Dissipation in ferrofluids: Mesoscopic versus hydrodynamic theory

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Part of the field dependent dissipation in ferrofluids occurs due to the rotational motion of the ferromagnetic grains relative to the viscous flow of the carrier fluid. The classical theoretical description due to Shliomis (Zh. Éksp. Teor. Fiz. **61**, 2411 (1971) [Sov. Phy JETP **34**, 1291 (1972)]) uses a mesoscopic treatment of the particle motion to derive a relaxation equation for the nonequilibrium part of the magnetization. Complementary, the hydrodynamic approach of Liu [Phys. Rev. Lett. **70**, 3580 (1993)] involves only macroscopic quantities and results in dissipative Maxwell equations for the magnetic fields in the ferrofluid. Different stress tensors and constitutive equations lead to deviating theoretical predictions in those situations, where the magnetic relaxation processes cannot be considered instantaneous on the hydrodynamic time scale. We quantify these differences for two situations of experimental relevance, namely, a resting fluid in an oscillating oblique field and the damping of parametrically excited surface waves. The possibilities of an experimental differentiation between the two theoretical approaches is discussed. [S1063-651X(99)15011-6]

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I. INTRODUCTION

The interplay between hydrodynamic behavior and magnetic field sensitivity gives rise to a variety of new physical effects and makes ferrofluids a fascinating subject of research with many interesting technical applications [1–3]. Practically all hydrodynamic properties can be modified or modulated by an external magnetic field. Although very similar effects are possible with electrically polarized fluids, the coupling of the fields to the hydrodynamics is more pronounced in the magnetic case. Since the experimental handling of strong magnetic fields is simpler than that of electric ones, ferrofluids are the experimenters choice to analyze the field affected hydrodynamic motion.

On the other hand, a thorough theoretical understanding of the many aspects of ferrofluid motion is a rather ambitious task [4,5]. This is in particular true if the motion is such that dissipation effects are important. The theoretical analysis must then account for the deviations from and the relaxation towards equilibrium in a consistent and quantitatively accurate way.

One of the interesting properties of ferrofluids is their tunable viscosity which can be controlled by an external magnetic field. This so called magnetoviscous effect was first observed experimentally by McTague [6]. Qualitatively it can be accounted for [7] by magnetic torques acting upon the suspended ferromagnetic particles: depending on the relative orientation between magnetic field and local vorticity the particle rotation is hindered. The friction at the coated particle surfaces generates an extra dissipation and thus leads to an enhanced effective viscosity. This additional rotational friction is described by a ''rotational viscosity'' η_R. Based on the theory of rotational Brownian motion of noninteracting rigid dipoles, Shliomis [4] developed a theoretical description of this effect. The basic ingredient is a relaxation equation for the nonequilibrium part of the magnetization resulting from a stochastic description of an ensemble of ferromagnetic grains. This treatment allows us to derive an analytic expression for the magnetic field dependence of η_R [4]. We will refer to this approach as the *mesoscopic* theory since it starts with a rather detailed characterization of the processes occurring on a 10 nm scale in order to determine macroscopic properties such as the viscosity.

Quite complementary, the same phenomena can be described within the framework of nonequilibrium thermodynamics. Appropriate magnetic field variables must then be introduced into the set of relevant thermodynamic quantities. New dissipative currents show up when the system is driven out of equilibrium. They are coupled to the thermodynamic forces by additional Onsager coefficients. This program was carried out by Liu and co-workers in a series of papers [5,8,9] and gave rise to what is called the *hydrodynamic* Maxwell theory. Making no reference at all to the microscopic mechanism of dissipation this approach has the appealing feature of being very general and applicable to electrically or magnetically polarizable continuous media. As in every hydrodynamic theory the Onsager coefficients are free parameters which have to be determined experimentally or from a microscopic theory.

The detailed quantitative relation between the two theoretical approaches is presently not completely clear and there has been some controversy in the literature over recent years. In order to contribute to a clarification of this issue we investigate in the present paper the consequences of both the mesoscopic and the hydrodynamic theory for two simple experimental setups involving ferrofluids. On the one hand, we analyze the influence of an oscillating oblique magnetic field on a ferrofluid at rest, on the other hand we investigate the damping of parametrically driven surface waves on the ferrofluid in a constant external magnetic field perpendicular to the undisturbed surface. Both situations are easily accessible to experiments. Interestingly the two theoretical approaches give rise to *different* results for some of the relevant quantities. We finally discuss whether these differences are pronounced enough to allow an experimental decision between the theories.

II. EQUATIONS OF MOTION

The fluid motion is governed by the hydrodynamic balance equations for mass and linear momentum. For an incompressible ferrofluid (density ρ) with a velocity field $\mathbf{v}(\mathbf{r},t)$ these equations read as

$$\nabla \cdot \mathbf{v}=0, \tag{1}$$

$$\rho(\partial_t+\mathbf{v}\cdot\nabla)\mathbf{v}=\nabla\cdot\Pi+\rho\mathbf{g}, \tag{2}$$

where Π_{ij} is the stress tensor containing reactive and dissipative contributions (see later) and $\mathbf{g}=(0,0,-g)$ is the acceleration due to gravity. The two theoretical approaches under consideration differ in their expressions for the stress tensor and the field equations. It will turn out that they produce different results when the magnetic relaxation time τ (usually between 10^{-3} and 10^{-6} s) cannot be neglected with respect to the time scale, $1/\omega$ say, of the hydrodynamic motion. In those cases it is argued that the local magnetization $\mathbf{M}(\mathbf{r},t)$ deviates from its local equilibrium value $\mathbf{M}_{eq}(\mathbf{r},t)$ giving rise to an additional dissipation, which we shall denote here as ''magnetodissipation.'' This phenomenon is responsible for the field dependent viscosity [4,6,7], the magnetovortical resonance [10,11], or the spectacular negative viscosity effect [12–14] detected recently in ferrofluids.

In order to simplify the mathematics we make throughout this paper the following approximations which do not remove the basic differences between the two approaches: (i) The magnetic field $\mathbf{H}$ within the ferrofluid is weak enough to warrant a linear relationship $\mathbf{M}_{eq}=\chi\mathbf{H}$, where the susceptibility is supposed to be proportional to the density ρ. (ii) The local nonequilibrium magnetization $\mathbf{M}$ deviates only slightly from $\mathbf{M}_{eq}$. (iii) Magnetodissipation is treated in the so called low-frequency limit characterized by $\tau\omega\ll 1$.

A. Field equations in the mesoscopic theory

Following Shliomis [4] (see also Ref. [1]) the stress tensor for ferrofluids appears in the form

$$\Pi_{ij}^{mes}=-\left(p+\frac{\mu_0}{2}H^2\right)\delta_{ij}+H_iB_j+\eta(\nabla_i v_j+\nabla_j v_i)$$
$$+\frac{\mu_0}{2}\varepsilon_{ijk}(\mathbf{M}\times\mathbf{H})_k. \tag{3}$$

In Eq. (3) occurs the pressure field $p(\mathbf{r},t)$, the Maxwell stress tensor with the induction $\mathbf{B}=\mu_0(\mathbf{H}+\mathbf{M})$, and the usual dissipative contribution proportional to the shear viscosity η. Moreover, the last term on the right-hand side of Eq. (3) arises if the magnetization exhibits an off-equilibrium component perpendicular to $\mathbf{H}$. It is related to the magnetodissipation and guarantees the symmetry of the stress tensor if the local directions of $\mathbf{H}$ and $\mathbf{B}$ do not coincide. The appearance of $\mathbf{M}$ in the stress tensor (3) requires an extra constitutive equation for the off-equilibrium component $\delta\mathbf{M}=\mathbf{M}-\chi\mathbf{H}$ of the magnetization. Under the above simplifications (i)–(iii) the following relation can be established [2]:

$$\delta\mathbf{M}=-\tau\chi\left[\partial_t\mathbf{H}+(\mathbf{v}\cdot\nabla)\mathbf{H}-\frac{\nabla\times\mathbf{v}}{2}\times\mathbf{H}\right], \tag{4}$$

where τ denotes a relaxation time.

The magnetic part of the problem is treated in the framework of the magnetostatic approximation

$$\nabla\times\mathbf{H}=0, \tag{5}$$

$$\nabla\cdot\mathbf{B}=0. \tag{6}$$

With this simplification and the above expression for the stress tensor (3) the hydrodynamics obeys the Navier-Stokes equation, which assumes the form

$$\rho\partial_t\mathbf{v}+\rho(\mathbf{v}\cdot\nabla)\mathbf{v}=-\nabla p+\rho\mathbf{g}+\eta\nabla^2\mathbf{v}+\mu_0(\mathbf{M}\cdot\nabla)\mathbf{H}$$
$$+\frac{\mu_0}{2}\nabla\times(\mathbf{M}\times\mathbf{H}). \tag{7}$$

B. The field equations of the hydrodynamic theory

The complete set of hydrodynamic Maxwell equations is given by Ref. [5]

$$\partial_t\mathbf{D}=\nabla\times\mathbf{H}-\mathbf{j}_{el}, \quad \partial_t\mathbf{B}=-\nabla\times\mathbf{E}, \tag{8}$$
$$\nabla\cdot\mathbf{D}=\rho_{el}, \quad \nabla\cdot\mathbf{B}=0,$$

where $\mathbf{E}$ and $\mathbf{D}$ denote the electric and displacement fields. For nonconducting ferrofluids there are no electric charges or currents, thus $\mathbf{j}_{el}=0$ and $\rho_{el}=0$. The fields $\mathbf{H}$ and $\mathbf{E}$ split into reactive and dissipative parts

$$\mathbf{H}=\mathbf{H}^R+\mathbf{H}^D \quad \text{and} \quad \mathbf{E}=\mathbf{E}^R+\mathbf{E}^D,$$

where $\mathbf{H}^R=\partial\epsilon/\partial\mathbf{B}$ and $\mathbf{E}^R=\partial\epsilon/\partial\mathbf{D}$ are derivatives of the thermodynamic energy density ϵ and contain only equilibrium information. The dissipative fields $\mathbf{H}^D$ and $\mathbf{E}^D$ represent the off-equilibrium currents and are functions of the thermodynamic forces. For isotropic isothermal situations and neglecting off-diagonal couplings they depend only on the electromagnetic fields in the local rest frame and are given by [5]

$$\mathbf{H}^D=-\frac{\alpha}{\mu_0}\nabla\times(\mathbf{E}^R+\mathbf{v}\times\mathbf{B})$$

and

$$\mathbf{E}^D=\frac{\beta}{\epsilon_0}\nabla\times(\mathbf{H}^R-\mathbf{v}\times\mathbf{D}), \tag{9}$$

where the parameters α and β are Onsager coefficients. In the present paper we restrict ourselves to ferrofluids with a negligible dielectric susceptibility $\chi_{el}\simeq 0$ (e.g., hydrocarbon based ferrofluids) giving rise to $\beta\simeq 0$ (see also Ref. [15]) implying $\mathbf{E}^D=0$. Moreover, the ratio between time and space derivatives of fields, respectively, is $\omega/(ck)$ and for hydrodynamic frequencies $\omega\lesssim 10^3$ s^{-1} and wavelengths $1/k\sim 10^{-3}$ m^{-1} we may neglect the time derivative of $\mathbf{D}$ in the Maxwell equations (8). This means that the magnetic field may be assumed to follow the fluid motion instantaneously. In this way we recover the magnetostatic field equations

$$\nabla\cdot\mathbf{B}=0, \tag{10}$$

$$\nabla\times\mathbf{H}=0.$$

At a liquid-air interface these fields are subjected to the usual continuity conditions for the normal component of **B** and the tangential component of **H**.

Using incompressibility we get from Eqs. (8) and (9) the following constitutive relation for the dissipative field:

$$\mathbf{H}^D=\frac{\alpha}{\mu_0}[\partial_t\mathbf{B}+(\mathbf{v}\cdot\nabla)\mathbf{B}-(\mathbf{B}\cdot\nabla)\mathbf{v}], \tag{11}$$

which is the analog to $\delta\mathbf{M}$ in the approach of Shliomis. For later use we introduce the *reactive* contribution to the magnetization

$$\mathbf{M}^R=\frac{1}{\mu_0}\mathbf{B}-\mathbf{H}^R. \tag{12}$$

This definition differs from the ''real'' magnetization $\mathbf{M}=\mathbf{B}/\mu_0-\mathbf{H}$ in using only the reactive part $\mathbf{H}^R$ of the magnetic field. The advantage of this formulation is that for the present case of constant and scalar susceptibility the fields $\mathbf{B},\mathbf{H}^R$, and $\mathbf{M}^R=\chi\mathbf{H}^R$ are all parallel to each other [16]. The fact that **H** and hence **M** are not parallel to **B** is the very reason for the magnetodissipative effects [4–6] investigated here.

With the assumption of the susceptibility being proportional to the density ρ, the stress tensor of the fluid simplifies to [5,17]

$$\Pi_{ij}^{hyd}=-\left[p+\frac{\mu_0}{2}(H^R)^2\right]\delta_{ij}+H_i^RB_j+\eta(\nabla_jv_i+\nabla_iv_j). \tag{13}$$

The fluid dynamics is therefore governed by the Navier-Stokes equation, which using Eq. (13) acquires the form

$$\rho\partial_t\mathbf{v}+\rho(\mathbf{v}\nabla)\mathbf{v}=-\nabla p+\rho\mathbf{g}+\eta\nabla^2\mathbf{v}+\mu_0M^R\nabla H^R +\mathbf{B}\times(\nabla\times\mathbf{H}^D). \tag{14}$$

The fourth term on the right-hand side of the Navier-Stokes equation was simplified to involve only the magnitudes of the vectors $\mathbf{M}^R$ and $\mathbf{H}^R$ by exploiting the fact that they are parallel.

C. Comparison between the two approaches

Let us first emphasize that the two descriptions introduced above are fully equivalent if the magnetic relaxation processes can be considered to be instantaneous ($\tau=\alpha=0$) on the time scale of the hydrodynamic motion. In this case of vanishing magnetodissipation we have $\delta\mathbf{M}=\mathbf{H}^D=0$, i.e., $\mathbf{M}^R=\mathbf{M}\|\mathbf{H}=\mathbf{H}^R$, and thus the stress tensors (3) and (13) or, respectively, the Navier-Stokes equations (7) and (14) coincide.

Differences between the two approaches arise due to the treatment of the magnetodissipation occurring at finite $\delta\mathbf{M}$ and $\mathbf{H}^D$, respectively. By comparing Eqs. (3) and (13) and expressing the antisymmetric part of the tensor element $H_i^DB_j$ in Eq. (13) by $(1/2)\varepsilon_{ijk}(\mathbf{H}^D\times\mathbf{B})_k=-(\mu_0/2)\varepsilon_{ijk}(\mathbf{M}\times\mathbf{H})_k$ one arrives at

$$\Pi_{ij}^{hyd}-\Pi_{ij}^{mes}=\mu_0\mathbf{H}\cdot\mathbf{H}^D\delta_{ij}-\frac{1}{2}(H_i^DB_j+H_j^DB_i), \tag{15}$$

where terms of order $(H^D)^2$ have been dropped. The stress tensors in Eq. (15) differ by a diagonal element and a symmetric nondiagonal contribution. For incompressible fluids the isotropic increment just leads to a renormalization of the pressure, which we won't pursue here. The nondiagonal increment, however, gives rise to a dissipative force which might be detectable in an experiment (see below).

It is also instructive to compare the constitutive equations, which determine the dissipative fields: From the identity

$$\mu_0(\mathbf{H}+\mathbf{M})=\mathbf{B}=\mu_0(\mathbf{H}^R+\mathbf{M}^R)=\mu_0(1+\chi)\mathbf{H}^R \tag{16}$$

one finds that the dissipative field contributions $\delta\mathbf{M}$ and $\mathbf{H}^D$ coincide up to a prefactor:

$$\mathbf{H}^D=-\frac{1}{1+\chi}(\mathbf{M}-\chi\mathbf{H})=-\frac{1}{1+\chi}\delta\mathbf{M}. \tag{17}$$

The associated constitutive relations

$$\delta\mathbf{M}=-\tau\chi\left[\partial_t\mathbf{H}+(\mathbf{v}\cdot\nabla)\mathbf{H}-\frac{1}{2}(\nabla\times\mathbf{v})\times\mathbf{H}\right] \tag{18}$$

$$\mathbf{H}^D=\frac{\alpha}{\mu_0}[\partial_t\mathbf{B}+(\mathbf{v}\cdot\nabla)\mathbf{B}-(\mathbf{B}\cdot\nabla)\mathbf{v}], \tag{19}$$

however, are not fully compatible: Omitting the contributions of $O(\alpha\omega)^2$, Eq. (19) can be re-cast as

$$\mathbf{H}^D=\alpha(1+\chi)[\partial_t\mathbf{H}+(\mathbf{v}\cdot\nabla)\mathbf{H}-(\mathbf{H}\cdot\nabla)\mathbf{v}]. \tag{20}$$

By comparing the two first terms on the right-hand side of Eqs. (18) and (20) and using Eq. (17) one can relate the characteristic relaxation times by [18]

$$\alpha=\tau\frac{\chi}{(1+\chi)^2}. \tag{21}$$

The third terms in Eqs. (18) and (20) describe different physics. This is easy to see by considering a homogeneous stationary magnetic field: According to the mesoscopic theory, Eq. (18), only a rotational flow is able to drive a magnetodissipative field. Contrary in the hydrodynamic approach, Eq. (20) states that a flow gradient parallel to the magnetic field suffices to excite a finite $\mathbf{H}^D$. It is interesting to point out that the deviation between the constitutive equations, Eqs. (18) and (20), drops out for a solid body rotation flow field $\mathbf{v}=\omega\times\mathbf{r}$, which is often investigated in the ferrofluid literature.

To summarize this section: We observe two differences between the classical description of Shliomis and the hydrodynamic Maxwell theory of Liu: One in the stress tensors Π_{ij} and a second in the constitutive equations for the dissipative fields $\delta\mathbf{M}$ and $\mathbf{H}^D$, respectively. In the following we investigate the possibilities of an experimental differentiation between the two theoretical approaches.

III. FERROFLUID AT REST IN AN OBLIQUE OSCILLATING MAGNETIC FIELD

We consider a resting ferrofluid layer exposed to a linearly polarized magnetic field $\mathbf{H}^{(2)}=(H_x^{(2)},0,H_z^{(2)})\cos\omega t$ directed oblique to the surface. Here and in the following, fields in the air carry the superscript ''(2),'' while fields within the ferrofluid are denoted without superscript.

The stress tensor in the air above the ferrofluid is taken as

$$\Pi_{ij}^{(2)}=-\left[p^{atm}+\frac{\mu_0}{2}(H^{(2)})^2\right]\delta_{ij}+H_i^{(2)}B_j^{(2)}, \qquad (22)$$

where we assume a constant atmospheric pressure p^{atm}.

For a fluid at rest the constitutive equations (18) and (19) coincide and thus eventual discrepancies between the two theories will result from the deviating stress tensors. To test possible differences we evaluate the tangential force $f_t=\Pi_{xz}-\Pi_{xz}^{(2)}$ acting upon a free surface element. By using the magnetic interface conditions for $\mathbf{H}$ and $\mathbf{B}$ and owing to the fact that isotropic tensor elements do not contribute to tangential forces we obtain

$$f_t^{mes}=\frac{\mu_0}{2}(\delta M_xH_z-\delta M_zH_x)=-\frac{\mu_0}{2}\tau\chi[H_z\partial_tH_x-H_x\partial_tH_z] \qquad (23)$$

and

$$f_t^{hyd}=-H_x^DB_z=-\frac{\alpha}{\mu_0}B_z\partial_tB_x. \qquad (24)$$

The computation of f_t requires knowledge of $\mathbf{H}$ and $\mathbf{B}$ within the ferrofluid. From the respective constitutive equations and the interface conditions we find

$$\mathbf{H}^{mes}=\frac{1}{2}\left[H_x^{(2)},0,\frac{H_z^{(2)}}{1+\chi-i\omega\tau\chi}\right]e^{i\omega t}+\text{c.c.} \qquad (25)$$

$$\mathbf{B}^{hyd}=\frac{\mu_0}{2}\left[\frac{H_x^{(2)}}{\frac{1}{1+\chi}+i\omega\alpha},0,H_z^{(2)}\right]e^{i\omega t}+\text{c.c.} \qquad (26)$$

Evaluating the tangential stresses (23) and (24) up to leading order in $\tau\omega$ or, respectively, $\alpha\omega$ we get

$$f_t^{mes}=O((\tau\omega)^2) \qquad (27)$$

and

$$f_t^{hyd}=\frac{\mu_0}{2}\alpha\omega(1+\chi)H_x^{(2)}H_z^{(2)}\sin 2\omega t+O((\alpha\omega)^2). \qquad (28)$$

At the considered accuracy level, the mesoscopic approach states that the resting ferrofluid is in equilibrium, while the hydrodynamic Maxwell theory predicts a residual tangential surface force oscillating with twice the excitation frequency. This force drops out in the static limit $\omega\to 0$ or when the applied field is directed normally or tangentially to the surface. The maximum effect is achieved for an inflection angle of 45°, where the product of the incident field components $H_x^{(2)}H_z^{(2)}$ is maximum. We conclude that the resting fluid cannot be in equilibrium, i.e., the force will drive a convective motion. With the aid of appropriate tracer particles exposed to the surface of the fluid we expect the acceleration to be easily detectable in an experiment.

IV. SURFACE WAVES WITH MAGNETODISSIPATION

In this section we investigate a setup, which aims at probing the different constitutive relations [Eqs. (18) and (19)]. As outlined in Sec. II C a nonrotational flow profile is most appropriate for this purpose. Surface waves on a deep fluid are a classical example: The associated flow field is purely potential, except in a thin boundary layer along the system boundaries. By increasing the system dimensions this parasitic influence can be limited to the viscous skin layer beneath the free surface. Our aim is to derive the complex dispersion relation for low frequency small amplitude surface waves with magnetodissipation taken into account.

A. Surface waves without magnetodissipation

In this subsection we review the present knowledge on small amplitude surface waves on a ferrofluid in a static magnetic field $\mathbf{H}_{ext}$ normal to the surface. With $z=\zeta(x,t)\propto e^{i(kx-\omega t)}$ we denote the surface deflection in vertical z direction depending on the horizontal coordinate x and on time t. The real part of the complex valued function $\omega(k)=\omega'(k)+i\omega''(k)$ reflects the wave dispersion, while the imaginary part accounts for the decay rate.

For a nonviscous ($\eta=0$) ferrofluid $\omega''(k)$ vanishes identically and the inviscid wave dispersion (indicated by the subscript, ''0'') is given by [1,19]

$$\omega_0^2(k,H_{ext})=gk-\frac{\mu_0}{\rho}\frac{\chi^2}{(1+\chi)(2+\chi)}H_{ext}^2k^2+\frac{\gamma}{\rho}k^3. \qquad (29)$$

Here γ denotes the coefficient of surface tension. The magnetic field leads to a negative contribution proportional to k^2. If H_{ext} is sufficiently strong ω_0^2 becomes negative, indicating the onset of the Rosensweig instability.

The effect of viscosity on the dispersion has been investigated theoretically in Refs. [20,21] under the assumption of an infinitely fast magnetic relaxation (vanishing magnetodissipative effect). It is found that the expression for $\omega(k)$ can no longer be given explicitly, rather the following implicit relation applies,

$$\mathcal{D}(k,\omega)=\omega^2-\omega_0^2(k)+X(k,\omega)=0 \qquad (30)$$

where the viscous contribution reads

$$X(k,\omega)=4i\omega\nu k^2+4(\nu k^2)^2\left\{\frac{q}{k}-1\right\}. \qquad (31)$$

Here $\nu=\eta/\rho$ is the kinematic viscosity and $q=\sqrt{k^2-i\omega/\nu}$. Note that the viscous contribution does not depend on the magnetic field and therefore coincides with the expression for nonmagnetic fluids [22]. This is because the magnetic contributions to the stress tensor are purely conservative at vanishing $\delta\mathbf{M}$ or $\mathbf{H}^D$. In the mathematical derivation of Eqs.

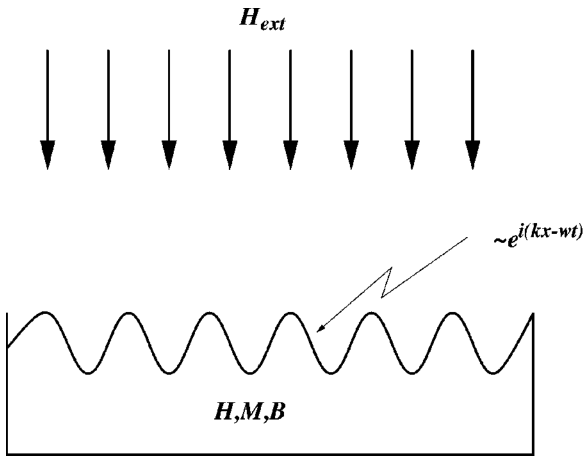

FIG. 1. Sketch of the experimental setup. A ferrofluid of infinite lateral extension and infinite depth is exposed to a stationary magnetic field $\mathbf{H}_{ext}$ directed normally to the surface.

(30) and (31) this is reflected by a decoupling of the magnetostatic from the hydrodynamic problem. Later we will see that this simplification no longer applies if magnetodissipative corrections are taken into account.

B. Formulation of the problem with magnetodissipation included

The investigation requires solving the equations of motion together with appropriate boundary conditions at the free liquid air interface. We consider a horizontally unbounded ferrofluid layer of infinite depth (from $z=0$ to $z=-\infty$) in contact with air at its free surface. The layer is exposed to an external stationary magnetic field $\mathbf{H}_{ext}=(0,0,H_{ext})$ directed perpendicularly to the undisturbed surface (see Fig. 1).

The balances of the normal and tangential stresses at the liquid air interface are expressed by the conditions

$$n_i(\Pi_{ij}-\Pi_{ij}^{(2)})n_j=-\gamma(\nabla_j n_j)n_i\,, \tag{32}$$

$$t_i(\Pi_{ij}-\Pi_{ij}^{(2)})n_j=0, \tag{33}$$

where $\mathbf{t}(\mathbf{r},t)$ and $\mathbf{n}(\mathbf{r},t)$ denote local unit vectors tangential and normal to the surface, respectively. The stress tensor in the air above the ferrofluid is given by Eq. (22). The discontinuity of the normal stress condition arises by virtue of the finite surface tension γ. The kinematic surface condition requires

$$\partial_t\zeta+u\,\partial_x\zeta=w, \tag{34}$$

where $\mathbf{v}=(u,0,w)$. The second lateral space direction y need not be considered.

As usual the boundary conditions for the magnetic field at the free surface read as

$$(\mathbf{B}-\mathbf{B}^{(2)})\cdot\mathbf{n}=0, \tag{35}$$

$$(\mathbf{H}-\mathbf{H}^{(2)})\cdot\mathbf{t}=0. \tag{36}$$

C. The motionless state and linearized equations for the perturbations

As reference state (index 0) we consider the motionless ($\mathbf{v}=\mathbf{0}$) fluid with a flat ($\zeta=0$) surface. The associated magnetic quantities are

$$\mathbf{B}_0=\mathbf{B}_0^{(2)}=(0,0,\mu_0H_{ext}), \tag{37}$$

$$\mathbf{M}_0=\left(0,0,\frac{\chi}{1+\chi}H_{ext}\right),$$

$$\mathbf{H}_0=\left(0,0,\frac{1}{1+\chi}H_{ext}\right),$$

$$\mathbf{H}_0^{(2)}=(0,0,H_{ext}),$$

$$\mathbf{H}^D=\delta\mathbf{M}=0.$$

From the momentum balance with the appropriate boundary condition we get for the pressure

$$p_0(x,z)=-\rho gz-\frac{\mu_0}{2}\left[\frac{\chi}{1+\chi}\right]^2H_{ext}^2. \tag{38}$$

Next we study the time evolution of small perturbations by linearizing the field equations and their boundary conditions around the basic state solution. To this end we introduce small deviations denoted by $\mathbf{v}=(u,0,w)$, δp, ζ, $\mathbf{b}$, $\mathbf{b}^{(2)}$, $\mathbf{h}$, $\mathbf{h}^{(2)}$, $\mathbf{m}$. Linearizing the Navier-Stokes equations (7) and (14) we, respectively, find for the mesoscopic and the hydrodynamic approach

$$\rho\partial_t\mathbf{v}=-\nabla\,\delta p+\eta\Delta\mathbf{v}+\mu_0M_0\partial_z\mathbf{h}+\frac{\mu_0}{2}\nabla\times[\mathbf{M}_0\times\mathbf{h}+\mathbf{m}\times\mathbf{H}_0] \tag{39}$$

and

$$\rho\partial_t\mathbf{v}=-\nabla\,\delta p+\eta\Delta\mathbf{v}+\mu_0M_0\nabla h_z+\mu_0H_{ext}(\nabla H_z^D-\partial_z\mathbf{H}^D). \tag{40}$$

Here we have exploited the fact that $\mathbf{H}_0$ and $\mathbf{M}_0$ are both parallel to the z direction. The linear magnetostatic field equations remain unchanged

$$0=\nabla\cdot\mathbf{b}=\nabla\cdot\mathbf{b}^{(2)} \tag{41}$$

$$0=\nabla\times\mathbf{h}=\nabla\times\mathbf{h}^{(2)}. \tag{42}$$

The boundary conditions (35) and (36) are linearized by using the lowest order expressions

$$\mathbf{n}=\left(-\frac{\partial\zeta}{\partial x},0,1\right)\quad\text{and}\quad\mathbf{t}=\left(1,0,\frac{\partial\zeta}{\partial x}\right) \tag{43}$$

to get

$$b_z=b_z^{(2)}, \tag{44}$$

$$h_x-h_x^{(2)}=M_0\frac{\partial\zeta}{\partial x} \tag{45}$$

for the magnetic interface conditions. The kinematic surface condition simplifies to

$$\partial_t \zeta = w, \tag{46}$$

while we get

$$0 = -\rho g \zeta + \delta p + \mu_0 M_0 m_z - 2\eta \partial_z w + \gamma \partial_x^2 \zeta \tag{47}$$

for the normal stress balance at the surface. Introducing the dimensionless magnetodissipative perturbation parameter

$$\kappa = \frac{\alpha \mu_0}{\eta} H_{ext}^2 = \frac{\tau \mu_0 \chi}{\eta (1+\chi)^2} H_{ext}^2, \tag{48}$$

the condition for the tangential stress reads in the mesoscopic approach

$$\frac{\kappa}{4}(\partial_x w - \partial_z u) = (\partial_z u + \partial_x w), \tag{49}$$

whereas for the hydrodynamic Maxwell theory we simply find

$$B_0 H_x^D = \eta(\partial_z u + \partial_x w). \tag{50}$$

To elucidate the time evolution of the perturbed state we assume for all lateral field dependencies a plane wave behavior $\propto \exp[i(kx-\omega t)]$, e.g., $\mathbf{v} = \overline{v}(z)\exp[i(kx-\omega t)]$ or $\zeta = \overline{\zeta}\exp[i(kx-\omega t)]$, etc. The associated z dependence (if appropriate) is indicated by a bar. We now first determine the magnetic fields from Eqs. (41) and (42) in terms of the surface deflection ζ and the flow field $\mathbf{v}$ using the boundary conditions (44) and (45). Then we solve for the velocity field and impose the hydrodynamic boundary conditions (49) and (50), respectively. The condition for nontrivial solutions of Eq. (47) finally yields the desired complex dispersion relation $\omega(k)$. The explicit calculations are sketched in the Appendix.

D. Results

The expressions we find for the implicit dispersion relation $\mathcal{D}(k,\omega)$ are of the form

$$0 = \mathcal{D}^{mes}(\omega,k) = \omega^2 - \omega_0^2(k) + 4i\omega\nu k^2 + 4(\nu k^2)^2\left\{\frac{\tilde{q}}{k} - 1\right\} - \kappa(\nu k^2)^2 \frac{\chi}{2+\chi}\left\{\frac{\tilde{q}}{k} - 1\right\}, \tag{51}$$

$$0 = \mathcal{D}^{hyd}(\omega,k) = (\omega^2 + 2i\omega\nu k^2)\frac{1 - \frac{2\nu k^2}{i\omega} + \kappa\frac{\nu k^2}{i\omega}\frac{\chi}{\chi+2}}{1+\kappa\frac{\nu k^2}{i\omega}} - \omega_0^2(k) + 2(2+\kappa)\nu^2 k^3 \hat{q}\frac{1+\frac{\kappa}{\chi+2}}{1+\kappa\frac{\nu k^2}{i\omega}}, \tag{52}$$

with

$$\tilde{q}^2 = k^2 - \frac{i\omega}{\nu(1+\kappa/4)}, \tag{53}$$

$$\hat{q}^2 = \frac{k^2 - \frac{i\omega}{\nu}}{1+\kappa}. \tag{54}$$

We analyze these results in the following for a ferrofluid with a magnetic susceptibility and viscosity appropriate for surface wave experiments [e.g., APG J12 (Ref. [23]) Ferrofluidics]. In a moderate external field up to $H_{ext} = 15$ kA/m, which is below the Rosensweig threshold, the constant susceptibility approximation holds with an error better than 4%. At this field strength the perturbation parameter given by Eq. (51) is smaller than 0.06. For a quantitative investigation of the magnetodissipative effect it is therefore sufficient to expand $\mathcal{D}(k,\omega)$ up to first order in κ giving

$$\mathcal{D}(k,\omega) = \omega^2 - \omega_0^2 + X(k,\omega) + \kappa X_M(k,\omega,H_{ext}) + O(\kappa^2), \tag{55}$$

with the magnetodissipative contributions

$$X_M^{mes}(k,\omega,H_{ext}) = (\nu k^2)^2\left\{\frac{i\omega}{2\nu k q} - \frac{\chi}{2+\chi}\left\{\frac{q}{k} - 1\right\}\right\} \tag{56}$$

in the mesoscopic theory and

$$X_M^{hyd}(k,\omega,H_{ext}) = \frac{2i\omega\nu k^2}{2+\chi} + 2(\nu k^2)^2\left(\frac{q}{k} - 1\right) \times \left[\frac{4+\chi}{2+\chi} + 2i\frac{\nu k^2}{\omega}\right] \tag{57}$$

in the hydrodynamic approach. We observe a different scaling behavior with the shear visosity ν. The magnetodissipative correction according to Eq. (56) scales with $\nu^{3/2}$, while it is only proportional to ν in Eq. (57). This deviating proportionality traces back to the constitutive equations. According to Eq. (18) magnetodissipation is associated with rotational flow and thus confined to the thin boundary layer beneath the surface, where damping is proportional to $\nu^{3/2}$ [24]. This is in contrast to Eq. (19), where the dissipative field $\mathbf{H}^D$ is finite over the whole fluid layer. In particular, dissipation within the convective bulk, which is known [25] to scale with ν, provides the leading contribution to Eq. (57).

Due to the smallness of κ, the magnetodissipative contribution $\kappa X_M(k,\omega)$ modifies the ordinary viscous shear damping $X(k,\omega)$ only slightly. Moreover, when studying the field dependence of the damping rate $\omega''(k,H_{ext})$ one has to account for appreciable wave-number shifts resulting from the nondissipative magnetic contribution

$$-\frac{\mu_0}{\rho}\frac{\chi^2}{(1+\chi)(2+\chi)}H_{ext}^2 k^2, \tag{58}$$

in $\mathcal{D}(k,\omega)$. Since $X(k,\omega)$ in turn is largely k dependent, the reactive term (58) strongly feeds back into the effective field dependence of $\omega''(H_{ext},k)$ and therefore masks the tiny

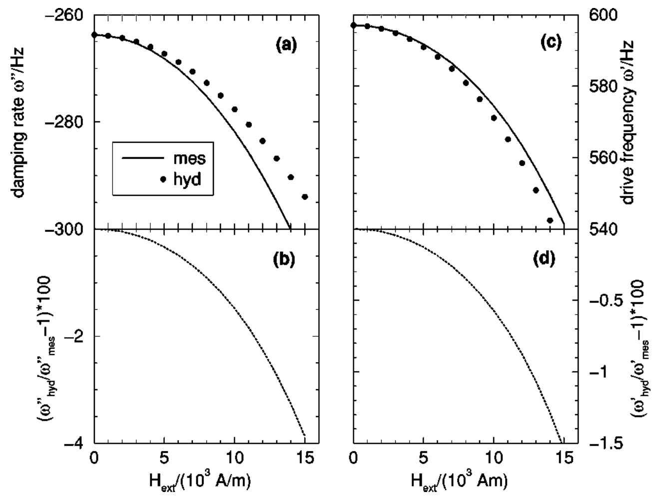

FIG. 2. (a) The decay rate $\omega''(k,H_{ext})$ for free waves on the surface of a ferrofluid exposed to a normal magnetic $\mathbf{H}_{ext}$. Solid lines (dots) denote the mesoscopic (hydrodynamic) theory. For the maximum investigated field intensity $H_{ext}=15$ kA/m the relative deviation (b) does not exceed 4%. (c), (d) demonstrate how the wave excitation frequency ω' must be adapted to keep the the inviscid contribution $\omega_0(k,H_{ext})$ constant during the variation of the field intensity. For the evaluation of the curves we used Eqs. (56) and (57) and the material specifications for the ferrofluid APG J12 with an estimated magnetic relaxation time of $\tau=3.8\times10^{-5}$ s.

magnetoviscous contribution X_M. To circumvent this difficulty one has to adapt the wave number k such that $\omega_0^2(k,H_{ext})$ as given by Eq. (29) is held constant when varying H_{ext}. This can be accomplished by simultaneously tuning the wave excitation frequency ω' during the H_{ext}-scan. Figure 2(a) depicts the field dependence of ω'' evaluated according to this protocol. As expected, the damping rate increases with the field strength. The predictions of the two approaches deviate by less than 4% [Figs. 2(a) and 2(b)]. With regard to the approximations made and the uncertainty of the material specifications this tiny difference is presumably too small to be resolved in a surface wave experiment. For completeness we show in Figs. 2(c) and 2(d) how the excitation frequency ω' must be adapted to guarantee a constant ω_0 during the variation of the field intensity.

V. SUMMARY AND DISCUSSION

The aim of the present paper was to compare two different theoretical approaches to the problem of magnetic field dependent dissipation in ferrofluids. On the one hand, we considered the theory given by Shliomis resting on a mesoscopic treatment of the rotation of the magnetic particles. On the other hand we applied the hydrodynamic Maxwell theory advocated by Liu which treats field dissipation within the standard framework of irreversible thermodynamics. For experimental arrangements in which the magnetic relaxation processes can be considered infinitely fast, the two descriptions are equivalent. This is, e.g., the case if the ferrofluid is composed of ferromagnetically soft particles, where the associated magnetic relaxation is usually very rapid as it is dominated by the fast Néel process.

On the other hand, for ferrofluids composed of materials with a high specific magnetic anisotropy (e.g., elementary cobalt or cobalt ferrites) the magnetic relaxation process is largely dominated by the Brownian rotation of the particles in the carrier matrix. In high viscosity carrier fluids the associated Brownian relaxation time can be large and reach the time scale of the hydrodynamic motion. This is the situation in which magnetodissipation must be accounted for. We have shown that this effect is treated differently by the approaches of Shliomis and Liu due to the use of different stress tensors as well as distinct constitutive equations for the dissipative fields.

In a ferrofluid layer at rest the constitutive equations coincide. If the surface is exposed to an oblique linearly polarized magnetic field, the differing stress tensors lead to different statements about whether the resting ferrofluid is in equilibrium or not: Whereas the mesoscopic approach gives no deviation from equilibrium the hydrodynamic theory predicts an onset of convection. We expect these predictions to be easy to verify or falsify in an experiment.

On the other hand, an experimental check of the validity of the constitutive equations is probably more subtle. The magnetodissipative contribution is usually very small as compared with the ordinary shear dissipation. Clearly, it becomes most pronounced if the ferrofluid is in solid body rotation, where shear dissipation drops out. Unfortunately, this special flow geometry—when introduced into the constitutive equations of Shliomis and Liu—leads to the *same* dissipative fields. We therefore looked for a nonrotational flow geometry, which is easy to realize and which fulfills the requirements of small amplitude low frequency motion. As a possible candidate we propose the flow field of free surface waves. On the basis of the two theoretical approaches at hand we worked out the associated complex dispersion relations. In the final expressions for the magnetodissipative contribution we observe a different scaling behavior with the shear viscosity ν, which could be traced back to the deviation between the constitutive equations. On the basis of the material specifications for a real ferrofluid and the experimental parameters for a low frequency surface wave experiment we computed the damping rate and evaluated the difference between the predictions. Unfortunately, the resulting deviation is less than 4% and therefore smaller than the expected experimental error.

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APPENDIX

In this Appendix we sketch the derivation of the dispersion relations for surface waves within the two theoretical approaches.

1. The mesoscopic approach

Due to Eq. (42) we introduce the scalar magnetic potentials by

$$\mathbf{H}=-\nabla\psi,\quad \mathbf{H}^{(2)}=-\nabla\psi^{(2)}, \tag{A1}$$

which obey the following relations:

$$\nabla^2\psi=-\frac{\chi\tau}{2(1+\chi)}H_0\nabla^2 w,\quad \nabla^2\psi^{(2)}=0. \tag{A2}$$

Equation (A2) has been derived by using Eq. (6) and the constitutive equation (18). Invoking once more the low frequency approximation, the latter occurs in the form

$$\mathbf{m}=\chi\mathbf{h}+\delta\mathbf{M}=\chi\left[\mathbf{h}+\frac{\tau}{2}(\nabla\times\mathbf{v})\times\mathbf{H}_0\right]. \tag{A3}$$

The potentials ψ and $\psi^{(2)}$ are solved by

$$\psi=-\frac{\tau\chi}{2(1+\chi)}H_0 w+\overline{\psi}e^{kz}e^{i(kx-\omega t)}, \tag{A4}$$

$$\psi^{(2)}=\overline{\psi}^{(2)}e^{-kz}e^{i(kx-\omega t)}. \tag{A5}$$

The boundary conditions (44), (45) yield the as yet unknown prefactors

$$\overline{\psi}=-\left\{\frac{\chi}{2+\chi}M_0-\frac{\tau\chi}{2+\chi}H_0\left[\nu k^2\left(1-\frac{\tilde{q}}{k}\right)-i\omega\right]\right\}\overline{\zeta} \tag{A6}$$

$$\overline{\psi}^{(2)}=\left[\frac{1+\chi}{2+\chi}M_0+\frac{\tau\chi}{2+\chi}H_0\nu k^2\left(1-\frac{\tilde{q}}{k}\right)\right]\overline{\zeta}, \tag{A7}$$

where $\tilde{q}=\sqrt{k^2-i\omega/[\nu(1+\kappa/4)]}$. From Eq. (A4) and the expressions for the coefficients (A6) and (A7) it can be seen that the solution of the magnetic field equations couple to the velocity field w. This coupling is removed in the absence of the magnetodissipation ($\tau=0$) and the result of the standard inviscid calculation [1] is recovered. These considerations complete the solution of the magnetic field problem in terms of the hydrodynamic variables.

We now turn to the solution of the hydrodynamic problem. From the linearized equations (39) and (40) we eliminate the gradient terms by operating twice with curl on the Navier-Stokes equation. Projecting on the z axis and using again Eq. (A3) we find

$$\left[\rho\partial_t-\left(1+\frac{\kappa}{4}\right)\eta\nabla^2\right]\nabla^2 w=0. \tag{A8}$$

Introducing again the plane-wave ansatz for the lateral dependence of w, the vertical dependence is determined by

$$(\partial_z^2-k^2)(\partial_z^2-\tilde{q}^2)\overline{w}=0, \tag{A9}$$

with

$$\tilde{q}^2=k^2-\frac{i\omega}{\nu(1+\kappa/4)}. \tag{A10}$$

The proper solution of Eq. (A9) remaining bounded for $z\to-\infty$ is

$$\overline{w}(z)=Ae^{kz}+Be^{\tilde{q}z}, \tag{A11}$$

with the so far undetermined coefficients A and B. It is always understood that the real part of $\tilde{q}$ is positive. Using the balance equation for normal stresses (47) and the kinematic surface condition (46) allows for us to express A and B in terms of $\overline{\zeta}$, resulting finally in Eq. (51).

2. The hydrodynamic approach

Again Eq. (42) allows to introduce scalar magnetic potentials

$$\mathbf{H}^R+\mathbf{H}^D=-\nabla\psi, \tag{A12}$$

$$\mathbf{H}^{(2)}=-\nabla\psi^{(2)}. \tag{A13}$$

The linearization of Eq. (11) is in the present case

$$\mathbf{H}^D=\frac{\alpha}{\mu_0}\partial_t\mathbf{B}-\alpha H_{ext}\partial_z\mathbf{v}. \tag{A14}$$

From this equation and Eq. (41) we find that due to the incompressibility of the flow, the magnetic potentials must be harmonic. Therefore

$$\psi=\overline{\psi}e^{kz}e^{i(kx-\omega t)}, \tag{A15}$$

$$\psi^{(2)}=\overline{\psi}^{(2)}e^{-kz}e^{i(kx-\omega t)}. \tag{A16}$$

Using the boundary conditions (44) and (45) we find

$$\overline{\psi}=\frac{\frac{\alpha}{k}\mu_r H_{ext}\partial_z\overline{w}(0)-(1-i\mu_r\alpha\omega)\overline{\zeta}M_0}{1+\mu_r-i\mu_r\alpha\omega}, \tag{A17}$$

$$\overline{\psi}^{(2)}=\frac{\frac{\alpha}{k}\mu_r H_{ext}\partial_z\overline{w}(0)+\mu_r\overline{\zeta}M_0}{1+\mu_r-i\mu_r\alpha\omega}, \tag{A18}$$

where $\partial_z\overline{w}(0)$ denotes the derivative of $\overline{w}(z)$ at $z=0$ and $\mu_r=1+\chi$. Again it can be seen that the solution of the magnetic field equations couples to the velocity field w. In the absence of the magnetodissipation ($\alpha=0$) this coupling is removed and the result of the standard inviscid calculation [1] is recovered. Moreover, considering the low frequency approximation $\alpha\omega\ll 1$ one finds that the *reactive* magnetic field $\mathbf{H}^R$ is exactly the same as in the absence of field dissipation. Eliminating from the linearized equations (39) and (40) the gradient terms by operating twice with curl on the Navier-Stokes equation and projecting on the z axis we find

$$(\rho\partial_t-\eta\nabla^2)\nabla^2 w=-\alpha H_{ext}\partial_t\partial_z\nabla^2 b_z+\alpha\mu_0 H_{ext}^2\partial_z^2\nabla^2 w. \tag{A19}$$

From the fact that ψ is harmonic, $\nabla^2 b_z$ can be replaced by

$$\nabla^2 b_z=\mu_0\mu_r\frac{\alpha H_{ext}}{1-i\mu_r\alpha\omega}\partial_z\nabla^2 w. \tag{A20}$$

Using again the plane-wave ansatz for the lateral dependence of w, the z dependence is determined by

$$(\partial_z^2-k^2)(\partial_z^2-\hat{q}^2)\overline{w}=0, \tag{A21}$$

with

$$\hat{q}^2=\frac{k^2-\frac{i\omega}{\nu}}{1+\kappa}, \tag{A22}$$

with the real part of $\hat{q}$ positive. The proper solution for $\overline{w}(z)$ remaining bounded for $z\rightarrow-\infty$ is hence

$$\overline{w}(z)=Ae^{kz}+Be^{\hat{q}z}, \tag{A23}$$

with the so far undetermined coefficients A and B. Using now $\alpha\omega\ll 1$ and expressing $\overline{\zeta}$ in terms of A and B by using the kinematic boundary condition the requirement of nontrivial solutions in A and B for the two stress boundary conditions results finally in Eq. (52).

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