Sound damping in ferrofluids: Magnetically enhanced compressional viscosity

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I. INTRODUCTION

Ferrofluids are colloidal suspensions of mono- or subdomain ferrimagnetic nano-sized particles suspended in a carrier liquid. Under the influence of an external magnetic field the fluid behaves paramagnetically. Among the more remarkable flow phenomena of ferrofluids are the enhanced effective shear viscosity in a static magnetic field or the viscosity decrease in response to an AC-field. Both are due to the so-called magneto-dissipative effect, which occurs when the experimental time scale compares to the magnetic relaxation time. In those situations the actual magnetization deviates significantly from its equilibrium value . Then the increment feeds back to the linear momentum balance, via the magneto-viscous stress element

\[
\Delta \Pi_{ij} = \frac{\mu_0}{2} \varepsilon_{ijk} [\mathbf{H} \times (\mathbf{M} - \mathbf{M}^\text{eq})]_k,
\]

leading to the appearance of an enhanced shear viscosity.

So if are the only magneto-dissipative term, one must conclude that sound in magnetized ferrofluids does not experience any additional damping, but this is incorrect: The recently derived ferrofluid dynamics contains, in addition to the expression of Eq. , also a new, diagonal magneto-viscous stress element, which accounts for the additional energy loss of sound waves if the medium is magnetized. It is natural to interpret this fact as a magnetically enhanced compressional viscosity, in close analogy to the magnetically enhanced shear viscosity first observed by McTague. Note that although this term was derived in as a stringent result of energy and momentum conservation, it is not contained in the standard ferrofluid-dynamics.

While the well-known tensor element of is non-vanishing only if the deviation and the field point in different directions, the new, diagonal stress element—writable as \( \mathbf{H} \cdot (\mathbf{M} - \mathbf{M}^\text{eq}) \delta_{ij} \) in the case of linear constitutive relationship—remains non-vanishing even if both are parallel to each other. This is exactly the situation characteristic for the propagation of sound. Provided the sound frequency does not greatly exceed the inverse magnetic relaxation \( \tau \) of the ferrofluid, a perceptible extra damping is predicted, several orders of magnitude larger than estimated by previous works.

It is worth pointing out that the new diagonal stress element may of course be disregarded in incompressible flow situations. In these cases the pressure \( p(\mathbf{r}, t) \) is determined by the condition \( \nabla \cdot \mathbf{v} = 0 \). A diagonal stress element such as the above term then re-normalizes \( p \), but leaves the velocity profile \( \mathbf{v}(\mathbf{r}, t) \) unchanged. Consideration of flow configurations such as those in sound, on the other hand, require the elimination of the incompressibility condition. This is done by adding the continuity equation for mass and adopting a viscous term proportional to \( \nabla \cdot \mathbf{v} \) in the Navier-Stokes equation.

\[ 
\]
The concept of pressure is rather ill-defined. The fact that the energy spent (or gained) by compressing a magnetized ferrofluid depends on the direction at which the force is applied implies that one needs to employ the more general concept of stress, and must especially be careful in handling the diagonal stress, as will be outlined below.

Several recent investigations on sound propagation in magnetizable fluids follow a more mesoscopic approach, considering relative particle motions within clusters, aggregates or chains. Taketomi attributes the anisotropy of the sound attenuation coefficient to two types of motions performed by the ferrous colloidal particles in the fluid, rotational and translational. Nahmad-Molnari et al. investigated the propagation of collective modes through a magnetorheological slurry (with micron-sized grains) and observed two independent modes. The slower one, with a large amplitude, was considered to be a compressional mode similar to what is found in porous fluid-saturated media. Later, Brand and Pleiner attributed this mode to a wave propagating along the particle chains.

Others authors pursue a macroscopic, hydrodynamic treatment similar to the approach employed here. Parsons considered ferrofluids subject to strong magnetic fields and took the vector of saturated magnetization as similar to the director in nematic liquid crystals. As a result, he found that magnetically induced relative corrections to the sound velocity are small, around $10^{-5}$. As discussed by Henjes, he worked with a purely mechanical pressure ignoring electromagnetic contributions in the diagonal stress. Using proper hydrodynamics, Henjes found that the magnetically induced corrections to the sound velocity are small, again of order $10^{-5}$. She also argued that since typical magnetic relaxation times are of the order of $\tau \approx 10^{-6}s$, magneto-dissipation is small for acoustic sound frequencies up to $\omega/(2\pi) = 20 \text{kHz}$. This is correct, but it does not imply that magneto-dissipation can be entirely ignored, because all dissipation mechanisms derive from fast characteristic times, and the question is one of relative weight. Summarizing, previous theoretical investigations on sound propagation in ferrofluids do not account for magneto-dissipation. The present manuscript does, and the result is: In the hydrodynamic frequency regime, $\omega \tau \approx 1$, even moderate magnetic fields will induce extra damping of approximately 10%.

II. THE STARTING EQUATIONS

To quantify the damping in compressional flow situations, propagation of sound waves through homogeneously magnetized ferrofluids will be investigated. To streamline the consideration and to focus on the basic physics, we shall implement the following simplifications:

(i) Only the leading order magnetic field effect $O(H^2)$ on the attenuation of sound is considered. This especially implies the linear constitutive relation, $\mathbf{M}^\text{eq} = \chi \mathbf{H}$. Moreover, the complication that sound in magnetized ferrofluids is generally accompanied by shear waves need not be considered. Although this coupling gives rise to rather surprising phenomena, it contributes only at $O(H^4)$ to the dispersion of sound (see below). (iii) We consider sound propagation and shear diffusion in the adiabatic limit. Adiabaticity means $\delta \equiv \delta(s/\rho) = 0$, rather than $\delta T = 0$ as is the case in the isothermal limit. Adiabaticity is valid because in ferrofluids, shear diffusion and sound are usually fast processes on the time scale of heat conduction: The Prandtl number $P$, given by the quotient of characteristic thermal diffusion time over viscous diffusion time, or equivalently, by kinematic viscosity over heat diffusivity, $P = \nu/\kappa$, is usually of the order of 10-100. (Depending on the ferrofluid, we have $\nu \approx 10^{-6} - 10^{-3} m^2/s$, and $\kappa \approx 10^{-7} - 10^{-5} m^2/s$.) The same argument holds for solutal diffusion processes which are slower than shear diffusion and sound by a factor $P/L$, where $L$ is the Lewis number. For ferrofluids we typically have $L = O(10^{-4})$.

Below it will be discussed in more details that the magnetic susceptibility $\chi$, usually taken as a function of $T$ and $\rho$, must then be considered as a function of entropy per unit mass $s$, in addition to $\rho$. Adiabaticity is a valid approximation here because in ferrofluids, shear diffusion and sound are usually fast processes on the time scale of heat conduction. (iv) Sound waves up to the MHz-range are weakly damped. The spatial decay length $\alpha^{-1}$ of the complex wave number $k = \omega/c + i\alpha$ exceeds the wave number $2\pi c/\omega$ by many orders of magnitude. Under those circumstances it is the custom to account for all damping mechanisms to linear order. (v) This paper focuses on sound attenuation. The tiny correction to the sound velocity is disregarded. An order of magnitude estimate for the magnetically induced correction yields $\Delta c \simeq (\mu_0 \chi H^2/\rho)^{1/2}$. Even at the highest magnetic field strength considered here, one gets $\Delta c/c < 10^{-4}$.

The unperturbed state of the ferrofluid is given by a homogeneously magnetized ferrofluid at rest, with density $\rho$ and equilibrium magnetization $\mathbf{M}^\text{eq} = \chi \mathbf{H}$, where $\chi$ is the magnetic susceptibility. To describe small amplitude sound excitations we introduce deviations from this state $\delta \rho$, $\mathbf{v}$, $\delta \mathbf{H}$, $\delta \mathbf{B}$ and $\delta \mathbf{M}$ proportional to a plane wave with wave vector $\mathbf{k}$. In particular the velocity field is taken as a longitudinal sound mode in the form

$$\mathbf{v} \propto \frac{k}{\Delta} e^{i (\mathbf{k} \cdot \mathbf{r} + \omega t)}. \tag{2}$$

The equations of motion governing the ferrofluid dynamics have recently been derived on the basis of the conservation laws and symmetries. The density field $\rho(\mathbf{r}, t)$ obeys as usual the continuity equation

$$\partial_t \rho + \nabla_j (\rho u_j) = 0. \tag{3}$$

The equation for the magnetization reads

$$\frac{d}{dt} M_i - \lambda_1 M_i v_{i4} - \lambda_2 M_j v^0_{ij} + (\mathbf{M} \times \Omega)_i = \sum_{\mu_0 \tau} - \frac{\mathbf{v}}{\mu_0 \tau} h_i, \tag{4}$$
where $d/dt = \partial_t + \mathbf{v} \cdot \nabla$ and $\mathbf{\Omega} = (\nabla \times \mathbf{v})/2$ is the vorticity of the flow. The contributions proportional to $\lambda_1$ and $\lambda_2$ appear with the applied field breaking the isotropy of the system. These two terms reflect the fact that in addition to the vorticity $\mathbf{\Omega}$—compressional and elongational flow, denoted respectively as $v_i = \nabla \cdot \mathbf{v}$ and $v_i^{\perp} = \frac{1}{2}(\nabla v_i + \nabla v_i - \frac{2}{3} \delta_{ij} v_{kk})$, contribute to the dynamics of $\mathbf{M}$. Further terms associated with the uniaxial symmetry (see the terms proportional to $\lambda_3$ and $\lambda_4$ in Eq. (13) of Ref. [9]) have been omitted on the left hand side of Eq. (3), as they are of higher order in the magnetic field. The increment

$$-\frac{\lambda}{\mu_0 \tau} \mathbf{h} = -\frac{\lambda}{\mu_0 \tau} (\mathbf{B}^\text{eq} - \mathbf{B}) = -\frac{1}{\tau} (\mathbf{M} - \chi \mathbf{H}), \quad (5)$$

with $\mathbf{B}^\text{eq} = \mu_0 \mathbf{M}(1 + \chi)/\chi$, accounts for magneto-dissipative relaxation, on the time scale given by $\tau$.

The magnetic field variables $\mathbf{H}, \mathbf{B}, \mathbf{M}$ are defined in SI-units as usual, with $\mu_0$ the vacuum permeability. For the evolution of the magnetic fields we adopt the static Maxwell equations $\nabla \cdot \mathbf{B} = \nabla \times \mathbf{H} = 0$. With the plane wave behavior similar to Eq. (2) the fluctuations are related in the following manner

$$\delta \mathbf{H} = -\delta \mathbf{M}_\parallel; \quad \delta \mathbf{B} = \mu_0 \delta \mathbf{M}_\perp. \quad (6)$$

Here the indices $\parallel$ and $\perp$ refer to the respective directions relative to the propagation direction $\mathbf{k}$ of the wave.

The balance equation for the linear momentum reads $\partial_t \rho \mathbf{v}_i + \nabla \rho \Pi_{ij} = 0$, with the stress tensor

$$\Pi_{ij} = -\rho \mathbf{v} s + \rho \mathbf{H} \cdot \mathbf{B} \quad (7)$$

$$+ (\lambda_1 - \frac{\lambda_2}{3}) h \cdot \mathbf{M} \delta_{ij} - \Pi_{ij}^\text{vis} + H_i B_j$$

$$- \frac{\lambda_2}{2} (M_i h_j + M_j h_i) + \frac{1}{2} (h_i M_j - h_j M_i).$$

To avoid misunderstandings of what is meant by the "pressure at non-zero magnetic field strength", the diagonal element is written in terms of the density of total energy $\rho$, the entropy density $s$, and the chemical potential $\mu$. The viscous stresses $\Pi_{ij}^\text{vis} = 2\eta \mathbf{v}^{\parallel}_i \mathbf{v}^{\parallel}_j + \eta \delta_{ij} \mathbf{v}_{kk}$ are taken as usual with the shear viscosity $\eta_1$ and the volume viscosity $\eta_2$. The terms proportional to $\lambda_1,2$ are counter terms to those of Eq. (3), they are constrained by the Onsager symmetry relations.

To make contact to previous formulations of the stress tensor [12] we have to switch for a moment to $T$ rather than $s$ as an independent variable. Then the square bracket in Eq. (6) can be recast in terms of the thermodynamic relation for the pressure at zero magnetic field $p_0(\rho, T)$

$$p_0 + \mu_0 \frac{H^2}{2} = (\lambda_1 - \frac{1}{3} \lambda_2 + 1) \mathbf{h} \cdot \mathbf{M} + \mu_0 M_\parallel^2 \frac{(1 - \rho \frac{\partial \chi}{\chi \partial \rho})}{2 \chi}, \quad (8)$$

where $\chi = \chi(\rho, T)$. Note that magneto-dissipation, proportional to $\mathbf{h} \cdot \mathbf{M}$, remains finite even if the transport coefficients $\lambda_1$ and $\lambda_2$ (not yet measured) should be negligibly small. As outlined in Sec. [13] this term arises cogently during the derivation of the stress tensor and accounts for magneto-dissipative processes if $\mathbf{h}$ is parallel to the equilibrium magnetization $\mathbf{M}^\text{eq}$. This is crucial for situations where $\mathbf{M}$ and $\mathbf{H}$ oscillate co-linearly but with a temporal phase lag. (Recall that the customary magneto-dissipative term given by Eq. (4) drops out if $\mathbf{M}$ and $\mathbf{H}$ are parallel to each other.)

### III. RESULTS

#### A. Dispersion of isothermal sound waves

We now return to the adiabatic formulation, where $\chi = \chi(\rho, s)$. Using Eq. (3) and the longitudinal plane wave velocity field [2] the magnetization fluctuation $\delta \mathbf{M} = \delta \mathbf{M}_\parallel + \delta \mathbf{M}_\perp$ is related to the density variations by

$$\delta \mathbf{M}_\parallel = \mathbf{M}_\parallel \left\{ \frac{(\rho/\chi) \partial \chi/\partial \rho - i \omega \tau (\lambda_1 + \frac{2}{3} \lambda_2)}{1 + \chi + i \omega \tau} \right\} \frac{\delta \rho}{\rho}, \quad (9)$$

$$\delta \mathbf{M}_\perp = \mathbf{M}_\perp \left\{ \frac{(\rho/\chi) \partial \chi/\partial \rho - i \omega \tau (\lambda_1 - \frac{1}{3} \lambda_2)}{1 + i \omega \tau} \right\} \frac{\delta \rho}{\rho}. \quad (10)$$

The real and imaginary part of $\delta \mathbf{M}$ is associated with magnetically induced corrections to the sound velocity and attenuation, respectively. Substituting Eqs. (9,10) into the divergence of the momentum balance yields the following complex dispersion relation for sound waves in magnetized ferrofluids

$$k = \frac{\omega}{c_s} + i \frac{\omega^2}{2 p c_s^3} \left( \frac{4}{3} \eta_1 + \eta_2 + \eta_m \right), \quad (11)$$

where $c_s^2 = \partial p_0(\rho, s)/\partial \rho$ is the square of the zero-field adiabatic sound velocity. Recall that – according to approximation (v) – magnetic corrections of $c_s$ are disregarded. The increment $\eta_m$ is given by

$$\eta_m = \mu_0 \tau H^2 \left[ \frac{\kappa_\parallel \cos^2 \theta}{(1 + \chi)^2 + (\tau \omega)^2} + \frac{\kappa_\perp \sin^2 \theta}{1 + (\tau \omega)^2} \right], \quad (12)$$

where

$$\kappa_\parallel = \left[ \frac{\rho \partial \chi}{\chi \partial \rho} + (1 + \chi) (\lambda_1 + \frac{2}{3} \lambda_2) \right]^2, \quad (13)$$

$$\kappa_\perp = \left[ \frac{\rho \partial \chi}{\chi \partial \rho} + (\lambda_1 - \frac{1}{3} \lambda_2) \right]^2. \quad (14)$$

$\eta_m$ can be interpreted as a "magnetic extra viscosity". The expression for $\eta_m$ is clearly anisotropic, and $\theta$ denotes the angle between the applied magnetic field $\mathbf{H}$ and the direction $\mathbf{k}$ of propagation. Note also that $\eta_m$ is
frequency dependent, being maximal at \( \tau \omega \to 0 \) and vanishing in the high frequency limit \( \tau \omega \gg 1 \). Eqs. (13,14) can be simplified if the magnetic susceptibility is proportional to the density thus \( \rho \partial \chi / \partial \rho \approx 1 \). For a rough estimate, take the data \( \chi \) for a high viscosity hydrocarbon based ferrofluid (APG 933, Ferrofluidics): \( \chi \approx 1.1, \eta_1 \approx 0.5 \text{PAs} \), and \( \tau \approx 0.55 \text{ms} \), in addition to \( \lambda_1 = \lambda_2 = \eta_2 = 0 \) (for lack of better information). Then a 100Hz-sound wave propagating in an applied magnetic field of say \( H = 10^4 \text{A/m} \) (this is a field strength at which most ferrofluids still obey linear constitutive relations) experiences a magneto-viscous extra damping of \( \approx 7\% \) at the parallel orientation \( \mathbf{k} \parallel \mathbf{H} \) (i.e. \( \theta = 0 \)) and almost 9\% at the transverse setup (\( \theta = 90^\circ \)). The same estimate applies to a ferrofluid of similar microscopic make-up, but at a viscosity of \( \eta_1 \approx 5 \text{mPAs} \) and a frequency of 10kHz. (Here we assume \( \tau \propto \eta_1 \), valid for Brownian particles, i.e. when the particle’s magnetic moment is fixed to the crystallographic orientation). Damping increments of this size should be detectable in a careful sound wave experiment. Moreover, by scanning the \( \theta \)-dependence of \( \eta_m \), it should be possible to obtain information on the transport coefficients \( \lambda_1 \) and \( \lambda_2 \).

**B. Adiabatic versus isothermal susceptibility**

Knowing the dependence of the magnetic susceptibility as a function of density and temperature, \( \chi(\rho, T) \), the derivative \( \rho \partial \chi / \partial \rho \) in the above equations can be expressed as follows

\[
\rho \frac{\partial \chi(\rho, s)}{\partial \rho} = \rho \frac{\partial \chi(\rho, T)}{\partial \rho} + T \frac{\partial \chi(\rho, T)}{\partial T} \frac{c_2^2 \alpha_v}{C_v},
\]

involving both the magneto-strictive and the magneto-caloric contributions. Here \( \alpha_v = -(1/\rho) \partial \rho(\rho, T)/\partial T \) denotes the thermal expansion coefficient, \( c_T \) the isothermal sound velocity, and \( C_v = T \partial s(T, \rho)/\partial T \) the specific heat at constant volume, of everyone of them evaluated at zero magnetic field. Eq. (17) indicates that adiabatic sound waves involve both magnetostrictive as well as magneto-caloric contributions. Here \( \alpha_v = -(1/\rho) \partial \rho(\rho, T)/\partial T \) denotes the thermal expansion coefficient and \( C_v = T \partial s(T, \rho)/\partial T \) the specific heat at constant volume, each of them evaluated at zero magnetic field. For a typical olefine-based carrier liquid the dimensionless factor \( c_2^2 \alpha_v / C_v \) can be estimated by 0.3.

**C. Enhanced compressional viscosity**

In order to classify the viscosity increment \( \eta_m \) [Eq. (12)] as an field dependent offset to either the shear viscosity \( \eta_1 \) or the volume viscosity \( \eta_2 \) we evaluate the entropy production in the present setup. Following Ref. [4] the total entropy production is given by

\[
R = 2 \eta_1 (\nabla \cdot \mathbf{v})^2 + \eta_2 (\nabla \cdot \mathbf{v})^2 + \frac{\chi}{\mu_0 \tau} h^2.
\]

Computing the magneto-viscous surplus [last term in (16)] up to first order in \( \omega \tau \) yields

\[
\frac{\chi}{\mu_0 \tau} h^2 = \mu_0 \tau \left[ \frac{\kappa_{\parallel} \cos^2 \theta}{(1 + \chi)^2} + \kappa_{\perp} \sin^2 \theta \right] (\nabla \cdot \mathbf{v})^2.
\]

The formal similarity of (17) with the second term of (14) suggests that the “magnetic extra viscosity” \( \eta_m \) according to Eq. (12) is to be interpreted as an enhanced compressional viscosity \( \Delta \eta_2(H) \).

**D. Comparison with experiments**

The experimental material on sound propagation in ferrofluids is rather scarce. The early measurements of Chung and Isler [8, 20] on a water-based ferrofluid seem to be the only available systematic study of the velocity and attenuation of sound (note, however, that Skujiel’s [21] later investigations reveal a strong dependence of the sound velocity on the type of the carrier liquid, i.e whether it is aqueous or organic). The experiments of Refs. [8, 20] were carried out with 2.25 MHz ultrasound, employing pulse-echo and continuous wave methods. The experimental data cover a wide magnetic field range from 0 up to 2500Gauss = 2 \times 10^5 \text{A/m}. Within the weak field subrange, where linear constitutive relations hold for most ferrofluids, \( H < 10^4 \text{A/m} \) say, the damping increment \( \alpha \) was found to increase by 1.8dB (\( \approx 20\% \)) at \( \theta = 0 \) but to decrease (anomalous sound attenuation) by almost 3.5dB (\( \approx 50\% \)) at \( \theta = 90^\circ \). Unfortunately no information was given whether demagnetization effects due to the cylindrical probe geometry had been taken into account here. However, we point out that the observed anomalous \( \theta \)-dependence does not even qualitatively comply with the present theory. Neither the observed history dependence of the experimental data (also detected by Gotoh et al. [22]) can be explained by the present approach. The latter peculiarities suggests that the small ultrasound wavelengths couple to microscopic inhomogeneities associated with particle chains or clusters in the ferrofluid suspension. Anisotropies, field dependencies and anomalies recorded by ultrasound experiments therefore seem to depend on mechanisms which are rather different from those covered by the present hydrodynamic analysis. Owing to the lack of other pertinent experimental data let us nevertheless try a quantitative comparison with the measurements of Isler and Chung [8]. For a rough estimate we take the viscosity of their aqueous ferrofluid by \( \eta_1 = 10^{-3} \text{ Pas} \) and the susceptibility by \( \chi \approx 1 \) (specifications are not given). Due to the high ultra-sound drive frequency the experiment was operated in the limit \( \omega \tau \gg 1 \) and thus the expected extra damping is quite small. Even if the magnetic relaxation time \( \tau \) is estimated to be as small as \( 10^{-6} \text{s} \), the prediction of Eq. (12) at \( \theta = 0 \) is two orders of magnitude smaller than the empiric value. We therefore conclude that for a reliable quantitative check...
of the present theory, experiments at acoustic frequencies (where $\omega \tau \simeq 1$) would be more suitable.

IV. DISCUSSION

The present analysis deals with the attenuation of sound in ferrofluids, which are exposed to a weak homogeneous magnetic field in any direction relative to the propagation. This has been accomplished by investigating the linear dispersion of a pure longitudinal velocity excitation. Recently, it has been pointed out \[17\] that density excitations (sound) and transverse velocity fluctuations (shear waves) in magnetized ferrofluids do not evolve separately as is the case at $H = 0$: At finite $H$, sound waves may produce shear excitations and vice versa. Clearly, if shear waves accompany sound this opens a new attenuation mechanism, which cannot be ignored. In the remainder of this section we shall argue why this cross-coupling remains without consequences for the present analysis: By taking the curl of the momentum balance one arrives at

$$\rho \partial_t \Omega - \eta \nabla^2 \Omega = \frac{1}{4} \nabla \times \nabla \times (h \times M). \quad (18)$$

Assuming that a sound emitter produces plane density waves $\delta \rho(t)$ within a magnetized ferrofluid, the right hand side of Eq. (18) can be recast as

$$\nabla \times \nabla \times (h \times M) = \frac{\tau \mu_0}{\chi} \frac{\partial \chi}{\partial \rho} \frac{M \perp \times M \parallel}{1 + \chi} \nabla^2 \delta \rho + \ldots,$$

thus acting as a magneto-dissipative source of vorticity $\Omega$. If the applied magnetic field is weak we have $\Omega \approx O(H^2)$. Via the term $\Omega \times M$ in Eq. (3) this sound-made vorticity induces a third order correction in $M$, which – at the considered accuracy level $O(H^2)$ – does not affect the sound dispersion. Note however, that a proper study of sound damping in strongly magnetized ferrofluids (as for instance undertaken in Ref. \[10\]) must not ignore the complication arising from the magneto-dissipative cross-coupling between compressional and shear excitations.

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