Reply to "Comment on 'Measuring the transverse magnetization of rotating ferrofluids'"

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In the preceding comment the authors present a theoretical analysis of experimental transverse magnetization data of a ferrofluid in a cylinder that is rotating in a magnetic field [Embs *et al.*, Phys. Rev. E **73**, 036302 (2006)]. This analysis of Weng and Chen is based on theoretically unfounded assumptions that imply unphysical properties of the velocity and vorticity fields of the rotating fluid, leading to wrong predictions for the magnetization. We show where the errors occurred. We uphold our main conclusion that the investigated single-relaxation-time models do not reproduce well the experimental results.

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We first discuss here the three major points where the analysis of Weng and Chen (WC) is wrong in our opinion: (i) misinterpretation of the field $\boldsymbol{\omega}$ [1], (ii) unwarranted boundary condition on $\boldsymbol{\omega}$, and (iii) diverging velocity field $\mathbf{v}$. There we shortly address also further inconsistencies in [2] and then we discuss under point (iv) the role of polydispersity before summarizing our reply.

I. MISINTERPRETATION OF THE FIELD $\boldsymbol{\omega}$

Below Eq. (1-WC) in [2] it is stated that "$\boldsymbol{\omega}$ is the angular velocity vector." However, $\boldsymbol{\omega}$ was introduced in [3] to be the averaged *rotation velocity of the magnetic particles* in the suspension. We do not comment on whether it is useful to introduce such an auxiliary field in a macroscopic field theory that ultimately should connect the macroscopic fields of velocity, magnetization, and the magnetic field. Here we show that WC draw the wrong conclusions from this field $\boldsymbol{\omega}$ that appears in the Shliomis approach and that is commonly eliminated immediately for good reasons with the relaxation equation (7-WC)

$$\rho j d_t \boldsymbol{\omega} = -4\kappa(\boldsymbol{\omega} - \hat{\boldsymbol{\omega}}) + \mathbf{m} \times \mathbf{h}. \tag{1}$$

Here ρ is the density and κ is the vortex viscosity that is related to the shear viscosity μ and the particle volume fraction φ by $\kappa = 3\mu\varphi/2$. Furthermore, j is the moment of inertia per unit mass and

$$\hat{\boldsymbol{\omega}} = \frac{1}{2} \nabla \times \mathbf{v} \tag{2}$$

is the local rotation velocity of the fluid defined by the physically much more relevant and better defined vorticity of the velocity field $\mathbf{v}$ of the suspension.

So $\boldsymbol{\omega}$ is fully determined via Eq. (7-WC) in terms of $\mathbf{h}$, $\mathbf{m}$, and $\mathbf{v}$ (and possibly an *initial* condition for $\boldsymbol{\omega}$). For example, under the commonly adopted quasistationary conditions one has

$$\boldsymbol{\omega} = \hat{\boldsymbol{\omega}} + \frac{1}{4\kappa} \mathbf{m} \times \mathbf{h}. \tag{3}$$

For the sake of clarity and simplicity we consider in the following such conditions to apply.

II. UNWARRANTED BOUNDARY CONDITION ON $\boldsymbol{\omega}$

The ten independent field variables for $\mathbf{v}$, $\mathbf{h}$, $\mathbf{m}$, and $p*$ (the magnetohydrodynamic pressure) are described by ten independent equations: The continuity equation $\nabla \cdot \mathbf{v} = 0$, the three components of the momentum balance (6-WC) [4], three independent relations from the Maxwell equations

$$\nabla \times \mathbf{h} = 0, \quad \nabla \cdot (\mathbf{h} + \mathbf{m}) = 0, \tag{4}$$

and, finally, the three components of some of the magnetization equations that are commonly used [5,6]. The latter typically contain $\mathbf{m}$, $\mathbf{h}$, and only first spatial derivatives of $\mathbf{v}$ like Eq. (3) for $\boldsymbol{\omega}$. Thus, the highest spatial derivatives are first order for $\mathbf{m}$ and $\mathbf{h}$ in Eq. (4) and second order for $\mathbf{v}$ in the momentum balance (6-WC). Consequently, one needs one boundary condition for both $\mathbf{m}$ and $\mathbf{h}$, but *two* for $\mathbf{v}$.

The fact that $\mathbf{m}$ vanishes outside the cylinder and that $\mathbf{h}(r \to \infty)$ is equal to the externally applied field together with the continuity of the normal component of $\mathbf{m}+\mathbf{h}$ and of the tangential components of $\mathbf{h}$ at the cylinder surface provides the necessary boundary conditions for the Maxwell equations.

The macroscopic velocity field $\mathbf{v}$ of the fluid should fulfill no-slip conditions at the wall of the cylinder—i.e., $\mathbf{v}(r=R) = \Omega R \mathbf{e}_\theta$, where Ω is the rotation frequency of the cylinder. And in addition, $\mathbf{v}$ must not diverge inside the cylinder:

$$|\mathbf{v}(r < R)| < \infty. \tag{5}$$

This is the second of the two conditions that are required for $\mathbf{v}$ with second-order derivatives appearing in the field equations.

WC, on the other hand, do not specify two boundary conditions for $\mathbf{v}$—and that is the major problem of the approach of WC. They instead replace the condition (5) by an unwarranted boundary condition on $\boldsymbol{\omega}$: namely, $\boldsymbol{\omega}(r=R) = \Omega \mathbf{e}_z$ (14-WC).

III. DIVERGING VELOCITY FIELD $\mathbf{v}$

With the wrong boundary conditions (14-WC) WC deduce that the velocity field should be

$$\mathbf{v} = \left(Br + \frac{C}{r} \right) \mathbf{e}_{\theta}, \tag{6}$$

with constants

$$B = \Omega - \frac{|\mathbf{m} \times \mathbf{h}|}{4\kappa}, \quad C = \frac{|\mathbf{m} \times \mathbf{h}|}{4\kappa} R^2 \tag{7}$$

instead of the rigid-body rotation field $\mathbf{v}=\boldsymbol{\Omega}\times\mathbf{r}=\Omega r\mathbf{e}_{\theta}$. Note, however, that the field (6) of WC diverges at the center, $r=0$, of the cylinder when $\mathbf{m}\times\mathbf{h}\neq 0$. And that was precisely measured by us [5].

Furthermore, we have no indication that the flow field in our cylinder of inner radius $R=3.2$ mm deviates from the rigid-body rotation $\mathbf{v}=\Omega r\mathbf{e}_{\varphi}$, which is understandable in view of the smallness of R and the resulting strong effect of the no-slip condition at R on the flow in the cylindrical volume. So it is the average rotation velocity of the magnetic particles $\boldsymbol{\omega}$ that adjusts itself to the vorticity $\boldsymbol{\Omega}$ of this flow; $\boldsymbol{\omega}=\boldsymbol{\Omega}+(\mathbf{m}\times\mathbf{h})/(4\kappa)$ according to Eq. (3), not the other way around. The assumption of WC that the nanoscopic magnetic particles rotate on average with the same frequency as the cylinder, $\boldsymbol{\omega}=\boldsymbol{\Omega}$ (16-WC), is unphysical in view of rotational dissipation.

Note that the vorticity $\hat{\boldsymbol{\omega}}$ of the unphysical velocity field (6) of WC is

$$(17-\mathrm{WC}): \quad \hat{\boldsymbol{\omega}} = \boldsymbol{\Omega} - \frac{1}{4\kappa}\mathbf{m} \times \mathbf{h}, \tag{8}$$

instead of $\hat{\boldsymbol{\omega}}=\boldsymbol{\Omega}$ as in the flow of a rigid body rotation. The use of $\hat{\boldsymbol{\omega}}$ (17-WC) instead of $\hat{\boldsymbol{\omega}}=\boldsymbol{\Omega}$ in the magnetization equations of Refs. [5,6] cancels the terms $\sim\mathbf{m}\times(\mathbf{m}\times\mathbf{h})$ that appear, e.g., in the models denoted by Sh72 and FK. Thereby Sh72 [5,6] would be transformed into the Debye model [5,6], the model Sh01 [5,6] would go over into the ML(S) model [5,6], and the Debye model [5,6] would acquire a term $\sim\mathbf{m}\times(\mathbf{m}\times\mathbf{h})$. The relation between the coefficient α_3 in Eq. (A7) of Ref. [5] and the coefficient α_3^{WC} in Eq. (18-WC) of Ref. [2] is $\alpha_3^{WC}=\alpha_3-\frac{1}{4\kappa}$. Thus, using an unphysical velocity field with a wrong vorticity WC arrive at magnetization equations (18-WC) with coefficients that are wrong [7].

IV. THE ROLE OF POLYDISPERSITY AND THE MAIN CONCLUSION OF [5]

We should like to stress that the first sentence "Contrary to the main conclusion…" in the abstract of WC is misleading: The main conclusion of [5] is that none of the single-relaxation-time models that were investigated in [5] is really able to reproduce well the experimental results. Amplitude correction factors of the order of about 1/10 would be needed—as also in the approach of WC. This discrepancy was explained (compare also [8]) by the polydispersity of the ferrofluid which is not properly accounted for by single-relaxation-time models.

V. SUMMARY

(a) The diverging velocity field (6) of WC is unphysical and has a wrong vorticity $\hat{\boldsymbol{\omega}}$, Eq. (8). (b) In the flow field $\mathbf{v}=\boldsymbol{\Omega}\times\mathbf{r}$ of a rigid body rotation one has $\hat{\boldsymbol{\omega}}=\boldsymbol{\Omega}$. (c) The assumption that the nanoscopic magnetic particles rotate on average with the same frequency as the cylinder, $\boldsymbol{\omega}=\boldsymbol{\Omega}$ (16-WC), is unphysical in view of dissipation and leads to wrong predictions for the magnetization of rotating ferrofluids.

[1] For the sake of an easy comparison with [2] we stick here to their notation despite the fact that we consider it to be ill chosen and prone to misinterpretations. So the vorticity is denoted as $\hat{\boldsymbol{\omega}}$, while the auxiliary field is denoted by $\boldsymbol{\omega}$. Furthermore, the magnetic field is $\mathbf{h}$ and the magnetization is $\mathbf{m}$ in Ref. [2].

[2] H. C. Weng and C. K. Chen, preceding paper, Phys. Rev. E **78**, 068301 (2008).

[3] M. I. Shliomis, Sov. Phys. JETP **34**, 1291 (1972).

[4] In (6-WC) a Kelvin-like force term is missing which, however, has no consequences for spatially constant $\mathbf{m}$ and $\mathbf{h}$.

[5] J. P. Embs *et al.*, Phys. Rev. E **73**, 036302 (2006).

[6] A. Leschhorn and M. Lücke, Z. Phys. Chem. **220**, 219 (2006).

[7] There is an additional algebraic mistake in WC's representation below Eq. (18-WC) of the magnetization equation MRS (2-WC) of Martsenyuk *et al.*, [9]. Writing it in the common form (18-WC) the correct coefficients are $\alpha_1=1/(F\tau)$, $\alpha_2=F+n$, and $\alpha_3=(6\varphi\mu)^{-1}+(1-A)/(AF\tau M^2)$ [respectively $\alpha_3=(1-A)/(AF\tau M^2)$, if one uses the relation $\omega=\Omega$ of WC instead of $\hat{\omega}=\Omega$].

[8] A. Leschhorn, J. P. Embs, and M. Lücke, J. Phys.: Condens. Matter **18**, 2633 (2006).

[9] M. A. Martsenyuk, Y. L. Raikher, and M. I. Shliomis, Sov. Phys. JETP **38**, 413 (1974).