# Macroscopic Account of Ferrofluid Rheology

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The Ferrofluid-Dynamics as given by Müller and Liu (including the well-accepted relaxation of magnetization) is shown to be capable of accounting for a broad range of non-Newtonian behavior in ferrofluids, including shear-thinning, shear-thickening, normal stress differences, viscous-elastic response, and a varying extensional viscosity.

Ferrofluid physics has enjoyed a concise understanding for a number of its varying phenomena since its inception. 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, a relaxation equation for the magnetization $\boldsymbol{M}$ and a torque $\frac{1}{2}\varepsilon_{ijk}(\boldsymbol{H}\times\boldsymbol{M})_k$ in the stress tensor $\Pi_{ij}$, the theory has been successfully applied in numerous circumstances [1]. Recently, denser and more strongly magnetized ferrofluids were found to display strikingly non-Newtonian behavior, including shear thinning and normal stress differences [2]. Experiments, microscopic theories [3] and simulations [4] all show this to be the result of magnetic particles forming chains or elongated domains in the presence of a field, making ferrofluids resemble polymer solutions. Facing the need for a similarly concise macroscopic theory for these ferrofluids, the general feeling is its construction would need *new physical ideas* from *concepts of polymer physics* [5], that the result would be a combination of the Shliomis theory with polymer fluid-dynamics. Unfortunately, in spite of a number of useful microscopic results, such a theory has not as yet crystallized.

In 2001, Müller and Liu [6] published a paper on the thermodynamic framework of ferrofluid-dynamics. Being perceived by some as a rival to the Shliomis theory, it stirred up unnecessary controversies, provoking the unfortunate sporting question of which the better theory is. A more relaxed view takes the main result to be the following: Ferrofluid-dynamics as given by Shliomis can be divided into two parts: structure and coefficients. While the structure is determined by general principles and always valid, the coefficients (such as the relaxation time) are the result of simplifying assumptions (especially about the particle shape and their lack of interaction), and therefore much more restricted in their range of validity. This is a consequential insight, because it implies one may take the same set of equations with different coefficients to account for any system entertaining a slowly relaxing magnetization, irrespective of particle shape, be it spherical or elongated, and independent of 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 "constituent particles," or better the building blocks, are extremely elongated, and the relaxation rate of magnetization is a composite quantity, restricted not only by how fast the chains may be oriented, but also how quickly particles can be transported and assembled, to form chains of the proper length, giving rise to an appropriate amount of magnetization. This remains true as long as the chains' dynamics is not independent from that of the magnetization.

It is hardly remarkable that polymer solutions and ferrofluids would differ in some fundamental ways. Being a negative statement, the term “non-Newtonian” lacks specificity, and there may well be different versions of it requiring different descriptions. Since polymer strands are entangled without shear, but get aligned along the flow by it, while magnetic chains are aligned along the field without shear, and broken into pieces by it, their similarity must be fairly restricted.

Characterized by *transient elasticity*, Polymers’ rheology may be accounted for by strain relaxing [7]. This is not necessary for non-Newtonian ferrofluids forming short chains, because the relaxation of magnetization suffices to fully account for its dynamics. We demonstrate this by considering

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

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

The first is the relaxation equation for the magnetization, the second (with $v_i$ the velocity and neglecting the term $\rho v_i v_j$) gives the total stress, defined by local conservation, $\dot{g}_i + \nabla_j \Pi_{ij} = 0$, of the momentum $g_i$. The force density on an infinitely extended plate in the $xz$-plane, being dragged along $\hat{\boldsymbol{x}}$ on top of a ferrofluid layer, is $\triangle\Pi_{xy}$, given by the difference between the stress of air and that of the ferrofluid. Taking the total viscosity as $\eta_1 + \eta_r \equiv -\triangle\Pi_{xy}/\dot{\gamma}$, the magneto-viscous contribution, $\eta_r$, may be evaluated for the external field $B_0$ along $\hat{\boldsymbol{y}}$, perpendicular to the plate. The result is

$$\eta_r^{\perp} = \frac{(1+\lambda_2)^2[4+(1-\lambda_2)^2\xi^2]}{[4(1+\chi)+(1-\lambda_2^2)\xi^2]^2}\tau\chi B_0^2. \quad (3)$$

For vanishing shear, $\xi \to 0$, the viscosity $\eta_r^{\perp}$ grows with $\tau, \chi B_0^2$ and $\lambda_2$. More generally, $\eta_r^{\perp}$ decreases monotonically with shear if $\chi < (1+3\lambda_2)/(1-\lambda_2)$, and displays shear-thickening otherwise.

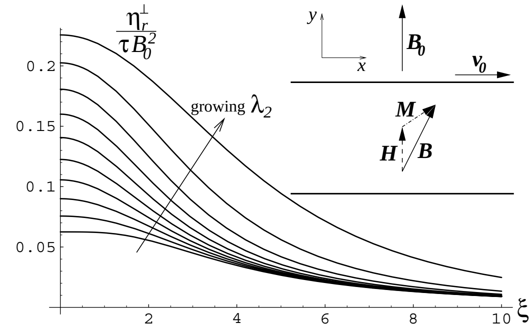


Figure 1: Magneto-viscous contribution to shear viscosity (in units of $\tau B_0^2$) as a function of $\xi \equiv \dot{\gamma}\tau$, from $\lambda_2 = 0$ to $0.9$, in $0.1$-steps, with $\chi$=1. Shear-thinning is obvious.

## References


[1] M.I. Shliomis, in S. Odenbach (Ed.), *Ferrofluids*, Lecture Notes in Physics, Vol **594**, Springer Pub, Berlin (2002).

[2] S. Odenbach and S. Turm, in S. Odenbach (Ed.), *Ferrofluids*, Lecture Notes in Physics, Vol **594**, Springer Pub, Berlin (2002); S. Odenbach, J. Phys.: Condens. Matter **16**, 1135 (2004).

[3] A.Y. Zubarev and L.Y. Iskakova, Phys. Rev **E 61**, 5415 (2000) and **E 65**, 61406 (2002); A.Y. Zubarev in S. Odenbach (Ed.), *Ferrofluids*, Lecture Notes in Physics, Vol **594**, Springer Pub, Berlin (2002).

[4] P. Ilg and M. Kröger, Phys. Rev **E 66**, 21501 (2002).

[5] M.I. Shliomis, Phys. Rev. **E 67**, 43201 (2003).

[6] H. W. Müller and M. Liu, Phys. Rev. **E 64**, 061405 (2001) and in S. Odenbach (Ed.), *Ferrofluids*, Lecture Notes in Physics, Vol **594**, Springer (2002).

[7] H.Temen, H.Pleiner, M.Liu, H.Brand, Phys. Rev. Lett. **84**, 3228 (2000).