Fluid Dynamics of Colloidal Magnetic and Electric Liquid

Mario Liu*
Institut für Theoretische Physik, Universität Hannover, 30167 Hannover, Germany
(Received 26 July 1994)

Consistent inclusion of the Maxwell equations, especially the recently derived dissipative fields, leads to two novel ponderomotive forces: one electric, the other magnetic. This completes the rigorous fluid dynamical description of ponderable liquids and renders mesoscopic variables such as domain spin or particle rotation superfluous.

PACS numbers: 75.50.Mm, 47.65.+a

The hydrodynamic theory of isotropic liquid consists of three balance equations for the conserved densities: energy, momentum, and particle number; the dynamics of the electromagnetic field is governed by the macroscopic Maxwell equations. Put together, there is no reason at all why these equations should not describe the dynamics of any fluid that is either exposed to an electromagnetic field or, equivalently, contains charges. The fluid may be insulating or conducting, polarizable, magnetizable, or both, as all these phenomena abide conservation laws and are contained in the macroscopic Maxwell equations. What is more, including the concentration as a variable, these statements become valid for a mixture, and at the price of a smaller hydrodynamic regime, also for suspensions: ferrofluid (including both the quasistationary and the micropolar variety [1]) and colloidal electric fluid [2].

This claim is bound to raise a few eyebrows, especially in the ferrofluid community. The hydrodynamics of ferrofluid [1,3,4] relies squarely on two additional variables, the magnetization $M$ and the spin $S$ of the suspended particles, to successfully interpret and understand the peculiar behavior of ferrofluid. In fact, the torque (or couple) produced by $M$ and $S$ are considered by many in the field as the most characteristic feature of ferrohydrodynamics.

Before going into the detailed arguments showing why the variables $M$ and $S$ are nevertheless dispensable, it is worth pointing out that if the opening statements are true, we would have at our hands a highly useful and desirable theory, which is simple, rigorous, and unifying. The theory is simple since it contains the smallest possible number of variables and, accordingly, the smallest number of equations of motion. The theory is rigorous, since it starts from generally valid input: conservation laws, Maxwell equations, and thermodynamics. (In contrast, $M$ and $S$ are not conserved.) Finally, the theory is unifying as it is valid for all the systems outlined above.

The crucial new step in the present approach is the systematic and consistent inclusion of the Maxwell equations, especially the dissipative fields $H^D$ and $E^D$ that were only recently derived and considered [5]. These dissipative fields appear when the magnetization and the polarization are not quite in equilibrium, and drive them toward it (i.e., they assume the same function as the diffusive heat current or the viscous stress). They also enter the Navier-Stokes equation and transmit momentum and angular momentum from the electromagnetic field to the material; in other words, they serve as dissipative ponderomotive forces there. For instance, the peculiar behavior of ferrofluid exposed to a rotating field [6] is easily understandable with the help of the magnetic part of this force. In what follows, the dissipative fields and forces will be introduced first. Then, concentrating on ferrofluids, the ramifications of the dissipative magnetic force will be considered, ending with a note on how to draw the analogy to the electric case. Finally, the complete set of hydrodynamic equations of ponderable systems is derived.

The structure of the macroscopic Maxwell equations,

$$\dot{D} = c \nabla \times H^M - j^c, \quad \dot{B} = -c \nabla \times E^M, \quad \nabla \cdot D = \rho^c, \quad \nabla \cdot B = 0, \quad (1)$$

imposes the interpretation that the field variables are $D$ and $B$. The first two are their equations of motion; the next two are constraints, to be satisfied at all times. Conversely, the two fields $H^M$ and $E^M$ appear only in the flux part of the equations of motion. They are the “currents,” which generally contain both reactive and dissipative parts,

$$H^M = H + H^D, \quad E^M = E + E^D. \quad (3)$$

In accordance with the concept of local equilibrium, the reactive fields $H$ and $E$ contain only equilibrium information and are given as $H = \partial \varepsilon / \partial B$, $E = \partial \varepsilon / \partial D$, respectively, $\varepsilon$ being the thermodynamic energy density [7]. Generally, $H$ and $E$ are nonlinear functions of all the thermodynamic variables, in particular of $D$ and $B$. The nonequilibrium information is confined to the dissipative fields $H^D$ and $E^D$, which are functions of thermodynamic forces, e.g., the temperature gradient $\nabla T$ or the shear flow $v_{ij} = (\nabla_i v_j + \nabla_j v_i)/2$. For the simple isotropic, diagonal case, we have [5]

$$H^D = -\alpha c \nabla \times E^0, \quad E^D = \beta c \nabla \times H^0. \quad (4)$$

As the rate of entropy production must not change with the frame, the dissipative fields depend only on the thermodynamic fields in the local rest frame: $E^0 = E + v \times B/c$ and $H^0 = H - v \times D/c$. The terms $H^D$ and $E^D$ are of different parity under time reversal than $H$ and $E$.}

Figure 0031-9007/95/74(22)/4535(4)$06.00 © 1995 The American Physical Society 4535
$E$, respectively; they therefore account for irreversibility and damping in Eqs. (1) [8]. A direct and noteworthy consequence of Eq. (3)—cf. Eq. (17) below—are the dissipative ponderomotive forces,

$$f_{\text{mag}}^D = B \times (\nabla \times H^D), \quad f_{\text{el}}^D = D \times (\nabla \times E^D).$$  

(5)

The size of these dissipative fields and forces depends on the magnitude of the transport coefficients $\alpha$ and $\beta$. These have been estimated, in a simple model [5], as $\alpha = \tau_M M/\beta$ and $\beta = \tau_P P/\beta D$, where $\tau$ denotes the relaxation time, of the magnetization if the index is $M$ and of the polarization if it is $P$. Obviously, $\alpha$ is especially large in ferrofluids, and $\beta$ in colloidal electric fluids. So these fluids are not only interesting in their own right; they also display prominently the rather general feature of the Maxwell equations to entertain dissipative magnetic and electric fields. Vice versa, the Maxwell equations play a much more direct role in their fluid dynamics than has been hitherto taken account of.

Neglecting $\beta$ (of the dissipative electric field) in a ferrofluid, the dissipative magnetic field, Eq. (4), can be written as

$$H^D = \alpha [\dot{B} - (B \cdot \nabla)v + B(\nabla \cdot v) + (v \cdot \nabla)B].$$  

(6)

(The last two terms depend on compressional flow and field inhomogeneity, respectively; they are usually small and will be neglected here.) Consider first solid body rotation $v = \Omega \times r$ in the presence of a static field $B$. With $(B \cdot \nabla)v = \Omega \times B$, the Navier-Stokes equation reduces to $\theta \dot{\Omega} = \int r \times f_{\text{mag}}^D = \int \alpha [\Omega(\cdot B)B - B^2 \Omega]$, with $\theta$ denoting the moment of inertia. Although the relaxation rate $\theta^{-1} \int \alpha B^2$ is for a mm-scale sphere ($\alpha = 10^{-5}$ s) 2 orders of magnitude smaller than the eddy-current rate $\int \sigma B^2/\rho c^2$ of a good conductor (with a conductance $\sigma = 10^9$ S/m an industrial-size effect), it is certainly measurable in ferrofluids. Next, consider shear flow in the presence of a static field. With $f_{\text{mag},i}^D = \alpha B_i B_k (\nabla_i v_j - \nabla_j v_i)$, the shear diffusion equation becomes $\rho \ddot{v} = \eta v \nabla v$, where $\eta = \eta + \alpha B^2 \cos^2 \varphi$, and $\varphi$ denotes the angle between $B$ and the direction of the gradient. Obviously, the viscosity increases (as observed) when exposed to a field. (Note, however, that the viscosity is a tensor in the presence of fields, hence the field dependence may in general be more complicated.)

Things are just as simple if the external field is time dependent, yet the story of understanding the spin-up of ferrofluid due to a rotating field has been long and winding indeed. First, theorists did not accept that there was a spin-up at all; then, for a long stretch of time, theory and experiment could not agree on the sense of fluid rotation, as ferrofluid was frequently observed to rotate against the external field (and admittedly also against the intuition). Only recently did Rosensweig, Poppelwell, and Johnston, in a series of thought- and artfully designed experiments, compellingly show that the key to the understanding lies in the combination of a surface couple and the fluid’s capillary form [6]. Not surprisingly, part of the dissipative ponderomotive force containing $B$ of Eq. (6), $f_{\text{mag}}^D = B \times (\nabla \times \alpha \dot{B})$, is such a surface coupling: It vanishes inside the ferrofluid, since the $B$ field is essentially homogeneous, but not at the fluid surface, where $\alpha$ is discontinuous. More appropriately, one should, as in Eq. (19) below, incorporate $f_{\text{mag}}^D$ into the boundary condition:

$$\eta(\nabla_n v_i - v_i/R) = \alpha B_n (B_i - B \nabla_i v_i),$$  

(7)

where the subscripts $n$ and $t$ refer to the field’s normal and tangential components, respectively, and $R$ is the local curvature radius. This boundary condition expresses continuity of the total stress tensor (or momentum conservation) at a free surface even though the magnetization is (due to $B$) not quite in equilibrium. At a stick surface, the usual boundary condition $v_i = 0$ holds. They have to be satisfied by the solution of the shear flow equation $\rho \ddot{v} = \eta \nabla^2 v$. This is all we need for a quantitative understanding of the ferrofluid’s spin-up.

In a planar geometry, with a free surface at $x = -L$, a stick surface at $x = 0$, a static $B_n || \hat{x}$, and an oscillatory $B_t = \Delta B \exp(-i \omega t) \parallel \hat{y}$, the flow field for large penetration depths ($\delta_y = \sqrt{\eta \rho / \omega} \gg L$, $\eta = \eta + \alpha B^2$) is $v_y(x) = -i \omega a_x / \eta \Delta B$. For higher frequencies, substitute $x$ with $\sin q/x/\cos q L$ [where $q = \sqrt{\eta / \delta_y}$, and $2i \sin q = \exp(i q x) - \exp(-i q x)$ even though $q$ is complex, and similarly for $\cos q L$]. The flow field is unchanged for two free surfaces, at $x = \pm L$, except for an undetermined constant velocity.

Now take a counterclockwise rotating field, $B_n = B \cos \omega t$, $B_t = B \sin \omega t$. The boundary condition becomes $\alpha \omega B^2 (1 + \cos 2 \omega t)/2 = (\eta + \alpha B^2 \cos^2 \omega t) \nabla_n v_i$, which implies nonlinear drive. We therefore assume $\eta \gg \alpha B^2$ and expand in $\alpha B^2 / \eta$. Then the flow field remains linear and is given by the superposition,

$$v_y(x) = \frac{\alpha \omega B^2}{2 \eta} \left[ x + \frac{e^{-2 i \omega t} \sin q}{q \cos q L} \right],$$  

(8)

with $q = (1 + i)/\delta_y$, $\delta_y = \sqrt{\eta / \rho \omega}$. For $\delta_y \ll L$, the second term is small and simplifies as $\sin q x/(q \cos q L) \approx \exp[i q (x + L)].$ This term is nonvanishing only between $-L$ and $\delta_y - L$. The velocity at the free surface is then $v_y(-L) = -(\alpha \omega B^2/2 \eta)(L + \delta_y(i - 1)^{-1} \exp(-2i \omega t))$.

Clearly, magnetic excitation of a planar shear flow merits attention for its own sake, but it also provides understanding of the ferrofluid’s spin-up. Consider a sheet of circular flow between two concentric cylinders, of distance $D$ and with the free surface at the inner cylinder. The rotating flow field now has a different phase at each point of the fluid, given by the polar angle $\varphi$ of the location, $B_n = B \cos(\omega t - \varphi)$, $B_t = B \sin(\omega t - \varphi)$. So the above planar flow field needs to be averaged over $\varphi$ (leaving only the time-independent part) and bent into a circle. The result is a clockwise rotating free surface, with the velocity $v = -(\alpha \omega B^2/2 \eta D)$. By averaging $\cos^2(\omega t - \varphi)$ over $\varphi$, the effective viscosity $\eta_e = \eta + \alpha B^2/2$ is time independent. Hence the drive is linear even without an
The velocity of the free surface and hence of the bulk is estimated to be a few cm, as measured. [The observed instability in Fig. 3 of [6] may be due to the oscillatory part $\cos 2\omega t$.]

More generally, the shear diffusion equation may be solved in cylindrical coordinates, assuming symmetry with respect to $\varphi$. This yields the velocity field $v = \Omega r + \delta/r$ along the $\varphi$ direction, with two constants, $\Omega$ and $\delta$, to be determined by boundary conditions, at the stick surface $R_s$ and the free surface $R_f$. The result is $v(r) = \Omega(r^2 - R_s^2)/r$,

$$\Omega = \alpha B^2 \omega [4\eta R_s^2/R_f^2 + \alpha B^2(1 + R_s^2/R_f^2)]. \quad (9)$$

Clearly, $v(R_f) > 0$ if $R_f > R_s$, or corotation of the bulk fluid, while $v(R_f) < 0$ if $R_f < R_s$ counterrotation. If the two radii are close, the flow field becomes the bent planar solution of the last paragraph, with $D = R_s - R_f$. (Experimentally, the present situation may be realized by a freely rotating cylinder at $R_f$.) If one of the two radii $R_f$ and $R_s$ vanishes, then $\delta = 0$ to prevent divergence at $r = 0$. We have $\Omega = 0$ if the remaining one is $R_s$, and a corotating $\Omega = 0$ if $R_f$ remains. (The value of $\Omega$ depends on the residual damping.)

By substituting $D \rightarrow B, E \rightarrow H, \beta \rightarrow \alpha$, analogous results are produced for colloidal electric fluids.

The standard theory of ferrofluid dynamics [1,3,6] contains the mesoscopic quantities magnetization $M$ and spin $S$ as additional variables. The usual argument to include them is as follows: Given a time-dependent external field, the magnetization will generally lag behind the perturbation and hereby produce a macroscopically detectable coupling. However, this is not at all special. Many internal degrees of freedom are not considered independent; rather, they are taken to assume the appropriate equilibrium values. This is, of course, the concept of local equilibrium, the central premise of the hydrodynamic theory. The delay, on the other hand, is adequately taken care of by the transport coefficients and the dissipative terms they precede. A well-known example is the second viscosity that arises from a slowly relaxing scalar quantity [9]. In the present situation, this means that $M$ and $H$ have to be given the local equilibrium value $||B$, while the fact that $M$ and $H$ are not quite in equilibrium is accounted for by the dissipative field $H^D$.

Part of the success of standard ferrohydrodynamics is certainly due to the fact it lies halfway between macroscopic and mesoscopic scales; the other half is attributable to the judicious handling of the theory. $M$'s evolution with time needs to be solved by considering simultaneity its equation of motion and the Maxwell equations, since both are strongly coupled. But this is not always easy. So intuitive shortcuts are taken to substitute for calculations; e.g., the rotation rate $\omega_M$ of the particles is either simply set to that of the field $\omega_f$, or $\omega_M = k \omega_f$ is taken with $k = 0.75$ [6]. (In fact, the latter is hardly a viable choice; there is no inherent reason for all the crystallites to rotate in phase while out of sync with the external field.) Less of practical relevance may be the fact that the standard theory of ferrohydrodynamics does not usually contain the particle concentration as an independent variable, although it is conserved and as such a bona fide long lived degree of freedom.

Now to comments on Henjes’s recent work [10], who starts from similar preconceptions, and provides a perceptive account of the state of the art of ferrohydrodynamics. Her results, however, are different. The dissipative field, $H^D = \alpha[\mathbf{B}_i - (\mathbf{\Omega} \times \mathbf{B}_i) - \mathbf{B}_i \mathbf{v}_i]$ from Eq. (6), alters the Maxwell and Navier-Stokes equations, and leads to an entropy production rate $(H^D)^2/\alpha$; cf. Eq. (15) below. In contrast, Henjes postulates an antisymmetric part of the dissipative stress tensor, $\epsilon_{ijk} \Pi_{ij}^D = \alpha_{ij} \Omega_{ij}$, which leads to the entropy production rate $\alpha_{ij} \Omega_{ij}$. This can be made to agree with the above expression for a solid body rotation in a static field, but not for shear flow nor for a time-dependent field. Especially, the equilibrium state of a solid body rotation with the field corotating, in which no entropy is produced, is not correctly accounted for.

To retain the symmetry of the total stress tensor, Henjes requires the reactive stress to also possess an antisymmetric part, given by $\mathbf{H} \times \mathbf{B}$. As criticized, this goes beyond local equilibrium. More fundamental, however, is the objection that $\Omega$ is an equilibrium quantity and not a thermodynamic force; the entropy production must not be expanded in it [9].

In the following, the complete hydrodynamic theory is derived. We start with the statics. The total energy density $\varepsilon$ is a function of $B$ and $D$, and a function of (the densities of) the following quantities: entropy $s$, total momentum $g^{tot} = \rho \mathbf{v} + \mathbf{E} \times \mathbf{H}/c$, particle mass $\rho_1$, and liquid mass $\rho_2$. Instead of the last three variables, however, it is more convenient to take the [11] momentum density $g = g^{tot} - \mathbf{D} \times \mathbf{B}/c$, the total density $\rho = \rho_1 + \rho_2$, and the density difference $\rho_e = \rho_1 - \rho_2$ as the independent variables:

$$d\varepsilon = \mu \rho d\rho + \mu_e d\rho_e + Tds + \nu_i d\mathbf{g}_i + H_i d\mathbf{B}_i + E_i d\mathbf{D}_i. \quad (10)$$

In equilibrium, maximizing the total entropy with appropriate constraints, we obtain a constant chemical potential $\mu$ and the vanishing of all the thermodynamic forces: $\nabla T$, $\nabla \rho_e$, $\mathbf{v}_i \times \mathbf{H}$, $\mathbf{v}_i \times \mathbf{E}$ [5,9,12].

Given the thermodynamics, Eq. (10), having identified the conserved quantities (especially $g^{tot}$) and the thermodynamic forces, the hydrodynamic equations can now
be derived employing the “standard hydrodynamic procedure”: Calculate \( \dot{e} \) via Eq. (10) and require the result to become \( \nabla \cdot \mathbf{Q} \) (where \( \mathbf{Q} \) is the energy flux). This is indeed how the usual hydrodynamics \([9,11]\) and the Maxwell equations \([5]\) are derived. A simultaneous derivation is what has been carried out here. The resultant equations, in addition to the Maxwell equations, Eqs. (1)–(3), are

\[
\dot{\rho} + \nabla_i (\rho \mathbf{v}_i) = 0, \quad \partial_t \rho + \nabla_i (\rho \mathbf{v}_i) + \nabla_i (\Pi_{ij} - \Pi^0_{ij}) = 0, \quad (11)
\]

\[
\dot{s} + \nabla_i (s \mathbf{v}_i - f^D_i) = R / T, \quad \dot{\rho}_c + \nabla_i (\rho_c \mathbf{v}_i - j^D_i) = 0, \quad (12)
\]

with the reactive part of the stress tensor \( \Pi_{ij} \), the energy flux \( \mathbf{Q} \), and the rate of entropy production tensor \( R \) given as

\[
\Pi_{ij} = \left[ (s \mathbf{v}_j - E_i D_j - H_i B_j + (i \leftrightarrow j))/2 + (s \mathbf{v}_j - E_i D_j - H_i B_j + (i \leftrightarrow j))/2 \right]
\]

\[
+ E_i D_j + H_i B_j - (i \leftrightarrow j)) \delta_{ij}, \quad (13)
\]

\[
\mathbf{Q}_i = (Ts + \mu \rho + \mu_c \rho + g \mathbf{v}_i) \mathbf{v}_i - T \mathbf{D}_i - \mu_c \mathbf{E}_i - \nabla \cdot \mathbf{Q}_i + \mathbf{R}_i \theta_{ij} + \int \mathbf{E}_i \cdot \mathbf{D}_j \mathbf{v}_j + (j^c - \mathbf{v} \cdot \mathbf{j}) \cdot \mathbf{E}_i
\]

\[
+ \mathbf{E}_i \cdot (c \nabla \times \mathbf{H}_i) - \mathbf{H}_i \cdot (c \nabla \times \mathbf{E}_i), \quad (14)
\]

Note the symmetric form of the stress tensor \( \Pi_{ij} = \Pi_{ji} \), which follows directly from the rotational identity, \( \mathbf{E} \times \mathbf{D} + \mathbf{H} \times \mathbf{B} + \mathbf{v} \times \mathbf{g} = 0 \). The explicit form of the dissipative terms in Eqs. (1), (11), and (12) are obtained from the rate of entropy production, Eq. (15), of which the fluxes, \( f^D, \ldots, \mathbf{E}^D, -\mathbf{H}^D \), are expanded in the forces, \( \mathbf{F} \),...,

The Heaviside-Lorentz system of units employed in this paper is by far the most convenient one in the present context. It is essentially the Gaussian system, but with all four fields reduced, and all four sources increased, by the factor of \( 2 / \sqrt{\pi} \). In other words, \( b^0 = b / \sqrt{\pi} \), similarly for \( D, H, E \), while \( \rho^0 = 2 / \sqrt{\pi} \rho \), similarly for \( j, p, M \).

For linear constitutive relations, Eqs. (3) and (4) reduce to the linear response expression \( \mathbf{E}^M = \mathbf{B}(\mu \mathbf{g}^1 + \mu_c \mathbf{g}^2) \),

\[
\mathbf{E}^M = \mathbf{D}(\epsilon^1 + i \epsilon^2 \mathbf{e}) \mathbf{E}^M = D / E, \quad \mu = 1 / \omega \alpha, \quad \text{and} \epsilon = 1 / \omega \beta, \quad \text{to lowest order either in the frequency} \omega \text{or the transport coefficients} \alpha, \beta.
\]

Finally, the boundary conditions: Barring surface charges and currents, the Maxwell equations, Eqs. (1) and (2), stipulate the continuity of the following quantities:

\[
\Delta D_{an}, \Delta B_{an}, \Delta (H + H^D), \Delta (E + E^D) = 0, \quad (18)
\]

which, in conjunction with the continuity of the total stress tensor, \( \Delta (\Pi_{ij} - \Pi^0_{ij}) = 0 \), lead to

\[
D_{an} \Delta E^D + B_{an} \Delta H^D - \Delta \Pi^0_{ij} = 0, \quad (19)
\]

or Eq. (7) for the considered situation.

I would like to thank S. Symalla for a critical reading of the manuscript, and for pointing out mistakes there.

*Electronic address: liu@itp.uni-hannover.de

References:


[7] The Heaviside-Lorentz system of units employed in this paper is by far the most convenient one in the present context. It is essentially the Gaussian system, but with all four fields reduced, and all four sources increased, by the factor of \( 2 / \sqrt{\pi} \). In other words, \( b^0 = b / \sqrt{\pi} \), similarly for \( D, H, E \), while \( \rho^0 = 2 / \sqrt{\pi} \rho \), similarly for \( j, p, M \).

[8] For linear constitutive relations, Eqs. (3) and (4) reduce to the linear response expression \( \mathbf{E}^M = \mathbf{B}(\mu \mathbf{g}^1 + \mu_c \mathbf{g}^2) \),

\[
\mathbf{E}^M = \mathbf{D}(\epsilon^1 + i \epsilon^2 \mathbf{e}) \mathbf{E}^M = D / E, \quad \mu = 1 / \omega \alpha, \quad \text{and} \epsilon = 1 / \omega \beta, \quad \text{to lowest order either in the frequency} \omega \text{or the transport coefficients} \alpha, \beta.
\]


