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Macroscopic dynamics of uniaxial magnetic gels

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We present the derivation of the macroscopic equations for uniaxial ferrogels. In addition to the usual hydrodynamic variables for gels we introduce the magnetization and the relative rotations between the magnetization and the network as macroscopic variables. The relative rotations introduced here for a system with magnetic degrees of freedom are the analog of the relative rotations introduced by de Gennes in nematic elastomers for rotations of the director with respect to the elastomeric network. These variables give rise to a large number of static as well as dynamic effects due to their coupling to the magnetization, the strain field, and the density of linear momentum. A few of them are discussed for specific geometries, for example, the case of a shear-induced magnetization perpendicular to the preferred direction.

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I. INTRODUCTION

In recent decades gels and especially ferrogels became a promising class of materials for applications in many fields. Ferrogels are chemically cross-linked polymer networks that are generated using a ferrofluid as a component. As was shown in many publications [1–3], there exists a coupling between the elastic and magnetic degrees of freedom allowing one to control the mechanical behavior by applying external magnetic fields. This might lead to many different applications from soft actuators or micromanipulators in technical fields to applications in medicine where they might act as artificial muscles [4] or as carriers for drugs to guarantee controlled drug release. Frequently discussed is the application in hyperthermia due to the heating of magnetic gels in alternating external fields [5]. In our model we will assume that the particles show some kind of interaction with the polymer network although the mechanism is not yet well understood.

Until 2002 interest focused on isotropic ferrogels. The first attempt to generate anisotropic ferrogels was made by Mitsumata *et al.* [6]. They produced gels that contained barium ferrite particles of micrometer size that led to a remnant magnetization without applying an external field. This anisotropy seemed to affect the sound speed. The first anisotropic ferrogels using ferrofluids containing monodomain ferromagnetic particles were produced in 2003 by Zrínyi *et al.* [2] and Collin *et al.* [7], and showed anisotropic features qualitatively and quantitatively, respectively. To produce these uniaxial gels, the cross-linking process was performed in an external magnetic field. In this situation the nanosized ferromagnetic particles form columns and fibers that are larger than the network mesh size [7], because the stabilizing coating of the particles [8] was reduced in its efficiency due to the low *p*H value needed to start the cross-linking process. These chains are fixed in the network, interacting in a way that is so far only partially understood, leading to a frozen-in magnetization which in turn gives rise to several effects in external shear and magnetic fields. In this paper we want to consider this type of gel and we discuss several interesting effects.

To derive the equations describing the macroscopic dynamics of these uniaxial ferrogels we will use the hydrodynamic method. In this method we obtain the hydrodynamic equations by using symmetry and thermodynamic arguments. These equations hold in the long wavelength limit and for sufficiently low frequencies. We use this method because of its generality and its applicability to many different systems in the hydrodynamic regime. But one has to introduce some phenomenological parameters and transport coefficients that cannot be derived by this method. These parameters have to be determined by microscopic models or one has to measure them in experiments.

The method is not restricted to hydrodynamic variables only. In some systems nonhydrodynamic relaxation processes become so slow that they are comparable to macroscopic time scales. Then one must consider these macroscopic variables in the description as well. There is, however, no general rule to decide which processes behave this way. The identification of these macroscopic variables has to be done for each system separately.

In this work we generalize the set of hydrodynamic equations for isotropic ferrogels [9] to these applicable to uniaxial ferrogels. We then discuss some of the effects mediated by couplings between the frozen-in magnetization and the elastic polymeric network. We make predictions for uniaxial magnetic gels, which can be tested experimentally. So far we are not aware of any experimental work investigating the cross-coupling effects discussed here.

II. DERIVATION OF MACROSCOPIC EQUATIONS

A. Hydrodynamic and macroscopic variables

To set up the macroscopic equations for uniaxial ferrogels we start with the identification of the relevant variables [10]. We can separate them into three classes. The first class of variables, also called the hydrodynamic variables, contains those already known from a simple fluid, the mass density ρ, the energy density ε, and the momentum density $\mathbf{g}$. In our case we add another variable, the concentration of ferromagnetic particles c. To the second class belong the variables that are related to spontaneously broken continuous symmetries. In our case we take into account the magnetization **M** and the strain field ϵ_{ij}. The first one is related to the spontaneously broken rotational symmetry while the latter describes the spontaneously broken translational symmetry. We will introduce a unit vector m_i defined by $m_i = M_i/|\mathbf{M}|$ pointing in the direction of magnetization in analogy to the director n_i in nematic liquid crystals. But there is a significant difference. While both are even under parity, the unit vector of magnetization **m** is odd under time reversal. This will permit static as well as dynamic couplings to other variables that are odd under time reversal. We can then define the transverse Kronecker tensor $\delta_{ij}^{\perp} = \delta_{ij} - m_i m_j$ and we have, together with the Levi-Cevità symbol ϵ_{ijk}, three invariants of the system in terms of which the coupling tensors and the transport tensors can be expanded. In a last step we consider a variable first introduced by de Gennes for liquid crystalline elastomers [11], called the relative rotation $\tilde{\Omega}_i$. This variable belongs to the class of slowly relaxing variables and describes the relative rotation between the polymer network and the orientation of the magnetization. It is defined by

$$\tilde{\Omega}_i = \delta m_i - \Omega_i^{\perp} = \delta m_i - \frac{1}{2} m_j (\nabla_i u_j - \nabla_j u_i), \tag{1}$$

where we introduced a vector u_i describing the displacement field of the network and the variation of the orientation of the magnetization δm_j. Since m_i is a unit vector, $\mathbf{m} \cdot \delta \mathbf{m} = 0$. This variable is odd under time reversal and even under parity.

B. Statics and thermodynamics

To get the static properties of our system we formulate the local first law of thermodynamics relating changes in the entropy density σ to changes in the hydrodynamic and macroscopic variables discussed above. We find the Gibbs relation

$$\begin{aligned} d\varepsilon = {} & T\,d\sigma + \mu\,d\rho + \mu_c dc + v_i dg_i + H_i dB_i + h_i'^M dM_i \\ & + \Phi_{ij}^M d(\nabla_j M_i) + \Psi_{ij} d\epsilon_{ij} + W_i d\tilde{\Omega}_i . \end{aligned} \tag{2}$$

In Eq. (2) the thermodynamic quantities temperature T, chemical potential μ, relative chemical potential μ_c, velocity v_i, magnetic field H_i, the magnetic molecular fields $h_i'^M$ and Φ_{ij}^M, the elastic stress Ψ_{ij} and the molecular field W_i are defined as partial derivatives of the energy density with respect to the appropriate variables [10]. If we neglect surface effects and integrate Eq. (2) by parts we can obtain an expression for the Gibbs relation that we want to use throughout the rest of this paper:

$$\begin{aligned} d\varepsilon = {} & T\,d\sigma + \mu\,d\rho + \mu_c dc + v_i dg_i + H_i dB_i + h_i^M dM_i + \Psi_{ij} d\epsilon_{ij} \\ & + W_i d\tilde{\Omega}_i , \end{aligned} \tag{3}$$

where the molecular field h_i^M is given by $h_i^M = h_i'^M - \nabla_j \Phi_{ij}^M$.

To determine the thermodynamic conjugate variables we need an expression for the local energy density. This energy density must be invariant under time reversal as well as under parity and it must be invariant under rigid rotations, rigid translations, and covariant under Galilei transformations. In addition to that this energy density must have a minimum, because there exists an equilibrium state for the gel. Therefore the expression for the energy density needs to be convex. Taking into account these symmetry arguments we write down an expansion for the generalized energy density up to second order in the variables that describe deviations out of that equilibrium and considering several interesting third order terms including magnetostriction:

$$\begin{aligned} \varepsilon = {} & \frac{1}{2} B_i B_i + \frac{\alpha}{2} M_i M_i - M_i B_i - \frac{1}{2} \gamma_{ijkl} M_i M_j \epsilon_{kl} + \frac{1}{2} \mu_{ijkl} \epsilon_{ij} \epsilon_{kl} \\ & + \frac{1}{2} K_{ijkl} (\nabla_i M_j)(\nabla_k M_l) + \frac{1}{2} D_1 \tilde{\Omega}_i \tilde{\Omega}_i + D_2 (m_j \delta_{ik}^{\perp} + m_k \delta_{ij}^{\perp}) \\ & \times \tilde{\Omega}_i \epsilon_{jk} + \sigma_{ijk}^{\sigma} (\nabla_i M_j)(\nabla_k \delta\sigma) + \sigma_{ijk}^{\rho} (\nabla_i M_j)(\nabla_k \delta\rho) \\ & + \sigma_{ijk}^{c} (\nabla_i M_j)(\nabla_k \delta c) + \epsilon_{ij} (\chi_{ij}^{\sigma} \delta\sigma + \chi_{ij}^{\rho} \delta\rho + \chi_{ij}^{c} \delta c) + c_{\rho\rho} (\delta\rho)^2 \\ & + c_{\sigma\sigma} (\delta\sigma)^2 + c_{cc} (\delta c)^2 + c_{\rho c} (\delta\rho)(\delta c) + c_{\rho\sigma} (\delta\rho)(\delta\sigma) \\ & + c_{\sigma c} (\delta\sigma)(\delta c) + \frac{1}{2\rho} g_i g_i + c_{ijk} g_i (\nabla_j M_k) + a_c M^2 \delta c \\ & + a_{\sigma} M^2 \delta\sigma + a_{\rho} M^2 \delta\rho . \end{aligned} \tag{4}$$

Apart from the energy density of a normal fluid binary mixture, Eq. (4) contains the magnetic energy as well as the elastic energy. It is worth mentioning that we get—because of the negative time reversal property of the magnetization—a coupling between the curl of **M** and the momentum density mediated by the tensor c_{ijk}, which takes the form $c_{ijk} = (c_{\parallel} m_i m_l + c_{\perp} \delta_{il}^{\perp}) \epsilon_{ljk}$. This kind of coupling is very similar to one of the couplings appearing in superfluid ^{3}He-*A* first introduced by Graham [12]. In this system one defines an axial vector $\boldsymbol{l}$ parallel to the direction of the net orbital momentum of the helium pairs. This vector does not commute with the total angular momentum vector and therefore this variable breaks the continuous rotational symmetry spontaneously, similarly to the magnetization in our system. The source-free part of the momentum density of ^{3}He-*A* is proportional to the curl of this vector $\boldsymbol{l}$ while the proportionality is given by $\hbar/2m$ and a coupling tensor c_{ij}.

Because we discuss a uniaxial system, the tensors will have more independent constants than in the isotropic case of Jarkova *et al.* [9]. The tensor of the elastic energy, for example, now has five independent constants instead of only two and takes the form

$$\begin{aligned} \mu_{ijkl} = {} & \mu_1 \delta_{ij}^{\perp} \delta_{kl}^{\perp} + \mu_2 \left\{ \left(\delta_{ik}^{\perp} \delta_{jl}^{\perp} - \tfrac{1}{2} \delta_{ij}^{\perp} \delta_{kl}^{\perp} \right) + \left(\delta_{il}^{\perp} \delta_{jk}^{\perp} - \tfrac{1}{2} \delta_{ij}^{\perp} \delta_{kl}^{\perp} \right) \right\} \\ & + \mu_3 m_i m_j m_k m_l + \mu_4 (m_i m_j \delta_{kl}^{\perp} + m_k m_l \delta_{ij}^{\perp}) \\ & + \mu_5 \{ m_i m_k \delta_{jl}^{\perp} + m_i m_l \delta_{jk}^{\perp} + m_j m_k \delta_{il}^{\perp} + m_j m_l \delta_{ik}^{\perp} \} \end{aligned} \tag{5}$$

while the magnetostrictive tensor will have six independent constants

$$\begin{aligned} \gamma_{ijkl} = {} & \gamma_1 \delta_{ij}^{\perp} \delta_{kl}^{\perp} + \gamma_2 \left\{ \left(\delta_{ik}^{\perp} \delta_{jl}^{\perp} - \tfrac{1}{2} \delta_{ij}^{\perp} \delta_{kl}^{\perp} \right) + \left(\delta_{il}^{\perp} \delta_{jk}^{\perp} - \tfrac{1}{2} \delta_{ij}^{\perp} \delta_{kl}^{\perp} \right) \right\} \\ & + \gamma_3 m_i m_j m_k m_l + \gamma_4 m_i m_j \delta_{kl}^{\perp} + \gamma_5 m_k m_l \delta_{ij}^{\perp} \\ & + \gamma_6 \{ m_i m_k \delta_{jl}^{\perp} + m_i m_l \delta_{jk}^{\perp} + m_j m_k \delta_{il}^{\perp} + m_j m_l \delta_{ik}^{\perp} \} . \end{aligned} \tag{6}$$

The tensor K_{ijkl} describes contributions to the local energy density due to spatial changes of the direction of the magnetization as well as of its magnitude. We get six independent constants for this coupling,

$$K_{ijkl} = \frac{1}{2}K_1(\delta_{ij}^{\perp}\delta_{kl}^{\perp} + \delta_{il}^{\perp}\delta_{jk}^{\perp}) + K_2 m_p \epsilon_{pij} m_q \epsilon_{qkl} + K_3 m_k m_i \delta_{lj}^{\perp} + K_4 m_i m_j m_k m_l + K_5 m_j m_l \delta_{ik}^{\perp} + \frac{1}{4}K_6(m_i m_l \delta_{kj}^{\perp} + m_j m_k \delta_{il}^{\perp} + m_i m_j \delta_{kl}^{\perp} + m_k m_l \delta_{ij}^{\perp}). \tag{7}$$

There are two more contributions to the energy density due to the coupling between the strain field and the variables associated with the relative rotations. One is proportional to D_1 and the other proportional to D_2. One can interpret these coefficients as a measure for the coupling strength of the magnetic particles to the polymer network, although the microscopic mechanism of this interaction is not precisely understood so far.

Now we are left with the couplings between the scalars ρ, c, and σ and the strain field as well as with the coupling between the gradient of the scalars and the gradient of the magnetization. The tensors take the following form, respectively:

$$\chi_{ij}^{\xi} = \chi_{\parallel}^{\xi} m_i m_j + \chi_{\perp}^{\xi}\delta_{ij}^{\perp}, \tag{8}$$

$$\sigma_{ijk}^{\xi} = \sigma_1^{\xi} m_i m_j m_k + \sigma_2^{\xi} m_j \delta_{ik}^{\perp} + \sigma_3^{\xi}(m_i \delta_{jk}^{\perp} + m_k \delta_{ij}^{\perp}), \tag{9}$$

where ξ can be either ρ, σ, or c.

We now give the expressions for the conjugated variables in terms of the hydrodynamic and macroscopic variables. They are defined as partial derivatives with respect to the appropriate variable, while all the other variables are kept constant, denoted by ellipses at the parentheses in the following. We obtain

$$v_i = \left(\frac{\partial\varepsilon}{\partial g_i}\right)_{\ldots} = \frac{1}{\rho}g_i + c_{ijk}(\nabla_j M_k), \tag{10}$$

$$H_i = \left(\frac{\partial\varepsilon}{\partial B_i}\right)_{\ldots} = B_i - M_i, \tag{11}$$

$$h_i'^{M} = \left(\frac{\partial\varepsilon}{\partial M_i}\right)_{\ldots} = \alpha M_i - B_i - \gamma_{ijkl} M_j \epsilon_{kl} + 2a_c M_i \delta c + 2a_{\sigma} M_i \delta\sigma + 2a_{\rho} M_i \delta\rho, \tag{12}$$

$$\Phi_{ij}^{M} = \left(\frac{\partial\varepsilon}{\partial(\nabla_j M_i)}\right)_{\ldots} = K_{ijkl}(\nabla_k M_l) + \sigma_{ijk}(\nabla_k \delta\xi) + c_{ijk} g_k, \tag{13}$$

$$\Psi_{ij} = \left(\frac{\partial\varepsilon}{\partial\epsilon_{ij}}\right)_{\ldots} = -\frac{1}{2}\gamma_{ijkl} M_k M_l + \mu_{ijkl}\epsilon_{kl} + D_2(m_j\delta_{ik}^{\perp} + m_i\delta_{kj}^{\perp}) \times \tilde{\Omega}_k + \chi_{ij}^{\sigma}\delta\sigma + \chi_{ij}^{\rho}\delta\rho + \chi_{ij}^{c}\delta c, \tag{14}$$

$$W_i = \left(\frac{\partial\varepsilon}{\partial\tilde{\Omega}_i}\right)_{\ldots} = D_1\tilde{\Omega}_i + D_2(m_j\delta_{ik}^{\perp} + m_k\delta_{ij}^{\perp})\epsilon_{jk}, \tag{15}$$

$$\delta T = \left(\frac{\partial\varepsilon}{\partial\delta\sigma}\right)_{\ldots} = \chi_{ij}^{\sigma}\epsilon_{ij} + 2c_{\sigma\sigma}\delta\sigma + c_{\rho\sigma}\delta\rho + c_{\sigma c}\delta c + a_{\sigma}M^2, \tag{16}$$

$$\delta\mu = \left(\frac{\partial\varepsilon}{\partial\delta\rho}\right)_{\ldots} = \chi_{ij}^{\rho}\epsilon_{ij} + 2c_{\rho\rho}\delta\rho + c_{\rho c}\delta c + c_{\rho\sigma}\delta\sigma + a_{\rho}M^2, \tag{17}$$

$$\delta\mu_c = \left(\frac{\partial\varepsilon}{\partial\delta c}\right)_{\ldots} = \chi_{ij}^{c}\epsilon_{ij} + 2c_{cc}\delta c + c_{c\rho}\delta\rho + c_{c\sigma}\delta\sigma + a_c M^2. \tag{18}$$

We used integration by parts to obtain expression (3) for the local energy density, where the new molecular field h_i^M was given by $h_i^M = h_i'^{M} - \nabla_j\Phi_{ij}^M$. If we use Eqs. (12) and (13) we find

$$h_i^M = \alpha M_i - B_i - \gamma_{ijkl} M_j \epsilon_{kl} + 2a_c M_i \delta c + 2a_{\sigma} M_i \delta\sigma + 2a_{\rho} M_i \delta\rho - (\nabla_j K_{ijkl})(\nabla_k M_l) - K_{ijkl}(\nabla_j\nabla_k M_l) - (\nabla_j\sigma_{ijk})(\nabla_k\delta\xi) - \sigma_{ijk}(\nabla_j\nabla_k\delta\xi) - (\nabla_j c_{ijk})g_k - c_{ijk}(\nabla_j g_k). \tag{19}$$

C. Dynamic equations

To determine the dynamics of the variables we take into account that the first class of our set of variables contains conserved quantities that obey a local conservation law while the dynamics of the other two classes of variables can be described by a simple balance equation where the counterterm to the temporal change of the quantity is called a quasicurrent. As a set of dynamical equations we get

$$\partial_t\rho + \nabla_i g_i = 0, \tag{20}$$

$$\partial_t\sigma + \nabla_i(\sigma v_i) + \nabla_i j_i^{\sigma} = \frac{R}{T}, \tag{21}$$

$$\rho\partial_t c + (\rho v_i\nabla_i)c + \nabla_i j_i^c = 0, \tag{22}$$

$$\partial_t g_i + \nabla_j\{v_j g_i + \delta_{ij}[p + \mathbf{B}\cdot\mathbf{H}] + \sigma_{ij}^{th} + \sigma_{ij}\} = 0, \tag{23}$$

$$\partial_t M_i + (v_j\nabla_j)M_i + (\mathbf{M}\times\boldsymbol{\omega})_i + X_i = 0, \tag{24}$$

$$\partial_t\epsilon_{ij} + (v_k\nabla_k)\epsilon_{ij} + Y_{ij} = 0, \tag{25}$$

$$\partial_t\tilde{\Omega}_i + (v_k\nabla_k)\tilde{\Omega}_i + Z_i = 0, \tag{26}$$

where we introduced the vorticity $\omega_i = 1/2\epsilon_{ijk}\nabla_j v_k$ and

$$\sigma_{ij}^{th} = -\frac{1}{2}(B_i H_j + B_j H_i) + \frac{1}{2}(\Psi_{jk}\epsilon_{ki} + \Psi_{ik}\epsilon_{kj}). \tag{27}$$

In Eq. (27) we implemented the requirement that the energy density should be invariant under rigid rotations [10].

The pressure p in Eq. (23) is given by $\partial E/\partial V$ and reads for our system

$$p = -\varepsilon + \mu\rho + T\sigma + \mathbf{v}\cdot\mathbf{g}. \tag{28}$$

In the equation for the entropy density (21) we introduced R, the dissipation function which represents the entropy production of the system. Due to the second law of thermodynamics R must satisfy $R \geqslant 0$. For reversible processes this dissipation function is equal to zero while for irreversible processes it must be positive. In the following we will split the currents and quasicurrents into reversible parts (denoted with a superscript R) and irreversible parts (denoted with a superscript D).

D. Reversible dynamics

If we again make use of the symmetry arguments mentioned above and use Onsager's relations we obtain the following expressions for the reversible currents up to linear order in the thermodynamic forces:

$$g_i = \rho v_i - \rho c_{ijk}(\nabla_j M_k), \tag{29}$$

$$j_i^{\sigma R} = -\kappa_{ij}^R \nabla_j T - D_{ij}^{TR}\nabla_j \mu_c + \xi_{ij}^{TR}\nabla_l \Psi_{jl}, \tag{30}$$

$$j_i^{cR} = -D_{ij}^R \nabla_j \mu_c + D_{ij}^{TR}\nabla_j T + \xi_{ij}^{cR}\nabla_l \Psi_{lj}, \tag{31}$$

$$\sigma_{ij}^R = -\Psi_{ij} - c_{ijk}^{RJ} h_k^M - \nu_{ijkl}^R A_{kl} + \xi_{ijk}^{\sigma R} W_k, \tag{32}$$

$$\begin{aligned} Y_{ij}^R = {} & -A_{ij} + \xi_{ijk}^{YR} W_k + \frac{1}{2}\lambda^M[\nabla_i(\boldsymbol{\nabla}\times\mathbf{h}^M)_j + \nabla_j(\boldsymbol{\nabla}\times\mathbf{h}^M)_i] \\ & - \frac{1}{2}[\nabla_i\{\xi_{jk}^R \nabla_l \Psi_{kl} + \xi_{jk}^{TR}\nabla_k T + \xi_{jk}^{cR}\nabla_k \mu_c\} + (i \leftrightarrow j)], \end{aligned} \tag{33}$$

$$X_i^R = b_{ij}^R h_j^M + \lambda^M \epsilon_{ijk}\nabla_j(\nabla_l \Psi_{kl}) - c_{jki}^{RJ} A_{jk} + \xi_{ij}^{XR} W_j, \tag{34}$$

$$Z_i^R = \tau_{ij}^R W_j - \xi_{ij}^{XR} h_j^M - \xi_{kli}^{\sigma R} A_{kl} - \xi_{kli}^{YR}\Psi_{kl}. \tag{35}$$

Compared to the reversible currents and quasicurrents of isotropic ferrogels we have the additional quasicurrent of relative rotations with its counterterms in X_i^R, σ_{ij}^R, and Y_{ij}^R. These terms describe the dynamic coupling of relative rotations to the magnetization, the momentum density, and the network, respectively. The first coupling—mediated by the tensor ξ_{ij}^{XR}—is an additional term that exists neither in nematic liquid crystalline elastomers [13] nor in superfluid ^{3}He-A, while the second coupling—$\xi_{ijk}^{\sigma R}$—already appeared in nematic liquid crystalline elastomers. The third coupling—ξ_{ijk}^{YR}—is also an additional one and will be discussed in detail in the following section. The additional term in the momentum density already appeared in superfluid ^{3}He-A and we will discuss one of the consequences of this coupling later. The tensors in the currents for the entropy density and the concentration all have to be odd under time reversal, because the currents have to be odd under time reversal. They are all of the form

$$\alpha_{ij}^R = \alpha^R \epsilon_{ijk} m_k. \tag{36}$$

Furthermore, we find for the coupling terms in the stress tensor

$$c_{ijk}^{RJ} = c_1^{RJ}(m_i \delta_{jk}^\perp + m_j \delta_{ik}^\perp) + c_2^{RJ} m_k \delta_{ij}^\perp + c_3^{RJ} m_i m_j m_k, \tag{37}$$

$$\begin{aligned} \nu_{ijkl}^R = {} & \nu_1^R\{\epsilon_{ikp}\delta_{jl}^\perp + \epsilon_{jlp}\delta_{ik}^\perp + \epsilon_{ilp}\delta_{jk}^\perp + \epsilon_{jkp}\delta_{il}^\perp\} m_p \\ & + \nu_2^R\{\epsilon_{ikp} m_j m_l + \epsilon_{jlp} m_i m_k + \epsilon_{ilp} m_j m_k + \epsilon_{jkp} m_i m_l\} m_p, \end{aligned} \tag{38}$$

$$\xi_{ijk}^{\sigma R} = \xi^{\sigma R}(m_i \delta_{jk}^\perp + m_j \delta_{ik}^\perp). \tag{39}$$

The coupling terms in the quasicurrent for the relative rotations are

$$\xi_{ij}^{XR} = \xi^{XR}\epsilon_{ijk} m_k, \tag{40}$$

$$\xi_{kli}^{\sigma R} = \xi^{\sigma R}(m_k \delta_{li}^\perp + m_l \delta_{ki}^\perp), \tag{41}$$

$$\tau_{ij}^R = \tau^R \epsilon_{ijk} m_k, \tag{42}$$

$$\xi_{kli}^{YR} = \xi^{YR}(m_k \epsilon_{lip} + m_l \epsilon_{kip}) m_p. \tag{43}$$

We are now left with the tensor coupling the molecular field h_i^M to the magnetization quasicurrent which takes the form

$$b_{ij}^R = b^R \epsilon_{ijk} m_k. \tag{44}$$

E. Irreversible dynamics and entropy production

We can use the dissipation function R as a Liapunov functional to derive the irreversible currents and quasicurrents. One can expand the function R (R/T is the amount of entropy produced within a unit volume per unit time) into the thermodynamic forces using the same symmetry arguments as in the case of the energy density. We obtain

$$\begin{aligned} R = {} & \frac{1}{2}\kappa_{ij}(\nabla_i T)(\nabla_j T) + D_{ij}^T(\nabla_i T)(\nabla_j \mu_c) + \xi_{ij}^T(\nabla_i T)(\nabla_k \Psi_{jk}) \\ & + \frac{1}{2}D_{ij}(\nabla_i \mu_c)(\nabla_j \mu_c) + \xi_{ij}^c(\nabla_i \mu_c)(\nabla_k \Psi_{jk}) + \frac{1}{2}\nu_{ijkl} A_{ij} A_{kl} \\ & + \xi_{ijk}^\sigma A_{ij} W_k + c_{ijk}^J A_{ij} h_k^M + \frac{1}{2}\xi_{ij}(\nabla_k \Psi_{ik})(\nabla_l \Psi_{jl}) + \frac{1}{2} b_{ij} h_i^M h_j^M \\ & + \frac{1}{2}\tau_{ij} W_i W_j + \xi_{ij}^X W_i h_j^M, \end{aligned} \tag{45}$$

where we have again introduced some tensors. The tensors κ_{ij}, D_{ij}^T, ξ_{ij}^T, D_{ij}, ξ_{ij}^c, ξ_{ij}, and b_{ij} take the form

$$\alpha_{ij} = \alpha_\parallel m_i m_j + \alpha_\perp \delta_{ij}^\perp \tag{46}$$

while the tensors τ_{ij} and ξ_{ij}^X read

$$a_{ij} = a\,\delta_{ij}^\perp. \tag{47}$$

This is due to the fact that only the parts of the relative rotations perpendicular to the preferred direction contribute to the entropy production [13]. For the viscosity tensor ν_{ijkl} we obtain

$$\begin{aligned} \nu_{ijkl} = {} & \nu_1 \delta_{ij}^\perp \delta_{kl}^\perp + \nu_2\left\{\left(\delta_{ik}^\perp \delta_{jl}^\perp - \frac{1}{2}\delta_{ij}^\perp \delta_{kl}^\perp\right) + \left(\delta_{il}^\perp \delta_{jk}^\perp - \frac{1}{2}\delta_{ij}^\perp \delta_{kl}^\perp\right)\right\} \\ & + \nu_3 m_i m_j m_k m_l + \nu_4(m_i m_j \delta_{kl}^\perp + m_k m_l \delta_{ij}^\perp) \\ & + \nu_5(m_i m_k \delta_{jl}^\perp + m_i m_l \delta_{jk}^\perp + m_j m_k \delta_{il}^\perp + m_j m_l \delta_{ik}^\perp). \end{aligned} \tag{48}$$

We are left with the tensors ξ^{σ}_{ijk} and c^{J}_{ijk}, which take the form

$$a_{ijk} = a(m_i\epsilon_{jkl} + m_j\epsilon_{ikl})m_l. \tag{49}$$

To obtain the dissipative parts of the currents and quasicurrents we take the partial derivatives with respect to the appropriate thermodynamic force,

$$j_i^{\sigma D} = -\left(\frac{\partial R}{\partial(\nabla_i T)}\right)_{\ldots} = -\kappa_{ij}(\nabla_j T) - D^T_{ij}(\nabla_j \mu_c) - \xi^T_{ij}(\nabla_k \Psi_{jk}), \tag{50}$$

$$j_i^{cD} = -\left(\frac{\partial R}{\partial(\nabla_j \mu_c)}\right)_{\ldots} = -D_{ij}(\nabla_j \mu_c) - D^T_{ij}(\nabla_j T) - \xi^c_{ij}(\nabla_k \Psi_{jk}), \tag{51}$$

$$\sigma^D_{ij} = -\left(\frac{\partial R}{\partial(\nabla_j v_i)}\right)_{\ldots} = -\nu_{ijkl}A_{kl} - \xi^{\sigma}_{ijk}W_k - c^J_{ijk}h^M_k, \tag{52}$$

$$\begin{aligned} Y^D_{ij} = \left(\frac{\partial R}{\partial \Psi_{ij}}\right)_{\ldots} &= -\frac{1}{2}\{\nabla_i(\xi_{jn}\nabla_l\Psi_{nl} + \xi^T_{jn}\nabla_n T + \xi^c_{jn}\nabla_n\mu_c)\} \\ &\quad - \frac{1}{2}\{\nabla_j(\xi_{in}\nabla_l\Psi_{nl} + \xi^T_{in}\nabla_n T + \xi^c_{in}\nabla_n\mu_c)\}, \end{aligned} \tag{53}$$

$$Z^D_i = \left(\frac{\partial R}{\partial W_i}\right)_{\ldots} = \tau_{ij}W_j + \xi^X_{ij}h^M_j + \xi^{\sigma}_{kli}A_{kl}, \tag{54}$$

$$X^D_i = \left(\frac{\partial R}{\partial h^M_i}\right)_{\ldots} = b_{ij}h^M_j + \xi^X_{ij}W_j + c^J_{kli}A_{kl}. \tag{55}$$

III. SOME SIMPLE SOLUTIONS

In this section we discuss some experimental setups that could reveal some of the unusual cross-coupling effects of this class of materials.

A. Shear-induced magnetization

Our system differs qualitatively from the isotropic ferrogels by the macroscopic variables associated with relative rotations. These variables describe, as already mentioned, the relative rotations between the orientation of the magnetization and the polymer network. In this section we discuss an effect associated with these variables. We apply a constant shear flow and determine the change of magnetization. We assume that the direction of the frozen-in magnetization in the uniaxial ferrogel is parallel to the x direction while the shear is applied in the x-y plane as sketched in Fig. 1. Furthermore we assume spatial homogeneity. In this case the dynamic equations for the momentum density and the scalars ρ, σ, and c are satisfied automatically. Contributions due to magnetostriction effects are neglected. These effects are of higher order in the variables [cf. Eqs. (19) and (14)] while we focus on linear effects. These assumptions reduce the set of dynamic equations to

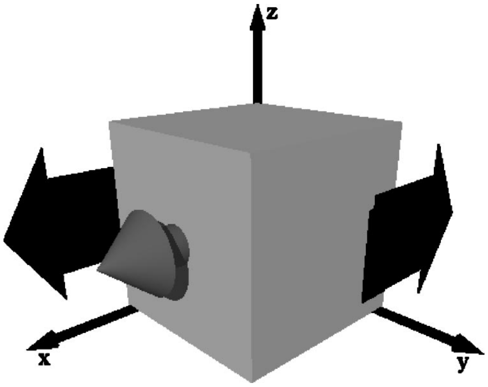

FIG. 1. Sketch of experimental setup. The conelike arrow represents the magnetization of the uniaxial gel while the flat arrows represent the external force.

$$\partial_t M_i + X_i = 0, \tag{56}$$

$$\partial_t \tilde{\Omega}_i + Z_i = 0, \tag{57}$$

$$\partial_t \epsilon_{ij} + Y_{ij} = 0. \tag{58}$$

Now we need to find the relevant expressions for the quasicurrents. In the quasicurrent for the magnetization we can discard the term $\lambda^M\epsilon_{ijk}(\nabla_l\Psi_{kl})$, because this term is of first order in the derivatives and does not contribute in a homogeneous system

$$X^R_i = b^R_{ij}h^M_j + \xi^{XR}_{ij}W_j - c^{RJ}_{jki}A_{jk}, \tag{59}$$

$$X^D_i = b_{ij}h^M_j + \xi^X_{ij}W_j + c^J_{kli}A_{kl}. \tag{60}$$

The same arguments hold for the quasicurrents of the relative rotations and of the strain field. Therefore we obtain

$$Z^R_i = \tau^R_{ij}W_j - \xi^{XR}_{ij}h^M_j - \xi^{YR}_{kli}\Psi_{kl} - \xi^{\sigma R}_{kli}A_{kl}, \tag{61}$$

$$Z^D_i = \tau_{ij}W_j + \xi^X_{ij}h^M_j + \xi^{\sigma}_{kli}A_{kl}, \tag{62}$$

$$Y^R_{ij} = \xi^{YR}_{ijk}W_k - A_{ij}, \tag{63}$$

$$Y^D_{ij} = 0. \tag{64}$$

To obtain a closed set of equations for the macroscopic variables, we substitute for the conjugate variables the expressions found in Sec. II B. Again we discard contributions due to magnetostriction and inhomogeneous contributions. Furthermore, we apply an external force, which is in our case a constant shear flow. We take the simple shear S_{kl} to be in the x-y plane,

$$S_{kl} = \nabla_k v_l \delta_{ky}\delta_{lx}. \tag{65}$$

Therefore we obtain

$$\Psi_{kl} = \mu_{klmn}\epsilon_{mn} + D_2(m_k\delta^{\perp}_{lm} + m_l\delta^{\perp}_{km})\tilde{\Omega}_m, \tag{66}$$

$$W_k = D_1\tilde{\Omega}_k + D_2(m_m\delta^{\perp}_{kn} + m_n\delta^{\perp}_{km})\epsilon_{mn}, \tag{67}$$

$$h_j^M = \alpha \delta M_j - \delta B_j = \left(\alpha - \frac{1}{\chi_0} \right) \delta M_j . \tag{68}$$

In the last expression we used the fact that in the case of a small frozen-in magnetization and no external magnetic fields the magnetic flux density **B** is only due to the intrinsic magnetization and that it can be expressed as $\mathbf{B}=\chi_0\mathbf{M}$. We will use α' as an abbreviation in the following,

$$\alpha' = \alpha - \frac{1}{\chi_0} . \tag{69}$$

We do not apply an external magnetic field. Therefore we can assume that the magnitude of the magnetization is not changed but only its direction. We can write

$$\mathbf{M} = M_0 (\mathbf{m} + \delta \mathbf{m}). \tag{70}$$

If we use the material tensors in our specific geometry we can derive the following set of equations for the different components of each macroscopic variable:

$$0 = \xi^{YR} (D_1 \tilde{\Omega}_z + 2 D_2 \epsilon_{xz}) - \frac{1}{2} A_{xy}^{ext} , \tag{71}$$

$$0 = - \xi^{YR} (D_1 \tilde{\Omega}_y + 2 D_2 \epsilon_{xy}) , \tag{72}$$

$$\begin{aligned} 0 = {} & b^R \alpha' M_0 \delta m_z + \xi^{XR} (D_1 \tilde{\Omega}_z + 2 D_2 \epsilon_{xz}) - c_1^{RJ} A_{xy}^{ext} \\ & + b_\perp \alpha' M_0 \delta m_y + \xi^X (D_1 \tilde{\Omega}_y + 2 D_2 \epsilon_{xy}) , \end{aligned} \tag{73}$$

$$\begin{aligned} 0 = {} & - b^R \alpha' M_0 \delta m_y - \xi^{XR} (D_1 \tilde{\Omega}_y + 2 D_2 \epsilon_{xy}) + b_\perp \alpha' M_0 \delta m_z \\ & + \xi^X (D_1 \tilde{\Omega}_z + 2 D_2 \epsilon_{xz}) + c^J A_{xy}^{ext} , \end{aligned} \tag{74}$$

$$\begin{aligned} 0 = {} & \tau^R (D_1 \tilde{\Omega}_z + 2 D_2 \epsilon_{xz}) - \xi^{XR} \alpha' M_0 \delta m_z \\ & + 2 \xi^{YR} (2 \mu_5 \epsilon_{xz} + D_2 \tilde{\Omega}_z) - \xi^{\sigma R} A_{xy}^{ext} + \tau (D_1 \tilde{\Omega}_y + 2 D_2 \epsilon_{xy}) \\ & + \xi^X \alpha' M_0 \delta m_y , \end{aligned} \tag{75}$$

$$\begin{aligned} 0 = {} & - \tau^R (D_1 \tilde{\Omega}_y + 2 D_2 \epsilon_{xy}) + \xi^{XR} \alpha' M_0 \delta m_y \\ & - 2 \xi^{YR} (2 \mu_5 \epsilon_{xy} + D_2 \tilde{\Omega}_y) + \tau (D_1 \tilde{\Omega}_z + 2 D_2 \epsilon_{xz}) \\ & + \xi^X \alpha' M_0 \delta m_z + \xi^\sigma A_{xy}^{ext} . \end{aligned} \tag{76}$$

From the first and second equations we find a relation between the components of relative rotations and the strain field, so that we can reduce this system to one with four equations for four variables, which is now shown in matrix form:

$$\begin{pmatrix} -\dfrac{Z_1}{N_1} & -\dfrac{Z_2}{N_1} & 0 & 0 \\ \dfrac{Z_2}{N_2} & -\dfrac{Z_1}{N_2} & 0 & 0 \\ -\dfrac{D_1 Z_3}{N_3} & \dfrac{D_1 Z_4}{N_3} & 0 & \dfrac{Z_5}{N_3} \\ -\dfrac{Z_4}{N_4} & -\dfrac{Z_3}{N_4} & \dfrac{Z_5}{D_1 N_4} & 0 \end{pmatrix} \times \begin{pmatrix} \delta m_y \\ \delta m_z \\ \epsilon_{xy} \\ \epsilon_{xz} \end{pmatrix} = A_{xy}^{ext} \begin{pmatrix} 1 \\ 1 \\ 1 \\ 1 \end{pmatrix} \tag{77}$$

with $Z_1=2\xi^{YR} b_\perp \alpha' M_0$, $Z_2=Z_1 b^R / b_\perp$, $Z_3=2\xi^{YR}\xi^X \alpha' M_0$, $Z_4=Z_3 \xi^{XR}/\xi^X$, $Z_5=8(\xi^{YR})^2(D_1\mu_5-D_2^2)$, $N_1=\xi^{XR}-2\xi^{YR}c_1^{RJ}$, $N_2=\xi^X+2\xi^{YR}c^J$, $N_3=2\xi^{YR}D_2+\tau^R D_1-2\xi^{YR}D_1\xi^{\sigma R}$, and $N_4=\tau+2\xi^{YR}\xi^\sigma$.

The solution takes the form

$$\delta m_y = - \frac{- b^R (\xi^X + 2 \xi^{YR} c^J) + b_\perp (\xi^{XR} - 2 \xi^{YR} c_1^{RJ})}{2 \xi^{YR} \alpha' M_0 (b_\perp^2 + b^{R2})} A_{xy}^{ext} , \tag{78}$$

$$\delta m_z = - \frac{b_\perp (\xi^X + 2 \xi^{YR} c^J) + b^R (\xi^{XR} - 2 \xi^{YR} c_1^{RJ})}{2 \xi^{YR} \alpha' M_0 (b_\perp^2 + b^{R2})} A_{xy}^{ext} . \tag{79}$$

For this experimental setup we thus predict a rotation of the magnetization out of the shear plane as well as out of the *x*-*z* plane, which is proportional to the applied external force as can be seen from Eqs. (78) and (79). This effect is due to the variables associated with relative rotations, because all contributions are proportional to either ξ^{XR}, ξ^X, or ξ^{YR}, which represent the dynamical coupling of relative rotations to the magnetization and the strain field, respectively. The change of the direction of the magnetization should be easily observable in the laboratory since Hall probes can sensitively measure magnetic fields.

B. The influence of $(\nabla \times \mathbf{M})$ on the mass current

As pointed out in Secs. II B and II D, there is an additional contribution to the momentum density due to the coupling of the curl of the magnetization to the momentum density via the tensor c_{ijk} (29). Here we want to discuss consequences of this unusual coupling. It is useful to discuss the Fourier transform of the energy density, where we focus on those terms which contain the momentum density and gradients of the magnetization:

$$\begin{aligned} \varepsilon_{\mathbf{k}} = \int & \left\{ \frac{1}{2\rho} g_i g_i + c_{ijk} g_i (\nabla_j M_k) + \frac{1}{2} K_{ijkl} (\nabla_i M_j)(\nabla_k M_l) \right\} \\ & \times e^{i\mathbf{k}\cdot\mathbf{r}} d^3 r . \end{aligned} \tag{80}$$

Since we do not consider an external magnetic field in the following, we can assume that the magnitude of the magnetization is constant. In this case the coupling tensor K_{ijkl} takes a form similar to the known Frank elastic tensor,

$$K_{ijkl} = \frac{1}{2}K_1(\delta^{\perp}_{ij}\delta^{\perp}_{kl} + \delta^{\perp}_{il}\delta^{\perp}_{jk}) + K_2 m_p \epsilon_{pij} m_q \epsilon_{qkl} + K_3 m_k m_i \delta^{\perp}_{lj}, \tag{81}$$

while the tensor c_{ijk} has the form discussed in Sec. II B,

$$c_{ijk} = (c_{\parallel} m_i m_l + c_{\perp}\delta^{\perp}_{il})\epsilon_{ljk}. \tag{82}$$

We introduce a new coordinate system in analogy to the one introduced by Brand *et al.* [14] for superfluid ^{3}He-A,

$$\hat{\mathbf{e}}_1 = \frac{\mathbf{k} - (\mathbf{k}\cdot\mathbf{m})\mathbf{m}}{|\mathbf{k} - (\mathbf{k}\cdot\mathbf{m})\mathbf{m}|}, \tag{83}$$

$$\hat{\mathbf{e}}_2 = \mathbf{m} \times \hat{\mathbf{e}}_1, \tag{84}$$

$$\hat{\mathbf{e}}_3 = \mathbf{m}. \tag{85}$$

One can expand the momentum density $\mathbf{g}$ and the change of the direction of the magnetization $\delta\mathbf{m}$ with respect to this basis:

$$\mathbf{g} = g_1\hat{\mathbf{e}}_1 + g_2\hat{\mathbf{e}}_2 + g_3\hat{\mathbf{e}}_3, \tag{86}$$

$$\delta\mathbf{m} = \delta m_1\hat{\mathbf{e}}_1 + \delta m_2\hat{\mathbf{e}}_2. \tag{87}$$

It is worth mentioning that the individual components have different properties under time reversal and parity,

$$\epsilon^{T}_{g_1} = -1, \quad \epsilon^{P}_{g_1} = +1, \tag{88}$$

$$\epsilon^{T}_{g_2} = +1, \quad \epsilon^{P}_{g_2} = +1, \tag{89}$$

$$\epsilon^{T}_{g_3} = +1, \quad \epsilon^{P}_{g_3} = -1, \tag{90}$$

$$\epsilon^{T}_{\delta m_1} = -1, \quad \epsilon^{P}_{\delta m_1} = -1, \tag{91}$$

$$\epsilon^{T}_{\delta m_2} = +1, \quad \epsilon^{P}_{\delta m_2} = -1. \tag{92}$$

Due to this different transformation behavior, the statics of the components g_2 and δm_1 are decoupled completely from the other components. We obtain the inverse susceptibility matrix of these two variables by taking the second order partial derivatives of the Fourier transformed energy density with respect to the appropriate variables,

$$\chi^{-1}_{ij}(\mathbf{k}) = \begin{pmatrix} \dfrac{1}{\rho} & +iM_0 c_{\perp} k_{\parallel} \\ -iM_0 c_{\perp} k_{\parallel} & M_0^2(K_1 k_{\perp}^2 + K_3 k_{\parallel}^2) \end{pmatrix}, \tag{93}$$

where $i,j \in \{g_2, \delta m_1\}$.

From Eq. (93) one can derive the static susceptibilities

$$\chi_{g_2 g_2} = \frac{\rho}{N}(K_1 k_{\perp}^2 + K_3 k_{\parallel}^2), \tag{94}$$

$$\chi_{\delta m_1 \delta m_1} = \frac{1}{M_0^2 N}, \tag{95}$$

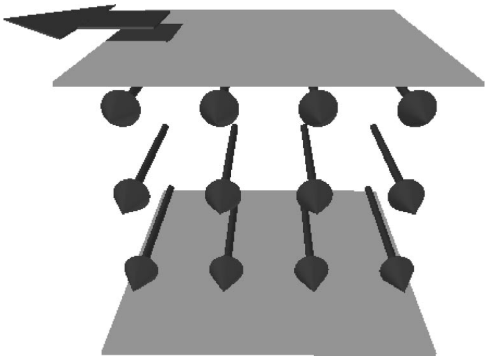

FIG. 2. Perturbation to measure ρ. The conelike arrows represent the orientation of the magnetization while the flat arrow represents the direction of the perturbation.

$$\chi_{g_2 \delta m_1} = \frac{i c_{\perp} k_{\parallel} \rho}{M_0 N}, \tag{96}$$

with $N = K_1 k_{\perp}^2 + K_3 k_{\parallel}^2 - \rho c_{\perp}^2 k_{\parallel}^2$. Now we will discuss the transverse momentum density correlation function in more detail. Therefore we determine the limiting cases for either setting $k_{\perp}$ or $k_{\parallel}$ to zero first,

$$\lim_{k_{\perp}\to 0}\lim_{k_{\parallel}\to 0} \chi_{g_2 g_2} = \rho, \tag{97}$$

$$\lim_{k_{\parallel}\to 0}\lim_{k_{\perp}\to 0} \chi_{g_2 g_2} = \frac{\rho}{1 - \rho c_{\perp}^2/K_3}. \tag{98}$$

Because we discussed the autocorrelation function of the transverse momentum density, Eqs. (97) and (98) give the inertia of the gel against velocity perturbations along the planes with a normal vector either perpendicular or parallel to the preferred direction, respectively. In the first case the usual inertia (Fig. 2) due to mass can be measured while in the second case (Fig. 3) an increase of the inertia can be observed.

The inverse susceptibility matrix of the other three components is obtained in the same manner and reads

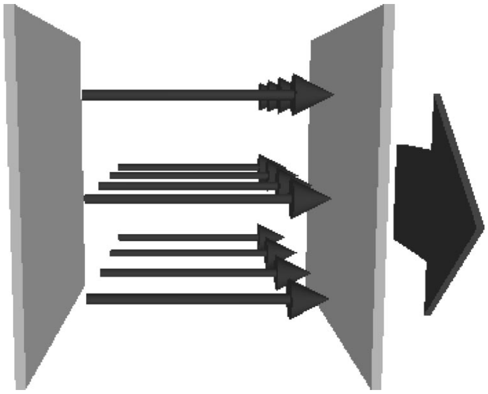

FIG. 3. Perturbation to measure $\rho(1-\rho c_{\perp}^2/K_3)^{-1}$. Again the conelike arrows represent the orientation of the magnetization while the flat arrow represents the direction of the perturbation.

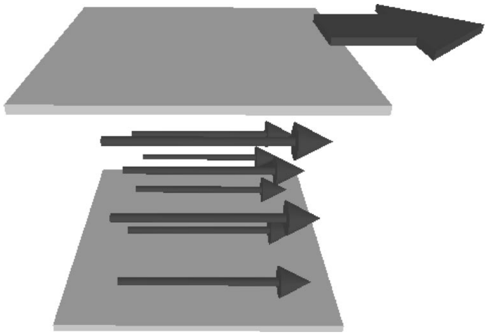

FIG. 4. Perturbation to measure $\rho(1-\rho c_\parallel^2/K_2)^{-1}$. The conelike arrows represent the orientation of the magnetization while the flat arrow represents the direction of the perturbation.

$$\chi_{ij}^{-1} = \begin{pmatrix} \dfrac{1}{\rho} & 0 & +iM_0 c_\perp k_\parallel \\ 0 & \dfrac{1}{\rho} & +iM_0 c_\parallel k_\perp \\ -iM_0 c_\perp k_\parallel & -iM_0 c_\parallel k_\perp & M_0^2(K_2 k_\perp^2 + K_3 k_\parallel^2) \end{pmatrix}, \tag{99}$$

where $i,j \in \{g_1, g_3, \delta m_2\}$.

We again obtain the static susceptibilities by inverting this matrix:

$$\chi_{g_1 g_1} = \rho[(K_2 k_\perp^2 + K_3 k_\parallel^2) - c_\parallel^2 k_\perp^2 \rho]N^{-1}, \tag{100}$$

$$\chi_{g_3 g_3} = \rho[(K_2 k_\perp^2 + K_3 k_\parallel^2) - c_\perp^2 k_\parallel^2 \rho]N^{-1}, \tag{101}$$

$$\chi_{g_1 g_3} = c_\perp k_\parallel c_\parallel k_\perp \rho^2 N^{-1}, \tag{102}$$

$$\chi_{g_1 \delta m_2} = i c_\perp k_\parallel \rho M_0^{-1} N^{-1}, \tag{103}$$

$$\chi_{g_3 \delta m_2} = i c_\parallel k_\perp \rho M_0^{-1} N^{-1}, \tag{104}$$

$$\chi_{\delta m_2 \delta m_2} = M_0^{-2} N^{-1}, \tag{105}$$

with $N = (K_2 k_\perp^2 + K_3 k_\parallel^2) - \rho c_\perp^2 k_\parallel^2 - \rho c_\parallel^2 k_\perp^2$. In analogy to the case of the variables g_2 and δm_1 we can evaluate the limiting expressions for the momentum density correlation function for setting either $k_\perp$ or $k_\parallel$ to zero, first. One obtains

$$\lim_{k_\perp \to 0} \lim_{k_\parallel \to 0} \chi_{g_3 g_3} = \frac{\rho}{1 - \rho c_\parallel^2 / K_2}, \tag{106}$$

$$\lim_{k_\parallel \to 0} \lim_{k_\perp \to 0} \chi_{g_3 g_3} = \rho, \tag{107}$$

$$\lim_{k_\perp \to 0} \lim_{k_\parallel \to 0} \chi_{g_1 g_1} = \rho, \tag{108}$$

$$\lim_{k_\parallel \to 0} \lim_{k_\perp \to 0} \chi_{g_1 g_1} = \frac{\rho}{1 - \rho c_\perp^2 / K_3}. \tag{109}$$

Here an increase of the inertia can be measured for velocity perturbations parallel to the preferred direction (Fig. 4) while for perturbations perpendicular to the frozen-in magnetization the usual inertia is observed (Figs. 5 and 6).

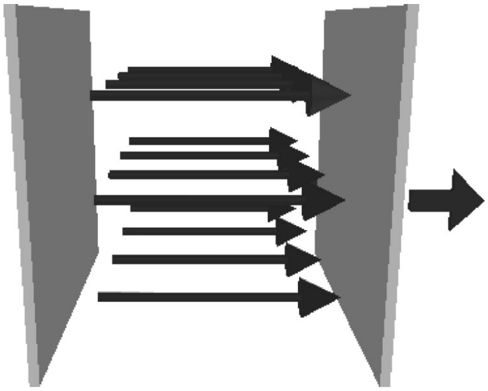

FIG. 5. Perturbation to measure ρ. Again the conelike arrows represent the orientation of the magnetization while the flat arrow represents the direction of the perturbation.

It is worth mentioning that there exists a correlation between the parallel and the transverse components of the momentum density given by Eq. (102).

C. Field-induced strain

As a last example we want to discuss the k^0 dynamics of the system, if we apply an oscillating external magnetic field perpendicular to the frozen-in magnetization. In our case we apply a magnetic field in the z direction (cf. Fig. 7). We again identify the relevant equations. At first one can neglect all the dynamic equations for the concentration c, the entropy density σ, the mass density ρ, and the momentum density $\mathbf{g}$, because these are true hydrodynamic variables and contain apart from the time derivative of the variable the gradient of the current related to that variable. From the quasicurrents of the remaining variables we consider only the contributions without gradient terms. We obtain for their reversible parts

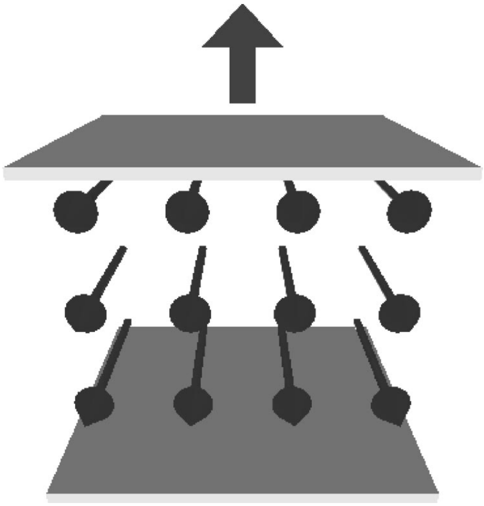

FIG. 6. Perturbation to measure ρ. The conelike arrows represent the orientation of the magnetization while the flat arrow represents the direction of the perturbation.

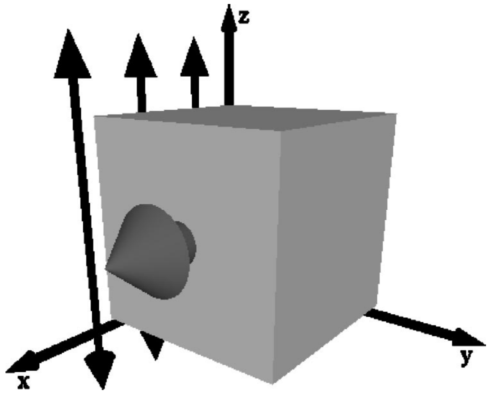

FIG. 7. Sketch of experimental setup. The big conelike arrow represents the magnetization of the uniaxial gel while the four smaller arrows represent the direction of the alternating external magnetic field.

$$Y_{ij}^{R} = \xi_{ijk}^{YR} W_k, \tag{110}$$

$$X_i^{R} = \xi_{ij}^{XR} W_j + b_{ij}^{R} h_j^{M}, \tag{111}$$

$$Z_i^{R} = -\xi_{ij}^{XR} h_j^{M} - \xi_{kli}^{YR} \Psi_{kl} + \tau_{ij}^{R} W_j. \tag{112}$$

And in the same manner for the irreversible parts

$$Y_{ij}^{D} = 0, \tag{113}$$

$$X_i^{D} = b_{ij} h_j^{M} + \xi_{ij} W_j, \tag{114}$$

$$Z_i^{D} = \tau_{ij} W_j + \xi_{ij}^{X} h_j^{M}. \tag{115}$$

One ends up with a rather complex set of equations. Here we concentrate on some qualitatively different features due to the dynamic couplings (and neglect magnetostriction).

After applying an oscillatory external field in the z direction, an oscillating magnetization in the z direction is induced due to Eqs. (68) and (69). This magnetization leads to an oscillation of the relative rotations (57), (61), and (62), where the maximum amplitudes are related by

$$\tilde{\Omega}_y \sim -\xi^{XR} \alpha' M_0 \delta m_z, \tag{116}$$

$$\tilde{\Omega}_z \sim \xi^{X} \alpha' M_0 \delta m_z. \tag{117}$$

The relative rotations couple to the strain field (58) and (63) and therefore lead to an oscillating strain in the plane perpendicular to the applied field as well as in the plane spanned by the frozen-in and the induced magnetization with

$$\epsilon_{xy} \sim D_1 \xi^{YR} \xi^{X} M_0 \delta m_z, \tag{118}$$

$$\epsilon_{xz} \sim D_1 \xi^{YR} \xi^{XR} M_0 \delta m_z. \tag{119}$$

These shear strains should be experimentally observable with piezoelectric transducers.

Second, we observe that, apart from the directly induced magnetization parallel to the applied field, a magnetization in the third direction, perpendicular to both the frozen-in and the applied field is induced as well (56) and (59)

$$M_0 \delta m_y \sim b^{R} H_z^{ext}. \tag{120}$$

This effect differs from the case of isotropic ferrogels studied by Jarkova *et al.* [9] where this effect appears in higher order of the magnetization, because in this case one has to induce a magnetization first. Experimentally one can measure these effects by using Hall probes.

For the last effect the crucial coefficient b^R can be calculated explicitly, using the microscopic theory of linear response. In this limit we make use of the results given by Forster, who used the projector formalism of Zwanzig and Mori. In the notation of [14] the temporal change of a macroscopic variable a_i is given by

$$\delta \dot{a}_i(\mathbf{x}t) = [-i\omega_{ij}(-i\boldsymbol{\nabla}) + \sigma_{ij}(-i\boldsymbol{\nabla})]\delta\lambda_j(\mathbf{x}t). \tag{121}$$

In this notation the λ_i are the thermodynamic forces of the system. The matrix ω_{ij} is called the frequency matrix while the matrix σ_{ij} is called the memory matrix.

At $t=0$ the external perturbation is switched on, and the system starts to relax toward the new equilibrium. The initial time derivative $\delta\dot{a}_i(\mathbf{k},t=0^{+})$ is given by [14]

$$\delta\dot{a}_i(\mathbf{k},t=0^{+}) = -i\omega_{ij}(\mathbf{k})\delta\lambda_j(\mathbf{k}). \tag{122}$$

Using the explicit expressions for the dynamic coupling tensors b_{ij}^{R} and ξ_{ij}^{XR}, one obtains for the dynamical equation of the magnetization

$$\dot{M}_i + \underbrace{b^{R}\epsilon_{ijk} m_k h_j^{M}}_{\omega_{\mathrm{MM}}} + \xi^{XR}\epsilon_{ijk} m_k W_j = 0. \tag{123}$$

In the last equation we discarded the contributions of the irreversible parts of the quasicurrent. This can be done because the frequency matrix ω_{ij} contains the instantaneous collisionless contributions which are purely reversible [14].

Explicitly, the frequency matrix for the autocorrelation of the magnetization reads, using Eq. (123),

$$\omega_{\mathbf{MM}} = \begin{pmatrix} 0 & ib^{R}m_z & -ib^{R}m_y \\ -ib^{R}m_z & 0 & ib^{R}m_x \\ ib^{R}m_y & -ib^{R}m_x & 0 \end{pmatrix}. \tag{124}$$

Using for the frequency matrix ω_{ij} of the variables M_i and M_j the representation

$$\omega_{ij}(\mathbf{k}) = \int \frac{d\omega}{\pi}\chi_{ij}'' = \frac{1}{\hbar}\int d\mathbf{x} \times e^{-i\mathbf{k}\cdot\mathbf{x}}\langle[M_i(\mathbf{r}-\mathbf{r}',t-t'),M_j(0,0)]\rangle \tag{125}$$

and taking into account the commutator for the magnetization

$$\langle[\hat{M}_i,\hat{M}_j]\rangle = i\hbar\epsilon_{ijk}\langle\hat{M}_k\rangle \tag{126}$$

we obtain for the frequency matrix in the microscopic theory

$$\omega_{\mathbf{MM}} = \begin{pmatrix} 0 & iM_0\langle\hat{m}_z\rangle & -iM_0\langle\hat{m}_y\rangle \\ -iM_0\langle\hat{m}_z\rangle & 0 & iM_0\langle\hat{m}_x\rangle \\ iM_0\langle\hat{m}_y\rangle & -iM_0\langle\hat{m}_x\rangle & 0 \end{pmatrix}. \tag{127}$$

Now we can compare these matrices obtained in two different ways. We can conclude that the coefficient b^R in the k^0

dynamics and in the linear and homogeneous regime is given by

$$b^R = M_0. \tag{128}$$

This result is also compatible with the results of Jarkova *et al.* for isotropic ferrogels, because if we set M_0 to zero, the coefficient b^R and thus the instantaneous response to the external field vanish.

IV. DISCUSSION AND CONCLUSIONS

Ferromagnetic gels are uniaxial, if the frozen-in magnetization denotes the only preferred direction. Such materials are potentially very interesting for a variety of applications. Here we investigate theoretically the thermodynamics and hydrodynamics of these systems. Uniaxial magnetic gels show on the one hand similarities to other anisotropic gels, like nematic elastomers, and to isotropic ferrofluids and ferrogels, but the combination of preferred direction, magnetic degree of freedom, and elasticity makes them unique and very peculiar.

Prominent features are the relative rotations between the magnetization and the elastic network, which couple dynamically flow, shear, and magnetic reorientation. As a result, shear flow in a plane that contains the frozen-in magnetization induces a rotation of the magnetization, not only within the shear plane, but also out of the shear plane. This behavior is qualitatively different from that of other types of materials. The basic results hold, even if the constant shear flow is replaced by an oscillating one, which is very likely done in actual experiments, although the formulas for that case will become much more complicated.

Another outstanding aspect of the hydrodynamics of this material is the difference between the mass current density (mass density times velocity) and the momentum density due to a nonvanishing magnetization vorticity. Unheard of in other classical condensed phases, it is known from some uniaxial quantum fluids, where, however, experiments on this aspect are impossible. In uniaxial ferromagnetic gels the static susceptibilities for momentum fluctuations (the long wavelength limit of the static momentum correlation functions) are given by the (bare) density for some geometries only, but show an increased renormalized effective density for other directions.

Finally, we looked at an oscillating external magnetic field that induces not only an oscillation of the magnetization in the direction of the external field, but also oscillating shear strains. The latter are found in planes that contain the frozen-in magnetization and either the external field or the third, perpendicular direction. In addition, the external field also induces a magnetization component perpendicular to both the field and the frozen-in magnetization. The reversible transport coefficient that governs this effect can be calculated by referring to the microscopic quantum mechanical spin-type dynamics for magnetic moments and using the projector formalism to evaluate the frequency matrix. This coefficient vanishes with the magnetization and is, thus, characteristic for this type of ferromagnetic gel.

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