Therefore, we introduce two density variables, ρ_1 and ρ_2 (as in a binary mixture), and two momenta $g_i^{(1)} = \rho_1 v_i^{(1)}$ and $g_i^{(2)} = \rho_2 v_i^{(2)}$. Compared to the minimal model, there are now two new variables, the concentration $\phi = \rho_2/\rho$ and the relative velocity $w_i = v_i^{(1)} - v_i^{(2)}$ that come together with the conserved variables, total density $\rho = \rho_1 + \rho_2$ and total momentum $g_i = g_i^{(1)} + g_i^{(2)}$. The concentration is even under T_R and S_I , while the relative velocity is odd under both.

This two-fluid description is different from the case of active polar gels in Section 3.1, where the second velocity is related to a broken symmetry and hence a class B variable. Here, the relative velocity w_i is simply a slowly relaxing variable of class C with the conjugate h_i^w ($\mu_C dC \rightarrow h_i^w dw_i$) and the balance equation $(\partial/\partial t)w_i + Y_i^w = 0$. The conjugate quantity to ϕ is the osmotic pressure Π and the (linearized) dynamic equation reads $\rho_0 (\partial/\partial t)\phi + \nabla_{ij} \phi = 0$ (for the general case, refer the study by Pleiner et al. [15]). The conjugates h_i^w and Π have the same symmetry signatures as w_i and ϕ , respectively. The currents j_i^ϕ and Y_i^w are odd under S_I , while the second law of thermodynamics requires $j_i^{\phi R}$ and Y_i^{wD} to be odd under T_R and $j_i^{\phi D}$ and Y_i^{wR} to be even.

The general problem of any two-fluid theory regarding the form of the transport velocities, e.g., how to generalize Eqs. 18–24, cannot be discussed here but refer the studies by Pleiner and Harden [48, 49]. However, as part of our two-fluid model of MRFs, we choose velocity $v_i^{(1)}$ to transport or convect the variables ρ_1 and $g_i^{(1)}$, while for the magnetization M_i and the strain U_{ij} , we take $v_i^{(2)}$ (and heat is transported by the mean velocity $v_i = g_i/\rho$). In Pleiner et al. [15], we show how this choice fits well into the general scheme without violating any thermodynamic rule, if one requires $v_i^{(c)} = (\rho_1/\rho)v_i^{(2)} + (\rho_2/\rho)v_i^{(1)}$ to be the transport velocity for ϕ and w_i .

In the statics, $w_i \sim h_i^w$ and does not couple to other degrees of freedom (in a linearized description), while the concentration couples to the magnetization and the (trace of the) strain, $\delta \Pi \sim (2\chi_m M_i^0 \delta M_i + \tilde{\chi}_u \delta U_{kk})$, and the counter terms (to make sure ε is a potential) are $h_i^M = 2\chi_m M_i^0 \delta \phi$ and $\Phi_{ij} = 2\tilde{\chi}_u \delta_{ij} \delta \phi$, with $\tilde{\chi}_u = M_{\delta}^2 \chi_u$ in accordance with the similar form of the elastic moduli.

In the dynamics, the relative velocity reversibly couples to the magnetization, $Y_i^{wR} = \xi_{ijk} \nabla_j h_k^M$ and $X_i^{MR} = \xi_{kji} \nabla_j h_k^w$, with ξ_{ijk} containing only odd powers of M_i , as well as irreversibly, $Y_i^{wD} = -\nabla_j (\gamma_{ijk} h_k^M)$ and $X_i^{MD} = \gamma_{kji} \nabla_j h_k^w$, where γ_{ijk} is even in M_i . The relative velocity also couples to flow $Y_i^{D} = -\nabla_j (v_{ijkl}^{(c)} A_{kl})$ and $\sigma_{ij}^D = -v_{ijkl}^{(c)} \nabla_l h_k^w$, where $v_{ijkl}^{(c)}$ is even in M_i . The concentration couples to magnetization, $j_i^{\phi D} = -\alpha_{kij} \nabla_j h_k^M$ and $Y_i^{MD} = -\nabla_k (\alpha_{ijk} \nabla_j \Pi)$, where α_{ijk} is odd in M_i in order to be dissipative.

A direct comparison with the one-fluid model can be made for a stationary shear flow in the parallel plate geometry. Assuming as boundary conditions at the moving

plate for the background flow, $v_x^{(1)} = \Gamma_x$, for the particle phase, $v_i^{(2)} = 0$, and for the strain, $\nabla_z U_{xz} = 0$, one finds a nonzero and asymmetric relative velocity field w_x , whose maximum depends on the external shear strain rate Γ_x in a complicated way. As a result, the flow profile (of $v_x^{(1)}$) in the flow channel is not linear (in contrast to the one-fluid case). In addition, the stress-strain relation, although quite similar to the one-fluid case for small and large Γ_x , differs for intermediate Γ_x values, in particular, the somewhat unphysical overshoot present in the one-fluid case is no longer there. The introduction of a two-fluid description considerably improves the quality of the macroscopic model.

Bridging the gap between MRFs and soft magnetorheological gels, we would like to refer to the recent review of the understanding of the interplay between single particle motion, internal deformation, and matrix properties, in particular, concerning the buckling of chains and the matrix deformations around inclusions [59].

5 Summary and perspective

In this compact review, we have first outlined the approach of macroscopic dynamics based on the use of linear irreversible thermodynamics and the behavior under symmetry operations including inversion, time reversal, and rotations. The variables of interest come in three groups: conservation laws, variables associated with spontaneously broken continuous symmetries, and macroscopic variables, which relax on a sufficiently long time scale to be of hydrodynamic interest.

One field covered is the interaction of nematic order with magnetism. It includes the description of isotropic and uniaxial magnetic elastomers and gels, as well as ferromagnetic nematics, as they have become available experimentally a few years ago. Stimulated by this progress, we also investigated the effect of additional tetrahedral/octupolar order. Another related topic is the study of elasticity with nonnematic order including polar and active polar gels, as well as tetrahedral and ferromagnetic tetrahedral gels.

To examine the effect of larger particles in a carrier fluid, we have presented a minimal one-fluid model of MRFs, the results of which are in accord with a large body of experimental literature. To account for segregation effects between the carrier fluid and the particles, we generalized our approach very recently and gave a two-fluid description of MRFs.

As a perspective, we just mention two classes of systems for which the presented approach will be useful. One is the field of active magnetic gels and elastomers. Quite recently, it has become clear that magnetotactic bacteria fall into this class of systems. Another direction for the near future is the study of fluid- or gel-like systems, which show simultaneously magnetic and electric order. We are thinking, for example, of a system, which is ferroelectric and ferromagnetic simultaneously. Candidates are the

ferromagnetic nematic liquid crystals already existing with a solvent, which contains ferroelectric particles. One might thus obtain a liquid multiferroic system with three types of order: ferromagnetic, ferroelectric, and nematic.

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