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Non-Newtonian behaviour in ferrofluids and magnetization relaxation

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Abstract

The relaxation of magnetization, a well-accepted part of ferrofluid dynamics, is shown to give rise to a broad range of non-Newtonian behaviour in ferrofluids, including shear thinning or shear thickening, normal stress differences, a visco-elastic response and a varying Trouton, or elongational, viscosity.

1. Introduction

Since its inception, the ferrofluid-dynamics has given us a concise understanding of a number of varying phenomena. The theory was derived by Shliomis assuming spherical, non-interacting magnetic particles, rotating against the viscosity of the carrier liquid. Consisting of two essential elements, (1) the relaxation equation for the magnetization $\mathbf{M}$ and (2) a torque $\frac{1}{2}\varepsilon_{ijk}(\mathbf{H}\times\mathbf{M})_k$ in the stress tensor Π_{ij}, the theory has been successfully applied in numerous circumstances, particularly negative viscosity [1, 2]. ($\mathbf{H}$, and $\mathbf{B}$ below, are the two magnetic fields.) Recently, denser and more strongly magnetized ferrofluids were found to display strikingly non-Newtonian behaviour, including shear thinning and normal stress differences [3]. Experiments, microscopic theories [4] and simulations [5] all show this to be the result of magnetic particles forming short chains in the presence of fields, making ferrofluids resemble polymer solutions. Facing the need for a similarly concise macroscopic theory for these ferrofluids, the general feeling is that its construction would require *'new physical ideas'* and *'concepts of polymer physics'* [6], and that the result would be a combination of the Shliomis theory with polymer fluid dynamics. There have been a number of rather useful microscopic results considering the influence of particle interaction on the dynamics [7], chain formation [4] and chain flexibility [8], but such a macroscopic theory has not yet been formulated.

In 2001, Müller and Liu [9] published a paper on the thermodynamic framework of ferrofluid dynamics, the main result of which is that ferrofluid dynamics as given by Shliomis can be divided into two parts: structure and coefficients. While the structure is determined by general principles that are always valid, the coefficients (such as the relaxation time) are the result of simplifying assumptions (especially the particle shape and lack of particle interaction), and are therefore much more restricted in their range of validity. This is a relevant insight, because it implies that one may take the same set of equations to account for any system having a slowly relaxing magnetization simply by choosing the appropriate coefficients depending

on particle shape (be it spherical or elongated) and the type of relaxation (Neél or Brown). Following this view to its logical end, one concludes naturally that the given set of equations also holds for chain-forming ferrofluids—although the chains as 'constituent particles' are rather elongated, and the relaxation rate of magnetization is a composite quantity, restricted not only by how fast the chains may be oriented but also by how quickly particles can be transported and assembled to form chains of the proper length.

It is hardly surprising that polymer solutions and chain-forming ferrofluids differ in some fundamental ways in their macroscopic dynamics. Being a negative statement, the term 'non-Newtonian' lacks specificity, and there may well be different versions of non-Newtonian behaviour (polymeric, ferrofluid one...) requiring different descriptions. Since polymer strands are entangled without shear, but get aligned along the flow by it [10], while magnetic chains are aligned along the field without shear, and are broken into pieces by it [3], their similarity must be rather restricted.

Although polymer solutions are amenable to many competing descriptions [10], they are in essence characterized by *transient elasticity*, with their rheology well accounted for by a relaxing strain field [11]. An important (and perhaps confusing) point here is that there are polymer-like magnetic fluids that also need the strain field as an extra variable: if the magnetic particles are large enough, they will, in the presence of a strong field, form long chains bridging the whole system. This is the jamming transition, after which the system is truly elastic [12], and needs the strain field for a description of its static and dynamic properties. Close to the transition, the strain should be a critical, relaxing variable, because two chains, neither quite bridging the system, temporarily get in the way of each other. This implies transient elasticity. Here we shall deal with magnetic fluids that are either incapable of the jamming transition, or far enough away from it, and refer to them as *ferrofluids*. The accepted term for magnetic fluids capable of forming long chains is *magneto-rheological fluids* (MRF), though a term such as *magnetic elastofluids* may be more descriptive.

The question we raise in this paper is whether the relaxation of magnetization suffices by itself to fully account for non-Newtonian ferrofluids forming short chains [13]. We do not aim for a full, quantitative answer to this conjecture, but only seek a qualitative answer, an understanding of whether the structure of ferrofluid dynamics is rich enough to support non-Newtonian behaviour. To do this, we consider a mono-disperse ferrofluid containing a single population of magnetic particles, all participating in chain formation. Such a ferrofluid is characterized by a single relaxation equation, that of the total magnetization. Furthermore, we assume a linear constitutive relation and incompressible flows, so fully analytical solutions for varying geometry and field orientation are possible. This is an important first step, because it establishes the fact, firmly and incontrovertibly, that some typical aspects of non-Newtonian behaviour are indeed the result of magneto-relaxation. In addition, the explicit solutions should also considerably ease the analysis and understanding of these aspects of ferrofluid non-Newtonian behaviour. On the other hand, the analytical results obtained below cannot be compared to experiments directly and quantitatively.

2. Ferrofluid dynamics

We start our consideration with the equations

$$\frac{\mathrm{d}}{\mathrm{d}t}M_i + (\mathbf{M}\times\boldsymbol{\Omega})_i - \lambda_2 M_j v_{ij} = -(M_i - M_i^{\mathrm{eq}})/\tau, \tag{1}$$

$$\Pi_{ij} = \tilde{P}\delta_{ij} - 2\eta_1 v_{ij} - H_i B_j + \tfrac{1}{2}[(M_i h_j - M_j h_i) - \lambda_2(M_i h_j + M_j h_i)]. \tag{2}$$

The first is the relaxation equation for the magnetization, the second (with v_i the velocity) gives the stress, defined by local momentum conservation $\dot{g}_i + \nabla_j(\Pi_{ij} + \rho v_i v_j) = 0$.

For $\lambda_2 = 0$, equation (1) describes magneto-relaxation in the co-rotating frame, as originally employed by Shliomis ($\boldsymbol{\Omega} \equiv \frac{1}{2}\nabla \times \mathbf{v}$ is the rate of rotation). Given our assumption of a linear constitutive relation, the equilibrium magnetization is simply $M_i^{\rm eq} = \chi H_i$. A finite λ_2, proposed in [9], accounts for the influence of an elongational flow, $v_{ij} \equiv \frac{1}{2}(\nabla_i v_j + \nabla_j v_i)$. It was measured using various ferrofluids by Odenbach and Müller, who obtained values between 0 and 0.88 [14], demonstrating its relevance. They also tentatively relate λ_2 to the chain length at zero shear, with $\lambda_2 \approx 0$ appropriate for single-particle ferrofluids, while 0.88 indicates chains averaging around five particles. (More coefficients, accounting for magnetic anisotropy, are permitted by symmetry, cf [9]. Although some may be necessary for quantitative comparisons with experiments, their inclusion here would only obfuscate the present, emphatically qualitative, consideration.)

The scalar $\tilde{P}$ in the total stress equation (2) contains all diagonal terms [9], not only the pressure P. It is not further specified, as it is relevant only for compressional flows such as considered in [15]. The transport coefficient η_1 is the shear viscosity without field, and the conjugate variable $\mathbf{h}$ is given as $\mathbf{h} \equiv \mathbf{M}/\chi - \mathbf{H} = \mathbf{M}\frac{1+\chi}{\chi} - \mathbf{B}$. (For nonlinear constitutive relations, we have $\mathbf{h} \equiv \mathbf{H}^{\rm eq} - \mathbf{H} = \mathbf{B}^{\rm eq} - \mathbf{B}$, where $\mathbf{H}^{\rm eq}(\mathbf{M})$ is the inverse of $\mathbf{M}^{\rm eq}(\mathbf{H})$, and $\mathbf{B}^{\rm eq}(\mathbf{M})$ that of $\mathbf{M}^{\rm eq}(\mathbf{B})$, see [9].) The two expressions preceded by λ_2 in equations (1) and (2),

$$\lambda_2 M_j v_{ij} = \tfrac{1}{2}\lambda_2 M_j(\nabla_i v_j + \nabla_j v_i), \qquad \tfrac{1}{2}\lambda_2(M_i h_j + M_j h_i),$$

are counter terms, which due to the Onsager reciprocity relation are necessarily present simultaneously. Similarly,

$$(\mathbf{M} \times \boldsymbol{\Omega})_i = \tfrac{1}{2}M_j(\nabla_i v_j - \nabla_j v_i), \qquad \tfrac{1}{2}(M_i h_j - M_j h_i),$$

are also counter terms, which share the material-independent coefficient of 1. Because $M_i h_j - M_j h_i = \varepsilon_{ijk}(\mathbf{M} \times \mathbf{h})_k$, and $\mathbf{h} = \mathbf{H}^{\rm eq} - \mathbf{H}$ with $\mathbf{H}^{\rm eq} \parallel \mathbf{M}$, the term $\frac{1}{2}(M_i h_j - M_j h_i) = -\frac{1}{2}\varepsilon_{ijk}(\mathbf{M} \times \mathbf{H})_k$ is the Shliomis torque. This torque is frequently falsely understood to be present only if magnetic grains rotate against the fluid matrix. Yet it is rather more general and always present. As mentioned above, it is the thermodynamically necessary counter-term to $\mathbf{M} \times \boldsymbol{\Omega}$ that merely transforms $\dot{\mathbf{M}}$ into the co-rotating local rest frame and has nothing to do with the difference between Neél and Brown relaxation.

The transport coefficients η_1, λ_2, τ are material-dependent parameters, and functions of thermodynamic variables such as density, concentration and temperature, but they are emphatically independent from shear. As discussed in [9], the dependence of transport coefficients on non-equilibrium quantities such as shear goes beyond the well-verified linear force-flux relation of Onsager, and may even lead to negative entropy production or violate other general principles. Moreover, it pre-empts any theory: simply taking the viscosity depending on shear and field as measured leaves no room, nor need, for a theory. Although a presumed shear dependence is an accepted and practical way to frame experimental results, see e.g. [14], real understanding can only be claimed if such dependence is eliminated by employing a more complete theory: experiments on magneto-vortical resonance [16] were accounted for by a shear-dependent relaxation time τ. Yet equal agreement with the data was achieved with a constant τ, after a finite λ_2 was introduced [9].

3. Different experimental situations

3.1. Shear thinning and shear thickening

Now we employ equations (1) and (2) to consider the first of four examples: simple shear, $\mathbf{v} = \dot{\gamma} y \hat{\mathbf{x}}$, with the velocity along $\hat{\mathbf{x}}$, the gradient along $\hat{\mathbf{y}}$, and the external field either perpendicular to the surface and along $\hat{\mathbf{y}}$ or parallel to the surface and $\hat{\mathbf{x}}$ (see figure 1). Since the

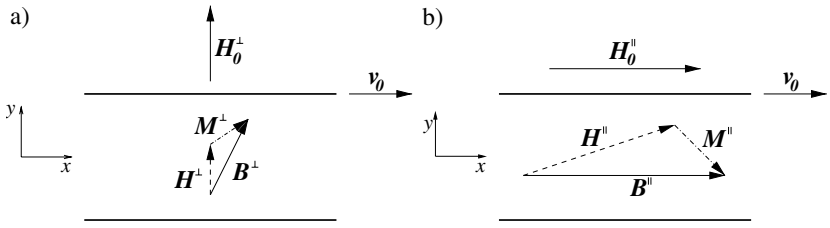

Figure 1. The orientation of the fields in a sheared ferrofluid, if the external field is either (a) perpendicular or (b) parallel to the surface.

equilibrium magnetization $\mathbf{M}^{\mathrm{eq}}$ is in the xy-plane in both cases, stationarity or $\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{M} = 0$ imply that equation (1) is a linear, 2×2 matrix equation, $\underline{\underline{A}}\mathbf{M} = \mathbf{M}^{\mathrm{eq}}$. Inverted, it reads

$$M_x = \frac{4M_x^{\mathrm{eq}} + 2\,(1+\lambda_2)\,\xi\,M_y^{\mathrm{eq}}}{4 + \left(1-\lambda_2^2\right)\xi^2}, \tag{3}$$

$$M_y = \frac{4M_y^{\mathrm{eq}} - 2\,(1-\lambda_2)\,\xi\,M_x^{\mathrm{eq}}}{4 + \left(1-\lambda_2^2\right)\xi^2}, \tag{4}$$

where

$$\xi \equiv \dot{\gamma}\tau \tag{5}$$

is the dimensionless shear rate. Equations (3) and (4) already contain the essence of shear thinning: the magnetization goes to zero in the limit of strong shear, $\xi \to \infty$, implying the vanishing of any magneto-viscous effect, because the second line of equation (2) also vanishes. This fact was partially recognized by Shliomis, who remarked in [2]: '*The reduction of the magnetization* [by shear] *leads in turn to some decrease in the rotational viscosity*'. We see that the magnetization is in fact eradicated, for arbitrary values of λ_2 except 1. Also there is considerable numerical correction from λ_2 at finite shear, especially if λ_2 approaches 1.

The force density on an infinitely extended plate in the xz-plane, being dragged along $\hat{\mathbf{x}}$ on top of a ferrofluid layer, is $\Delta\Pi_{xy} \equiv \Pi_{xy}^{\mathrm{air}} - \Pi_{xy}^{\mathrm{ff}}$. The stress of air, Π_{xy}^{air}, is $-H_x B_y$, and that of the ferrofluid, Π_{xy}^{ff}, is given by equation (2). (Because $H_x B_y$ is continuous, $\Delta\Pi_{xy}$ is calculated from the second line, in addition to the term $-2\eta_1 v_{ij}$.) Taking the total viscosity as $\eta_1 + \eta_r \equiv -\Delta\Pi_{xy}/\dot{\gamma}$, the magneto-viscous contribution η_r is evaluated by inserting equations (3) and (4) into (2) for the boundary condition of either perpendicular or parallel external field H_0.

Because of the typical geometry for viscometers, the experimentally more convenient configuration is given by $B_0 = H_0$ along $\hat{\mathbf{y}}$, perpendicular to the plate. As $B_y = H_0$, $H_x = 0$ are continuous, the internal fields are $\mathbf{B} = (M_x, H_0)$, $\mathbf{H} = (0, H_0 - M_y)$, with $\mathbf{M} = \mathbf{B} - \mathbf{H} = (M_x, M_y)$ and $\mathbf{M}^{\mathrm{eq}} = \chi\mathbf{H} = \chi(0, H_0 - M_y)$. Using these in equations (2), (3) and (4), we find

$$\eta_r^\perp = \frac{(1+\lambda_2)^2[4 + (1-\lambda_2)^2\xi^2]}{[4(1+\chi) + (1-\lambda_2^2)\xi^2]^2}\tau\chi H_0^2. \tag{6}$$

For vanishing shear, $\xi \to 0$, the viscosity $\eta_r^\perp$ grows with τ, χH_0^2 and λ_2. More generally, $\eta_r^\perp$ decreases monotonically with shear if

$$\chi \leqslant \frac{1+3\lambda_2}{1-\lambda_2}, \tag{7}$$

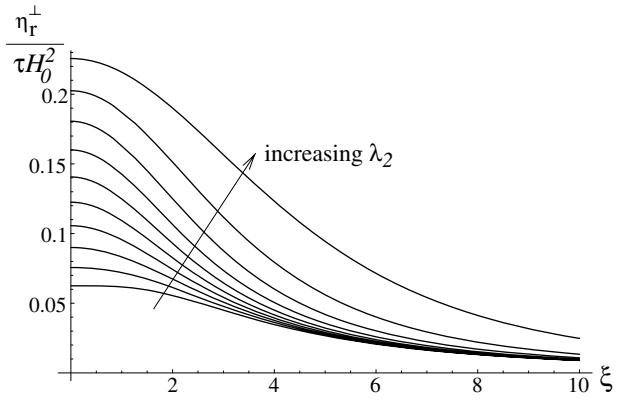

Figure 2. The magneto-viscous contribution to the shear viscosity, in units of τH_0^2, as a function of $\xi \equiv \dot{\gamma}\tau$, from $\lambda_2 = 0$ to $\lambda_2 = 0.9$ in steps of 0.1, for a perpendicular external field H_0, with χ =1. Shear thinning is obvious for the chosen parameters, but shear thickening is also possible.

and displays a maximum and partial shear thickening otherwise. Figure 2 shows a case in which equation (7) is satisfied, and the magneto-viscous contribution decays monotonically as a function of shear, for $\chi = 1$ and 10 different values of λ_2, from 0 to 0.9.

If the external field $H_0 = B_0$ is parallel to the plate, along $\hat{\mathbf{x}}$, again because B_y, H_x are continuous, the internal fields are $\mathbf{B} = (H_0 + M_x, 0)$, $\mathbf{H} = (H_0, -M_y)$, and $\mathbf{M}^{\rm eq} = \chi(H_0, -M_y)$. Using these in equations (2), (3) and (4), we find

$$\eta_{\rm r}^{\parallel} = \frac{(1-\lambda_2)^2\left[4\,(1+\chi)^2 + (1+\lambda_2)^2\,\xi^2\right]}{\left[4(1+\chi) + \left(1-\lambda_2^2\right)\xi^2\right]^2}\tau\,\chi\,H_0^2. \tag{8}$$

In the limit of low shear, $\xi \to 0$, $\eta_r^{\parallel}$ still grows with τ, χH_0^2, but now decreases with λ_2. (For $\lambda_2 = 0$ and $\chi \ll 1$, both shear viscosities are the same, $\eta_{\rm r}^{\perp} = \eta_{\rm r}^{\parallel}$, as they should be: this is the limit where the rotation of the magnetic particles against the fluid matrix holds, then any field orientation perpendicular to Ω is equivalent.) For finite shear, $\eta_r^{\parallel}$ decreases monotonically with ξ only if

$$\chi \geqslant \frac{3\lambda_2 - 1}{2(1-\lambda_2)} \tag{9}$$

is satisfied. In figure 3, this is the case only for $\lambda_2 \lesssim 0.6$. Assuming, in equations (7) and (9), that λ_2 lies between 0 and 1 (for which there are no general reasons, only model-independent ones), and χ is around 1, the monotonicity of the magneto-viscous contribution to the shear viscosity can be read off the map of figure 4.

The plots of figures 2 and 3 are of more conceptual interest, demonstrating shear thinning as a result of magneto-relaxation and quantifying the influence of λ_2. In an experiment, of course, λ_2 cannot be varied for a given field. Besides, both λ_2 and the relaxation time τ are functions of the external field, so $\eta/\tau H_0^2$ is not field-independent. But one may of course compare $\eta_r^{\perp}$ to $\eta_r^{\parallel}$ for given field, τ, and λ_2. Depending on λ_2, the normalized magneto-viscous shear contribution, as a function of the shear rate ξ, is given in figure 5.

Finally, we calculate the ratio between $\eta_r^{\perp}$ and $\eta_r^{\parallel}$ at vanishing shear,

$$\frac{\eta_{\rm r}^{\perp}\,(\xi \to 0)}{\eta_{\rm r}^{\parallel}\,(\xi \to 0)} = \frac{1}{(1+\chi)^2}\left(\frac{1+\lambda_2}{1-\lambda_2}\right)^2. \tag{10}$$

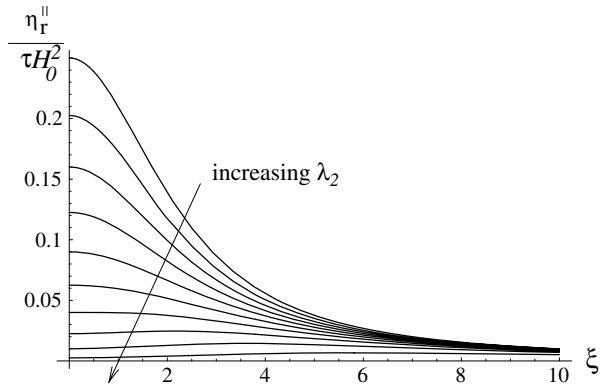

Figure 3. Magneto-viscous contribution to the shear viscosity as a function of $\xi \equiv \dot{\gamma}\tau$, from $\lambda_2 = 0$ to $\lambda_2 = 0.9$, for a parallel external field H_0, with $\chi = 1$. A growing λ_2 diminishes the effect, possibly because the chains are already aligned along the flow, in the same way as polymer strands at high shear. Monotonic shear thinning prevails only for $\lambda_2 \lesssim 0.6$.

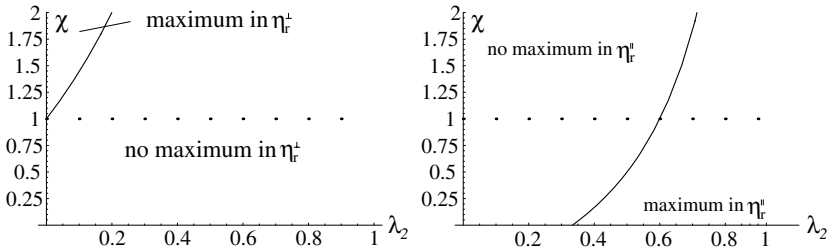

Figure 4. There is a maximum in the magneto-viscous contribution to the shear viscosity, and shear thickening occurs, if χ versus λ_2 lies to the left or right of the line, respectively. The dots represent parameters used for the above two figures for the perpendicular and parallel case.

3.2. Normal-stress differences

It is well known from polymer physics [10] that quite a number of non-Newtonian behaviours—including especially the Weissenberg (or rod-climbing) effect already seen in ferrofluids [3]—derive from finite normal-stress differences. As our second example, we therefore calculate normal-stress differences, $N_1 \equiv \Pi_{xx} - \Pi_{yy}$ and $N_2 \equiv \Pi_{yy} - \Pi_{zz}$. Note, however, that as a result of the Maxwell stress, N_1, N_2 are in general finite even for vanishing shear, $\xi = 0$, in ferrofluids. (An example is the magnetic force that leads to the elongation of spherical droplets.) We shall assume a geometry in which this contribution either vanishes or is compensated (say because the droplet is already elongated), and subtract from our result the normal stress difference at vanishing shear,

$$-\dot{\gamma}^2\Psi_1 \equiv N_1 \equiv \left[\Pi_{xx}(\dot{\gamma}) - \Pi_{yy}(\dot{\gamma})\right] - \left[\Pi_{xx}(0) - \Pi_{yy}(0)\right], \tag{11}$$

$$-\dot{\gamma}^2\Psi_2 \equiv N_2 \equiv \left[\Pi_{yy}(\dot{\gamma}) - \Pi_{zz}(\dot{\gamma})\right] - \left[\Pi_{yy}(0) - \Pi_{zz}(0)\right], \tag{12}$$

where Ψ_1, Ψ_2 are the so-called normal stress coefficients. They are calculated as

$$\Psi_1^{\perp} = -\frac{(1+\lambda_2)\left[4(1+\chi)(1-3\lambda_2) + (1+\lambda_2)(1-\lambda_2)^2\xi^2\right]\tau^2}{\left[4(1+\chi) + \left(1-\lambda_2^2\right)\xi^2\right]^2}\frac{\chi}{1+\chi}H_0^2, \tag{13}$$

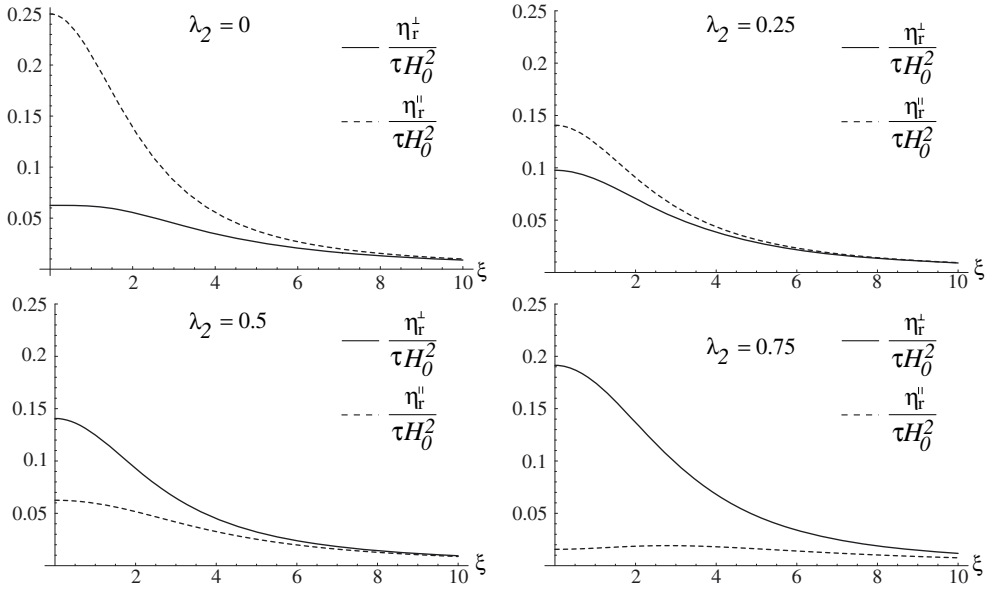

Figure 5. Comparing $\eta_r^{\perp}$ to $\eta_r^{\parallel}$ taking $\chi = 1$. Clearly, we have $\eta_r^{\perp} < \eta_r^{\parallel}$ for $\lambda_2 = 0$, but end up with $\eta_r^{\perp} > \eta_r^{\parallel}$ at larger values of λ_2. Both are equal for $\lambda_2 = 1/3$.

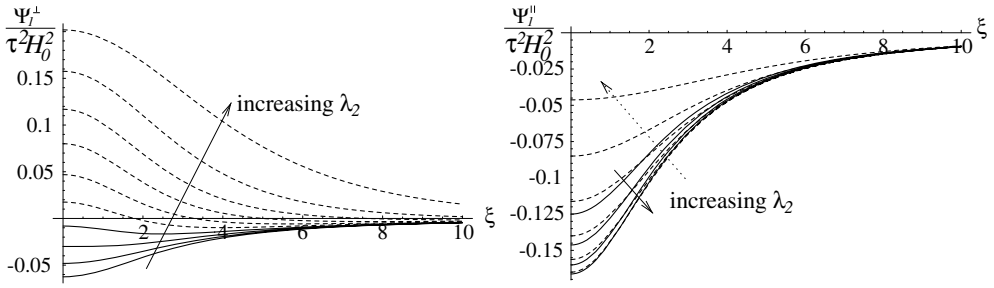

Figure 6. The first normal stress coefficients, $\Psi_1^{\perp}$ and $\Psi_1^{\parallel}$, respectively for normal and parallel external fields, in units of $\tau^2 H_0^2$, as functions of ξ. We have $\chi = 1$, and vary λ_2 from 0 to 0.9 in steps of 0.1, depicting all curves with $\lambda_2 > 1/3$ as dashed ones.

$$\Psi_1^{\parallel} = -\frac{(1-\lambda_2)\left[4\,(1+\chi)\,(1+3\lambda_2)+(1+\lambda_2)^2\,(1-\lambda_2)\,\xi^2\right]\tau^2}{\left[4\,(1+\chi)+\left(1-\lambda_2^2\right)\xi^2\right]^2}\chi H_0^2, \tag{14}$$

and are depicted in figure 6. The coefficients $\Psi_1^{\perp}$ and $\Psi_1^{\parallel}$ differ strongly at vanishing shear,

$$\Psi_1^{\perp}(\xi \to 0) = -\frac{1}{4}\,(1+\lambda_2)\,(1-3\lambda_2)\,\tau^2\frac{\chi}{(1+\chi)^2}H_0^2, \tag{15}$$

$$\Psi_1^{\parallel}(\xi \to 0) = -\frac{1}{4}\,(1-\lambda_2)\,(1+3\lambda_2)\,\tau^2\frac{\chi}{1+\chi}H_0^2, \tag{16}$$

and switch their behaviour especially, at $\lambda_2 = 1/3$: for smaller λ_2, $\Psi_1^{\perp}$ remains negative for all values of shear. For larger λ_2, there is a zero crossing.

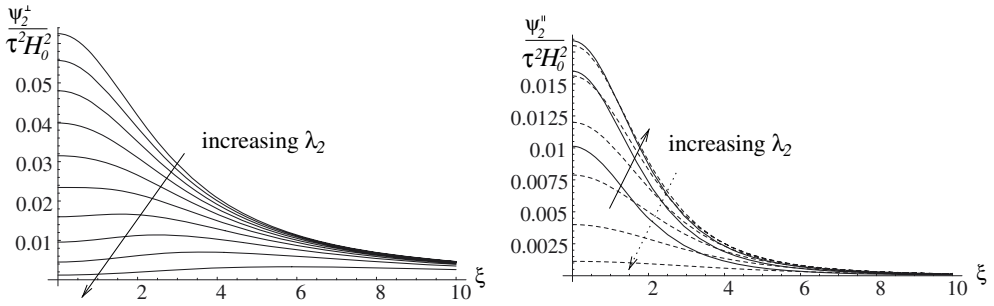

Figure 7. The second normal stress coefficients, $\Psi_2^{\perp}$ and $\Psi_2^{\parallel}$, respectively for normal and parallel external fields, in units of $\tau^2 H_0^2$, as functions of ξ. We have $\chi = 1$, and vary λ_2 from 0 to 0.9 in steps of 0.1, depicting all curves with $\lambda_2 > 1/3$ as dashed ones in the right figure.

The second normal stress coefficient, for the two cases of field orientation, are given as

$$\Psi_2^{\perp} = \frac{(1+\lambda_2)\,(1-\lambda_2)^2\left[4\,(1+\chi)+(1+\lambda_2)\,\xi^2\right]\tau^2}{\left[4\,(1+\chi)+\left(1-\lambda_2^2\right)\xi^2\right]^2}\,\frac{\chi}{1+\chi}\,H_0^2, \tag{17}$$

$$\Psi_2^{\parallel} = \frac{4\,(1+\chi)\,\lambda_2\,(1-\lambda_2)^2\,\tau^2}{\left[4\,(1+\chi)+\left(1-\lambda_2^2\right)\xi^2\right]^2}\,\chi\,H_0^2, \tag{18}$$

and are depicted in figure 7. If $\lambda_2 = 0$, we have $\Psi_2^{\perp}/\Psi_1^{\perp} = -1$ and $\Psi_2^{\parallel}/\Psi_1^{\parallel} = 0$ for all values of ξ.

3.3. Visco-elastic responses

Third, we probe the linear, visco-elastic response of ferrofluids at a given frequency ω, showing that it is, amazingly, exactly mappable onto the polymer case. Because of molecular entanglement, that needs a time τ_1 to disentangle, the polymer response to a small, oscillatory shear perturbation $\dot{\gamma}$ is viscous at low, but elastic at high, frequencies,

$$\Pi_{xy} = -[\eta_1 + K\tau_1/(1 - \mathrm{i}\omega\tau_1)]\dot{\gamma}, \tag{19}$$

with K denoting the shear elastic coefficient. Turning to ferrofluids, we consider the same geometry as above, linearize equation (1) with respect to the fluctuating components, $\delta\mathbf{M} \equiv \mathbf{M} - \mathbf{M}^{\mathrm{eq}}$ and $\mathbf{v}$, Fourier transform them, $\dot{\mathbf{M}} \to -\mathrm{i}\omega\delta\mathbf{M}$ and $\dot{\gamma} \to -\mathrm{i}\omega\gamma$, again yielding a linear, algebraic 2×2 matrix equation. The result is inverted and inserted into equation (2), leading again to equation (19), with K and τ_1, respectively, given as

$$K^{\perp} = \tfrac{1}{4}(1+\lambda_2)^2\chi\,H_0^2/(1+\chi)^2, \qquad \tau_1^{\perp} = \tau, \tag{20}$$

$$K^{\parallel} = \frac{1}{4}(1-\lambda_2)^2\chi(1+\chi)H_0^2, \qquad \tau_1^{\parallel} = \frac{\tau}{1+\chi}. \tag{21}$$

(H_0 still denotes the external field that is either perpendicular or parallel to the oscillating plate.) Taking $\omega\tau \to 0$ in equation (19) yields the enhanced viscosities at vanishing frequencies and small shear flow. So the results are the same as taking $\xi \to 0$ in equations (6) and (8).

3.4. The Trouton viscosity

Fourth, we consider the so-called *Trouton viscosity* [10]. The velocity field is now elogational, $\mathbf{v} = (-\frac{1}{2}\dot{\varepsilon}x, -\frac{1}{2}\dot{\varepsilon}y, \dot{\varepsilon}z)$, with $\boldsymbol{\Omega} \equiv 0$, and $\dot{\varepsilon}$ the parameter quantifying the flow rate (similar to

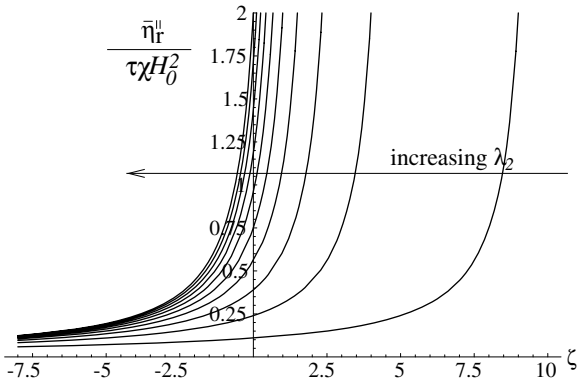

Figure 8. The magneto-viscous contribution to the Trouton, or elongational, viscosity $\bar{\eta}_r$, in units of $\tau\chi H_0^2$, as a function of the dimensionless elongation rate $\tau\dot{\varepsilon}$. λ_2 varies from 0.1 to 0.9 in steps of 0.1.

$\dot{\gamma}$ before). Since $\boldsymbol{\Omega}$ vanishes identically, there is no effect whatsoever without λ_2. Equation (1) is again inverted, leading to

$$M_x = \chi H_x/(1 + \tfrac{1}{2}\lambda_2\dot{\varepsilon}\tau), \tag{22}$$

$$M_y = \chi H_y/(1 + \tfrac{1}{2}\lambda_2\dot{\varepsilon}\tau), \tag{23}$$

$$M_z = \chi H_z/(1 - \lambda_2\dot{\varepsilon}\tau). \tag{24}$$

(Clearly, the possible divergences of M_i show that a linear constitutive relation fails here.) Inserting these expressions into equation (2), one can easily calculate the Trouton viscosity, as

$$\eta^{\mathrm{T}} = [(\Pi_{xx} - \Pi_{zz}) - (\Pi_{xx} - \Pi_{zz})|_{\dot{\varepsilon}=0}]/\dot{\varepsilon}. \tag{25}$$

The term $H_i B_j$ is now finite, and included. But the normal stress difference at vanishing flow, $\dot{\varepsilon} = 0$, is again subtracted, for the same reason as in equations (11) and (12).

We take the external field $B_0 = H_0$ to be along $\hat{\mathbf{z}}$, with the ferrofluid in the shape of a thin, long cylinder, also along $\hat{\mathbf{z}}$, implying that the ferrofluid column is either getting thinner and longer or, conversely, fatter and shorter, depending on the sign of $\dot{\varepsilon}$. Then all internal fields are also along $\hat{\mathbf{z}}$, with the internal H-field essentially constant, $H = H_0$, while the other fields are $B = H_0 + M$, $M^{\mathrm{eq}} = \chi H_0$. The magneto-viscous contribution $\bar{\eta}_r$ to the Trouton viscosity η^{T}, given as

$$\bar{\eta}_r \equiv \eta^{\mathrm{T}} - 3\eta_1 = \frac{\lambda_2\tau(1 + \lambda_2 - \lambda_2\tau\dot{\varepsilon})}{(1 - \lambda_2\tau\dot{\varepsilon})^2}\chi H_0^2, \tag{26}$$

is depicted in figure 8.

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