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RHEOLOGY OF FERROFLUIDS AND MAGNETORHEOLOGICAL FLUIDS

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INTRODUCTION

Fluids with microstructure often show fascinating flow behavior. In particular, the rheological behavior of ferrofluids – stable suspensions of nano-sized ferromagnetic particles in a carrier liquid – can be manipulated by external magnetic fields. Anisotropic viscosities are observed in ferrofluids depending on the relative orientation of the applied magnetic field to the flow field. For a plane shear flow, these are the so called Miesovitz viscosities. As has been noticed [1, 2], the stress tensor of ferrofluids can be described phenomenologically by the same set of viscosity coefficients used to describe uniaxial (or generally biaxial) nematic liquid crystals in the presence of an orienting field [3-7]. The microscopic mechanisms leading to the anisotropic viscosities are, however, rather different in these systems [2]. To understand the origin of various viscosity coefficients for ferrofluids, a simple model system is studied here. The model is formulated on the level of kinetic theory, in accordance with kinetic models of nematic fluids. General expressions for the viscosity coefficients are obtained from the model. For the special case of uniaxial symmetry, the expressions are worked out in detail and compared with the result of phenomenological approaches to uniaxial nematic liquid crystals as well as findings from simulations, other theoretical approaches and experiments.

EXAMPLE: MODEL FOR DILUTE FERROFLUIDS, BROWNIAN RELAXATION REGIME

To give an example for a model developed for dilute ferrofluids made of particles for which Brownian motion is the dominating relaxation mechanism, we consider a number n of identical, rigid, non-interacting ellipsoids per unit volume. The ellipsoids of revolution may be considered to represent aggregates (or chains) of magnetic spherical particles, whose shape (prolate, oblate) is also affected by concentration and, e.g., the strengths of flow and magnetic fields. Furthermore, we assume that the magnetic moment of each aggregate is parallel to its (single) symmetry axis. Thus, the state of the system is described by the probability distribution function f of finding a particle oriented in a certain direction at certain time. The kinetic equation for such particles in the presence of a general flow field and a magnetic field is of a Fokker-Planck

type equation for f , given e.g. in [7, 8] together with an expression for the full hydrodynamic stress tensor. Analytical solutions to the kinetic model are known only for special cases like e.g. spherical particles in the limit of weak magnetic fields. Furthermore, closed expressions for the hydrodynamic stress tensor in terms of low order moments of the distribution function f cannot in general be derived rigorously from the kinetic model due to closure problems.

RELATIONSHIP TO THERMODYNAMIC APPROACHES

Thermodynamic approaches to the dynamics of ferrofluids have also been proposed in the literature that do not rely on a specific kinetic model [9]. However, these approaches assume that the macroscopic magnetization is the only relevant additional variable, needed to describe the magnetization dynamics and the stress tensor of ferrofluids. Starting from the present kinetic model and assuming the macroscopic magnetization to be the only relevant macroscopic variable, the stress tensor is found to be given by a generalization of the corresponding expression given within the thermodynamic approach of Ref. [9] due to viscous contributions. The magnetization equation on the contrary is found to be a special case of the corresponding equation given in Ref. [9]. Explicit expressions of the phenomenological parameters in the magnetization equation and the stress tensor are derived from the kinetic model.

RESULTS FOR UNIAXIAL SYMMETRY

In the sequel, the assumptions of the previous section are released to the case of uniaxial symmetry. Note, that uniaxial symmetry includes the previous approximation as a special case. In the case of uniaxial symmetry, a hierarchy of orientational order parameters is defined and the hydrodynamic stress tensor is found to be of the same form as in uniaxial nematic liquid crystals. Explicit expressions for the viscosity coefficients in terms of the order parameters and a particle shape factor are worked out in detail. For example, in flow-free equilibrium, the order parameter of lowest degree equals the Langevin function $L(x) = \coth(x) - 1/x \in [0, 1]$ where x is a dimensionless magnetic field strength. In the low Mason number regime, where the as-

sumption of uniaxiality is best maintainable, the expressions for the viscosity coefficients reduce to explicit functions of the applied magnetic field and a particle shape factor. Special emphasis is paid to the ‘rotational viscosity’ frequently studied in ferrofluids. In addition, the existence and relevance of other viscosity coefficients, together with their experimental verification is pointed out. Additional viscosity coefficients enter the description, if the universal case, i.e., biaxial symmetry, is considered. For spherical particles, this case is studied in detail in [10].

BROWNIAN DYNAMICS SIMULATIONS AND COMPARISON TO RESULTS FOR UNIAXIAL SYMMETRY

The expressions for the viscosity coefficients in case of uniaxial symmetry are compared to Brownian dynamics simulation of the full kinetic model for plane shear flow. Good agreement between numerical results and the predictions for the viscosity coefficients are obtained for strong magnetic fields, $x > 1$, and shear rates small compared to the inverse Brownian relaxation time of the ellipsoidal particles. For weak magnetic fields and/or high shear rates, deviations from the analytical predictions are seen due to deviations from uniaxial symmetry. However, even in this regime, the assumption of uniaxial symmetry still leads to predictions for the viscosities that are in reasonable agreement with the numerical results of the Brownian dynamics simulation.

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