Comment on "Measuring the transverse magnetization of rotating ferrofluids"

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Contrary to the main conclusion of Embs *et al.* [Phys. Rev. E **73**, 036302 (2006)], we demonstrate with amplitude correction factors that the predictions of the magnetization model proposed by Shliomis [Sov. Phys. JETP **34**, 1291 (1972)] are well consistent with the experimental data for weakly nonequilibrium states and that the model proposed by Shliomis [Phys. Rev. E **64**, 063501 (2001)] is valid even far from equilibrium. A model on the basis of the weak-field magnetization equation of Müller and Liu [Phys. Res. E **64**, 061405 (2001)] with a "structure" modification is also shown to reproduce a wide range of experimental data. Our statement is confirmed by a more exact insight into the hydrodynamic problem of rotating ferrofluids.

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Shliomis [1] (Sh72) postulated a ferrohydrodynamic magnetization equation

$$\frac{d\mathbf{m}}{dt} + \mathbf{m} \times \boldsymbol{\omega} = \frac{\mathbf{m}_{eq} - \mathbf{m}}{\tau}, \tag{1}$$

where d/dt is the material derivative, $\mathbf{m}$ is the magnetization vector, $\mathbf{m}_{eq}$ is the equilibrium magnetization vector, $\boldsymbol{\omega}$ is the angular velocity vector, and τ is the relaxation time. The ferrofluid was assumed to be incompressible and nonconducting. Martsenyuk *et al.* [2] (MRSh) later proposed another equation derived microscopically from the Fokker-Planck (FP) equation [3,4]

$$\frac{d\mathbf{m}}{dt} + \mathbf{m} \times \boldsymbol{\omega} = \frac{\mathbf{m}(\mathbf{h}_l \cdot (\mathbf{h} - \mathbf{h}_l))}{\tau |\mathbf{h}_l|^2} - \frac{\mathbf{h}_l \times (\mathbf{m} \times \mathbf{h})}{A(\xi_l)\tau |\mathbf{h}_l|^2}, \tag{2}$$

where

$$A(\xi_l) = \frac{2L(\xi_l)}{\xi_l - L(\xi_l) - \xi_l L^2(\xi_l)}.$$

Here, $\mathbf{h}$ is the internal magnetic field vector, $\mathbf{h}_l$ is the "local equilibrium" or "effective" field vector, L is the Langevin function, and ξ_l is the Langevin argument of the local equilibrium field strength (a dimensionless field strength). Tsebers [5,6] performed a numerical simulation of magnetic moment dynamics and indicated that the MRSh model using the effective-field method (EFM) proposed by Leontovich [7] describes perfectly the magnetization in wide ranges of ξ_0 and $\hat{\omega}\tau$, where ξ_0 is the Langevin argument of the applied stationary uniform magnetic field strength and ϖ is the fluid vorticity. Shliomis *et al.* [8] made the same conclusion by comparing the results of tangential magnetostress under the EFM-based MRSh model with those obtained by numerical integration of the FP equation. In this work, the Sh72 model was justified for weakly nonequilibrium magnetization states (i.e., $\hat{\omega}\tau \to 0$). Recently, many authors have proposed modifications to obtain a more proper form than the Sh72 model or a simpler form than the MRSh model. Felderhof [9,10] (Feld) showed that irreversible thermodynamics in combination with the Maxwell equations leads to the equation

$$\frac{d\mathbf{m}}{dt} + \mathbf{m} \times \boldsymbol{\omega} = \gamma_H(\mathbf{h} - \mathbf{h}_l), \tag{3}$$

where γ_H is the Feld relaxation rate. Shliomis [11] (Sh01) later indicated that the Feld model leads to anomalous viscosity results and further revised Eq. (3) to have the form

$$\frac{d\mathbf{h}_l}{dt} + \mathbf{h}_l \times \boldsymbol{\omega} = \frac{\mathbf{h} - \mathbf{h}_l}{\tau}. \tag{4}$$

Comparing with the viscosity predictions of the Sh72 and MRSh models, Shliomis [12] showed that the new magnetization equation turns out to be valid even far from equilibrium and concluded that it should be recommended for further application. At almost the same time, Müller and Liu [13] (ML) proposed a new set of ferrohydrodynamic equations divided into two parts: structure and coefficients. Their magnetization equation for the weak-field case is

$$\frac{d\mathbf{m}}{dt} + \mathbf{m} \times \hat{\boldsymbol{\omega}} = \zeta(\mathbf{h} - \mathbf{h}_l), \tag{5}$$

where $\hat{\boldsymbol{\omega}}$ is the flow vorticity vector, related to the flow velocity vector $\mathbf{v}$ by $\hat{\boldsymbol{\omega}} = \nabla \times \mathbf{v}/2$, and ζ is the ML relaxation rate. Comparing with the experimental results of the transverse magnetization of a ferrofluid in a rotating cylinder (rotating ferrofluid) for a wide range of $\hat{\boldsymbol{\omega}}\tau$ with a fixed small value of ξ_0, Embs *et al.* [14] showed that the weak-field ML model with proper coefficient setting must be preferred. To our surprise, they also showed that the Sh01 model leads to strong disagreement with their measurements and that the Sh72 model is not valid even for weakly nonequilibrium states. Below, we argue the reduced forms of magnetization equations shown in their work and draw different conclusions from our findings.

Let r, θ, and z denote the usual cylindrical polar coordinates. Consider the same experimental setup. A cylinder of radius R rotates with the angular velocity vector $\boldsymbol{\Omega} = (0,0,\Omega)$ in the presence of an applied stationary uniform

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magnetic field vector $\mathbf{h}_0$ oriented in the perpendicular direction of $\mathbf{\Omega}$. The linear and angular momentum balance equations are, respectively,

$$\rho\frac{d\mathbf{v}}{dt} = -\nabla p^* + (\mu+\kappa)\nabla^2\mathbf{v} + 2\kappa(\nabla\times\boldsymbol{\omega}), \tag{6}$$

$$\rho j\frac{d\boldsymbol{\omega}}{dt} = -4\kappa(\boldsymbol{\omega}-\hat{\boldsymbol{\omega}}) + \mathbf{m}\times\mathbf{h}, \tag{7}$$

where p^* is a magnetohydrodynamic pressure [15,16]; ρ is the density; κ is the vortex viscosity, related to the shear viscosity μ and the particle volume fraction φ by $\kappa=3\mu\varphi/2$; and j is the moment of inertia per unit mass. The internal magnetic field vector $\mathbf{h}$ and the magnetization $\mathbf{m}$ within a ferrofluid could be related to each other via $\mathbf{h}=\mathbf{h}_0-n\mathbf{m}$, where n is the demagnetizing factor ($n=1/2$ in the experimental setup). The hydrodynamic equations (6) and (7) admit a steady solution in the form

$$\mathbf{v}=(0,u_\theta(r),0),\quad p^*=p^*(r),$$

$$\boldsymbol{\omega}=(0,0,\omega_z(r)),$$

$$\mathbf{h}=(h_r,h_\theta,0),\quad \mathbf{m}=(m_r,m_\theta,0), \tag{8}$$

and thus reduce to

$$Dp^*=\rho\frac{u_\theta^2}{r},$$

$$(\mu+\kappa)DD_*u_\theta - 2\kappa D\omega_z = 0,$$

$$4\kappa\omega_z - 2\kappa D_*u_\theta = m_rh_\theta - m_\theta h_r, \tag{9}$$

where $D=d/dr$ and $D_*=d/dr+1/r$. Embs *et al.* considered the rotating ferrofluid as a rigid body with velocity

$$u_\theta(r)=\Omega r. \tag{10}$$

Substituting Eq. (10) into the linear momentum balance shown in Eq. (9) gives

$$\omega_z(r)=A. \tag{11}$$

By using the no-slip boundary condition $\omega_z(R)=\Omega$, the unknown constant can be obtained as $A=\Omega$. Substituting Eqs. (10) and (11) into the angular momentum balance shown in (9) gives

$$m_rh_\theta - m_\theta h_r = 0. \tag{12}$$

It means that the magnetization vector $\mathbf{m}$ is always parallel to the internal magnetic field vector $\mathbf{h}$, i.e., the magnetic torque is neglected ($\mathbf{m}\times\mathbf{h}=0$). This is contradictory to the actual magnetic behavior in such a ferrohydrodynamic system.

Now we get a more exact insight into the hydrodynamic problem of rotating ferrofluids. Integrating Eq. (9) and considering the limiting case $D|\mathbf{m}\times\mathbf{h}|\to 0$ (small magnetic-torque gradient), we obtain

$$u_\theta(r)=Br+C/r,$$

$$\omega_z(r)=B+\frac{m_rh_\theta-m_\theta h_r}{4\kappa},$$

$$p^*(r)=\rho\int\frac{u_\theta^2}{r}dr. \tag{13}$$

Assuming that the fluid adheres to the cylinder with the conditions

$$u_\theta(R)=\Omega R,\quad \omega_z(R)=\Omega \tag{14}$$

gives

$$B=\Omega-\frac{m_rh_\theta-m_\theta h_r}{4\kappa},\quad C=\frac{m_rh_\theta-m_\theta h_r}{4\kappa}R^2. \tag{15}$$

Thus, the angular velocity vector is

$$\boldsymbol{\omega}=\mathbf{\Omega} \tag{16}$$

and the fluid vorticity vector is

$$\hat{\boldsymbol{\omega}}=\mathbf{\Omega}-\frac{\mathbf{m}\times\mathbf{h}}{4\kappa}. \tag{17}$$

It should be noted that the conditions (14) have been confirmed by Stokes [17] for the dynamics of fluids with microstructure.

The model equations for the magnetization in situations where $\mathbf{m}$ and $\mathbf{h}$ are spatially and temporally constant but not parallel to each other include either the magnetization vector $\mathbf{m}$ toward the equilibrium magnetization vector $\mathbf{m}_{eq}(\mathbf{h})=\chi(h)\mathbf{h}$ or the local equilibrium field vector $\mathbf{h}_l(\mathbf{m})=F(m)\mathbf{m}$ toward the internal field $\mathbf{h}$. By using the relations (16) and (17) and the setting $\zeta=1/F\tau$ [18] and $\gamma_H=\chi_0/\tau$ [19], where the initial susceptibility $\chi_0=\chi(h\to 0)$, the five well-known model equations (1)–(5) can be written in the common form

$$\mathbf{m}\times(\mathbf{\Omega}+\alpha_3\mathbf{m}\times\mathbf{h}_0)=\alpha_1(\mathbf{h}_0-\alpha_2\mathbf{m}), \tag{18}$$

with the following coefficients:

Sh72: $\alpha_1=\frac{\chi}{\tau},\quad \alpha_2=\frac{1}{\chi}+n,\quad \alpha_3=0$

(the Debye model of Embs *et al.*);

MRSh: $\alpha_1=\frac{1}{A(\xi_l)F\tau},\quad \alpha_2=F+n,\quad \alpha_3=0;$

Feld: $\alpha_1=\frac{\chi_0}{\tau},\quad \alpha_2=F+n,\quad \alpha_3=0$

[the ML(F) model of Embs *et al.*];

Sh01: $\alpha_1=\frac{1}{F\tau},\quad \alpha_2=F+n,\quad \alpha_3=0$

[the ML(S) model of Embs *et al.*];

$$\text{ML:}\quad \alpha_1 = \frac{1}{F\tau},\quad \alpha_2 = F + n,\quad \alpha_3 = -\frac{1}{4\kappa}.$$

Embs *et al.* has shown with amplitude correction factors of the order of 0.1 that the shapes of the curves for the transverse field strength h_y^{sensor} versus the angular velocity Ω are somewhat better reproduced by the Debye model (the present Sh72 model) and the ML(S) model (the present Sh01 model) and that only the ML(S) model reproduces well the curve for large values of Ω. Leschhorn *et al.* [20] showed that these corrections result from the polydispersity of ferrofluids. We may, therefore, conclude with amplitude correction factors that the Sh72 model is able to reproduce the experimental data for weakly nonequilibrium states and that the Sh01 model is valid even for strong nonequilibrium states. In addition, the coefficient α_3 has been shown by Embs *et al.* to have a strong effect on the discrepancy in the theoretical and experimental comparison. A "structure" modification for the weak-field magnetization equation of Müller and Liu [13] should, therefore, be done. We write Eq. (5) in a proper "structure":

$$\frac{d\mathbf{m}}{dt} + \mathbf{m} \times \boldsymbol{\omega} = \zeta(\mathbf{h} - \mathbf{h}_l). \tag{19}$$

For rotating ferrofluids, it can be written in the common form (18) with the same coefficients of the Sh01 model, i.e., its predictions are also well consistent with a wide range of experimental data. In Fig. 1, we plot possible approximations of the measurements of Embs *et al.* It is clear from the plot that the ML model is impertinent for the experiment. The MRSh model seems to be the best approximation, and the WC (the present) model provides another way of estimating the transverse magnetization.

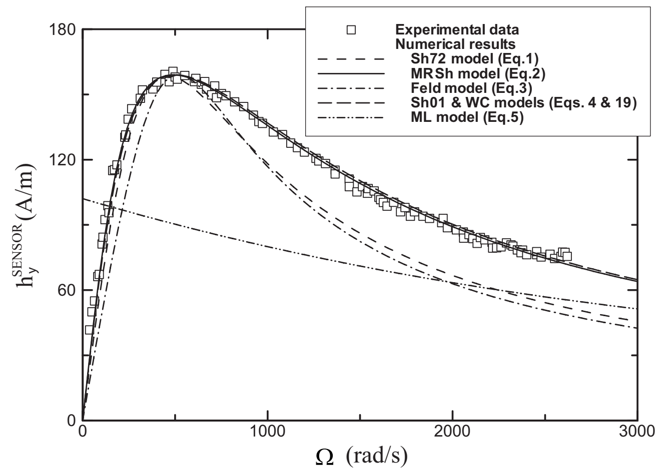

FIG. 1. Comparison of the experimental data of the transverse local equilibrium field strength measured by the sensor at the applied magnetic field $h_0 = 30$ kA/m with the numerical results according to different theoretical models. The fit parameters used for the Sh72, Feld, and Sh01 models are shown in Table I of Embs *et al.* [14], and those used for the MRSh, Weng-Chen (WC), and ML models are the same as the Sh01 model.

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