## **Off-Equilibrium, Static Fields in Dielectric Ferrofluids**

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The usual, frequency dependent permittivities  $\mu$  and  $\epsilon$  yield only an incomplete account of electric and magnetic dissipation when they are both important. Especially, a certain type of static field is ignored: Transverse, coupled, and dissipative. They vary exponentially in space, with a decay length that is macroscopic in select systems. [S0031-9007(98)05678-6]

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Linear response theory applies impeccable logic to derive the properties of the permittivities  $\epsilon$  and  $\mu$ . Without explicit reference, atomic scale distribution of charges and currents-both present and in the past-are accounted for. The real parts of  $\epsilon$  and  $\mu$  are even functions of the frequency  $\omega$ , they express the reactive response, such as the oscillatory motion of the microscopic charges in the presence of a periodic field. The imaginary parts are odd functions of  $\omega$ , they parametrize dissipation and absorption [1]. There is no doubt that a great deal of physics is captured by measuring  $\epsilon(\omega)$  and  $\mu(\omega)$ , and by calculating them for various systems. However, the diagonal structure of the constitutive relations, the fact that the electric field plays no role in the magnetic constitutive relation, and vice versa, the magnetic field does not partake in the electric one, is really an assumption that is hard to justify on general grounds, and as we shall see, is even in isotropic media not always correct.

When questioning whether these constitutive relations are (within their linear range of validity) general enough to cover all conceivable circumstances and any materials of interest, one must employ an independent macroscopic framework of at least equal rigor and standing. This is provided by the thermo- and hydrodynamic theory. Presuming local equilibrium, the hydrodynamic theory is valid for any field strength but confined to low frequencies. The linear response theory, on the other hand, is valid for arbitrary frequencies as long as the field is sufficiently weak. A comparison must therefore take place in the double limit of low frequencies and weak fields, where both ranges of validity overlap. Here, agreement in every detail must be expected, but is not found. And the discrepancy can be traced to the assumed diagonal structure of the constitutive relations, which contradicts basic thermo- and hydrodynamic considerations.

In the linear response theory, if electric and magnetic fields are static in a dielectric medium, they are also in equilibrium, and decoupled from each other. The hydrodynamic theory, on the other hand, allows them to be both time independent and dissipating—similar to a constant temperature gradient, or a constant electric field in a conductor. These stationary electric and magnetic fields are coupled and transverse; they start off from the boundary and extend over a certain distance into the bulk, defining a surface region. Although this distance is frequently tiny, and very ignorable, it turns macroscopic in magnetic systems such as some dielectric ferrofluids [2], where it is around 30 m. Clearly, one is hard pressed to find any bulk region here.

An equivalent statement is that spatial dispersion (i.e., spatial nonlocality and the wave vector dependence of  $\epsilon$  and  $\mu$ ) must not be neglected in these systems. This is plausible because the presence of a boundary makes itself felt over macroscopic distances, and because these stationary fields depend sensitively on the properties of the substance forming the boundary.

Take a slab of a dielectric ferrofluid, L = 1 cm wide. Expose this liquid to an oscillating electric or magnetic field, tangential to the slab, of a frequency  $\omega \ll c/L$ , and measure the internal field. Conventional wisdom holds that the result is a uniform internal field, E or H, that oscillates in phase with the external one and has the same magnitude, so D and B display a phase lag proportional to the imaginary part of the permeability,  $\epsilon''$ and  $\mu''$ . The hydrodynamic consideration includes the surface region mentioned above and depends on the type of the containing plates: If they are nonconducting, the phase lag has the same magnitude but an opposite sign. Consequently, irrespective of the experimental outcome, one of two theories feigns a negative  $\epsilon''$  and  $\mu''$ . If the plates are conducting, the prediction is even more striking, as the internal electric field should then be drastically reduced. These effects result directly from the existence of the dissipative, exponential fields, and probing them cuts both ways: If the hydrodynamic predictions are vindicated, the usual linear response theory is shown to be incomplete; if not, some basic considerations of the thermo- and hydrodynamic theory need scrutinizing.

In what follows—after a brief introduction to the perhaps less familiar hydrodynamic theory of electromagnetism—we shall first evaluate when, why, and how the linear response and the hydrodynamic theory differ, then discuss this experiment in appropriate details.

The hydrodynamic theory accounts for the complete low frequency behavior of systems that are polarizable, magnetizable, or charged [3]. It has been obtained by the time proven "standard procedure" to set up the hydrodynamic equations [4]. Though seemingly unorthodox

at times, it is rather elementary in its essence, and especially easy to comprehend by analogy. Consider a typical hydrodynamic equation, that of Navier-Stokes,  $\dot{g}_i$  +  $\nabla_j(\pi_{ij} + \pi_{ij}^D) = 0$ . The momentum density  $g_i = \rho v_i$ is a thermodynamic variable, odd under time inversion. The stress tensor  $\pi_{ij} + \pi_{ij}^{D}$  is the corresponding flux, with two parts: The reactive one is (if linearized) given by the pressure,  $\pi_{ij} = p \delta_{ij}$ , a thermodynamic derivative. It is even under time reversal, same as  $\dot{g}_i$ . The dissipative part of the flux is odd,  $\pi_{ij}^D = -\eta v_{ij} + \dots$ with  $v_{ij} \equiv \frac{1}{2} (\nabla_i v_j + \nabla_j v_i)$ . It breaks the time inversion symmetry of the Navier-Stokes equation and thereby accounts for dissipation and irreversibility. Generally, quantities such as  $v_{ii}$  or  $\nabla_i T$  are referred to as thermodynamic forces. Every thermodynamic variable has its thermodynamic force, the vanishing of which ensures equilibrium with respect to this variable. For instance,  $v_{ii} = 0$  ensures maximal entropy with respect to variations of the momentum density  $g_i$  for a given total momentum (i.e.,  $v_{ij} = 0$  is the respective Euler-Lagrange equation). If  $v_{ij}$ is finite, the entropy is not maximal, and the system not in equilibrium. There is then a dissipative momentum flux  $\sim v_{ii}$  that redistributes  $g_i$ , and an entropy production of the rate  $\sim v_{ij}^2$ . The coefficient in both cases is the viscosity  $\eta$ . Dissipative fluxes may also be proportional to other thermodynamic forces, and the constants are usually referred to as Onsager cross coefficients. Every statement in this paragraph has its analog in the next.

The Maxwell equations [5],  $\nabla \cdot \mathbf{D} = 0$ ,  $\nabla \cdot \mathbf{B} = 0$ ,

$$\dot{\mathbf{D}} = c\nabla \times \mathbf{H}^M, \qquad \dot{\mathbf{B}} = -c\nabla \times \mathbf{E}^M$$
 (1)

(of a stationary, dielectric substance), impose the analogy  $g \rightarrow \mathbf{D}$ , **B** and  $\pi + \pi^{\mathbf{D}} \rightarrow \mathbf{H}^{M}$ ,  $\mathbf{E}^{M}$ : The thermodynamic variables are **D** and **B**, being even and odd, respectively. Equations (1) are their equations of motion, while the two nontemporal Maxwell equations are constrains. The fields  $\mathbf{H}^{M}$  and  $\mathbf{E}^{M}$  appear only where fluxes do, they therefore split into reactive and dissipative parts,

$$\mathbf{H}^{M} = \mathbf{H} + \mathbf{H}^{D}, \qquad \mathbf{E}^{M} = \mathbf{E} + \mathbf{E}^{D}.$$
(2)

The reactive ones are (like the pressure) thermodynamic derivatives,  $\mathbf{H} \equiv \partial u / \partial \mathbf{B}$  and  $\mathbf{E} \equiv \partial u / \partial \mathbf{D}$ , *u* being the energy density. They contain only equilibrium information and are functions of all the thermodynamic variables. For weak fields, they reduce to  $\mathbf{H} = \mathbf{B}/\overline{\mu}$  and  $\mathbf{E} = \mathbf{D}/\overline{\epsilon}$ , where  $\overline{\mu}$  and  $\overline{\epsilon}$  depend on temperature and density, but not the frequency. The thermodynamic forces of  $\mathbf{D}$  and  $\mathbf{B}$  are  $c\nabla \times \mathbf{E}$  and  $c\nabla \times \mathbf{H}$ , respectively, the vanishing of which ensures maximal entropy with respect to variations in  $\mathbf{D}$  and  $\mathbf{B}$ —under the constraints of  $\nabla \cdot \mathbf{D} = 0$ and  $\nabla \cdot \mathbf{B} = 0$  [1,3]. So the dissipative fields  $\mathbf{H}^D$  and  $\mathbf{E}^D$  are proportional to these two forces, and possibly also to  $v_{ij}$  or  $\nabla_i T$ . For the isotropic case, symmetry considerations allow only

$$\mathbf{H}^{D} = -\alpha c \nabla \times \mathbf{E}, \quad \mathbf{E}^{D} = \beta c \nabla \times \mathbf{H} + \gamma \nabla T, \quad (3)$$
  
where  $\alpha$ ,  $\beta$ , and  $\gamma$  are Onsager coefficients. As  $\alpha c \nabla \times \mathbf{E}$  and  $\beta c \nabla \times \mathbf{H}$  are of opposite parity under time

reversal than  $\dot{\mathbf{D}}$  and  $\dot{\mathbf{B}}$ , respectively, they account for dissipation in the Maxwell equations, and contribute to the entropy production at the rate of  $\alpha (c\nabla \times \mathbf{E})^2$  and  $\beta (c\nabla \times \mathbf{H})^2$ . The coefficients are estimated [3] to be

$$\alpha = \tau_M(\overline{\mu} - 1)/\overline{\mu}, \qquad \beta = \tau_P(\overline{\epsilon} - 1)/\overline{\epsilon}, \quad (4)$$

while  $\gamma$ , probably smallish, may arise from the ordering of permanent dipoles along the heat current. As  $\gamma$  is in general a function of pressure,  $\gamma \nabla T$  does not necessarily have zero curl. And it may become a linear term via boundary conditions; cf. Eqs. (14) below.

This ends the review of the hydrodynamic Maxwell theory, and we turn to a comparison between the hydrodynamic and the linear response theory. Considering constant temperature, we may rewrite Eqs. (3) as

$$\mathbf{E}^{D} = \boldsymbol{\beta} \dot{\mathbf{D}} + \lambda^{2} \nabla \times \nabla \times \mathbf{E}, \mathbf{H}^{D} = \boldsymbol{\alpha} \dot{\mathbf{B}} + \lambda^{2} \nabla \times \nabla \times \mathbf{H},$$
(5)

where  $\lambda = c\sqrt{\alpha\beta}$ . If either  $\alpha$  or  $\beta$  is very small, both last terms may be neglected. Then, taking  $\mathbf{H} = \mathbf{B}/\overline{\mu}$ ,  $\mathbf{E} = \mathbf{D}/\overline{\epsilon}$ , and  $\partial_t \longrightarrow -i\omega$ , these two equations reduce to  $\mathbf{D} = \epsilon(\omega)\mathbf{E}^M$  and  $\mathbf{B} = \mu(\omega)\mathbf{H}^M$ , with

$$\boldsymbol{\epsilon}(\boldsymbol{\omega}) = \overline{\boldsymbol{\epsilon}}/(1 - i\boldsymbol{\omega}\boldsymbol{\beta}\overline{\boldsymbol{\epsilon}}), \quad \boldsymbol{\mu}(\boldsymbol{\omega}) = \overline{\boldsymbol{\mu}}/(1 - i\boldsymbol{\omega}\boldsymbol{\alpha}\overline{\boldsymbol{\mu}}).$$
(6)

And the hydrodynamic and linear response theory are demonstrated as equivalent to linear order in field and frequency. However, there are also cases in which  $\lambda$ cannot be neglected: Being essentially the relaxation time of magnetization and polarization,  $\alpha$  and  $\beta$  vary greatly, from  $\beta \approx 10^{-15}$  s for transparent dielectrics, to  $\alpha \approx$  $10^{-5}$  s for colloidal magnetic liquids; while water, with a permanent molecular dipole moment, is in the middle range,  $\beta \simeq 10^{-9}$  s. So a water-based ferrofluid should have a colossal  $\lambda \simeq 3 \times 10^3$  cm. Inserting Eqs. (5) into Eqs. (2), the three terms each are of order: unity,  $\omega \beta \overline{\epsilon}$ or  $\omega \alpha \overline{\mu}$ , and  $(q\lambda)^2$ —where the last one, with q the wave vector, is by no means always the smallest. If spatial dispersion quantified by  $\lambda$  cannot be neglected, it is sensible to return to the simpler hydrodynamic Maxwell equations, with the off-diagonal structure of Eq. (3), where  $\mathbf{H}^{D}$  is given by  $\mathbf{E}$ , and  $\mathbf{E}^{D}$  by  $\mathbf{H}$ .

In the conventional linear response theory, as in Eq. (5) with  $\lambda = 0$ , if a field configuration is stationary ( $\dot{\mathbf{D}}, \dot{\mathbf{B}} = \mathbf{0}$ ), it is also in equilibrium ( $\nabla \times \mathbf{E}, \nabla \times \mathbf{H} = 0$ ). Not so for the hydrodynamic theory: Taking (say)  $E_x$  and  $H_y \sim \exp(iqz - i\omega t)$  as the variables, Eqs. (1)–(3) reduce to a system of homogeneous, linear equations, and the solutions are obtained by setting the corresponding determinant to zero,

$$(\omega/\overline{c})^2 - q^2(1 - 2i\omega\tau) - q^4\lambda^2 = 0, \qquad (7)$$

$$\overline{c}^2 = c^2 / \overline{\mu}\overline{\epsilon}, \quad \tau = (\alpha \overline{\mu} + \beta \overline{\epsilon})/2, \quad \lambda^2 = \alpha \beta c^2.$$
 (8)

The roots are  $q_+ = \pm (1 + i\omega\tau)\omega/\overline{c}$ ,  $q_- = \pm i(1 - i\omega\tau)/\lambda$  for  $\omega\tau \ll 1$ ; and  $q_+^2 = i\omega\alpha\mu/\lambda^2$ ,  $q_-^2 = i\omega\beta\epsilon/\lambda^2$  for  $\omega\tau \gg 1$ . The first is clearly the electromagnetic wave, the last two a pair of diffusive modes;

the second survives the  $\omega \rightarrow 0$  limit,  $q_{-} = \pm i/\lambda$ , it is a stationary mode in which  $\nabla \times \mathbf{E}$ ,  $\nabla \times \mathbf{H}$ , and hence the entropy production, are finite. As announced, these stationary fields are coupled, dissipative, and transverse. One way to find the ratio of amplitudes of the associated fields is by setting the time derivative to zero in the second of Eqs. (1), leading to

$$E_x = \mathcal{T}_{\pm} e^{\pm z/\lambda}, \qquad H_y/E_x = \pm \sqrt{\alpha/\beta}.$$
 (9)

If  $\omega \neq 0$  (yet  $\omega \tau \ll 1$ ), the fields  $E_x$  and  $H_y$  retain their spatial dependence, but the amplitudes oscillate in time,  $\sim \exp(-i\omega t \pm z/\lambda)$ . The characteristic polynomial, Eq. (7), is rather typical of the hydrodynamic theory and accounts (with different definitions of  $\overline{c}$ ,  $\tau$ , and  $\lambda$ ) for collective modes in such diverse broken symmetry systems as superfluid <sup>3</sup>He and liquid crystals [6]. In analogy to these systems, we shall refer to  $q_- = \pm i/\lambda$  as the (electromagnetic) sq-mode.

A dielectric medium is an idealized concept, as the conductivity  $\sigma$  is never truly zero. The general spectrum of the sq-mode,  $\lambda^2 = \alpha \beta c^2/(1 + \sigma \beta)$ , remains unchanged from above if  $\sigma \beta \ll 1$ , i.e., if the relaxation time  $\tau_P$  of the polarization is much smaller than the charge relaxation time  $1/\sigma$  [5].

In substances in which  $\overline{\mu}$  or  $\overline{\epsilon} \gg 1$ , we should be able to find a frequency window, hydrodynamic yet  $\omega \tau \gg 1$ . Here, the two diffusive modes calculated above exist, where  $q_-$  is purely electric and  $q_+$  magnetic. They are independent and decoupled, in contrast to the single diffusive mode (with the coefficient  $c^2/\sigma$ ) in conductors.

We now proceed to derive the appropriate boundary conditions for the hydrodynamic Maxwell equations, without which no relevant discussions of concrete experimental situations is possible. The equations of motion for D and B, Eqs. (1), are an order higher in spatial derivative and possess additional collective modes; they need more boundary conditions to uniquely determine any solution. These are derived from the hydrodynamic equations [3], employing a recently developed procedure [7]; see also [6]. The resultant boundary conditions are rather general, valid where the hydrodynamic theory is, and depend on the same input as the bulk theory: conservation laws, broken symmetries, and irreversible thermodynamics. The microscopic information about the boundary is parametrized in surface Onsager coefficients, the magnitude of which is unknown. As any transport coefficient, they need to be determined either experimentally, for a given pair of substance forming the interface, or in a microscopic calculation employing a specific model. There are two types of boundary conditions: The first states the continuity of the normal component of the fluxes, of those variables that are defined and independent on both sides of the interface. These are obtained by integrating the hydrodynamic equations over an infinitesimally narrow slab around the interface. The second type are Onsager relations given by the surface entropy production  $R_s$ , the expression for which is extracted from the continuity of the total energy flux (that contains both material and field contributions [3]).

We study two kinds of interfaces, vacuum-ferrofluid (VFI) and conductor-ferrofluid (CFI). Both "vacuum" and "conductor" stand for an electromagnetically inert medium that is only weakly dissipative, with  $\alpha$  and  $\beta$ so small that  $\lambda$  is microscopic, and  $E^D$ ,  $H^D$  negligible for the given frequency. It behaves vacuumlike if nonconducting, and possesses both E and H as independent variables. If it is conducting,  $\sigma \gg \omega$ , only the magnetic field is retained as an independent variable, with the electric one given by  $\mathbf{E} = c\nabla \times \mathbf{H}/\sigma$ . (Since all substances have finite conductivities, the condition  $\sigma \gg \omega$ can always be satisfied by lowering the frequency. So conductor may be taken to stand for the low frequency limit.) Finally, "ferrofluid" stands for any strongly dissipative system that is nonconducting, dielectric, and magnetic, in which both  $E^D$  and  $H^D$  are important. The boundary conditions for the VFI and CFI are, respectively,

$$\Delta \mathbf{H}_{t}^{M} = 0, \quad \Delta \mathbf{E}_{t}^{M} = 0, \quad \zeta_{1} \mathbf{H}_{t}^{D} = \mp \mathbf{E}_{t}^{D} \times \hat{\mathbf{n}}; \quad (10)$$
  
$$\Delta \mathbf{E}_{t}^{M} = 0, \quad \mathbf{E}^{M} = \zeta_{2} \Delta \mathbf{H} \times \hat{\mathbf{n}}, \quad \mathbf{E} = \mp \zeta_{3} \mathbf{H}^{D} \times \hat{\mathbf{n}}; \quad (11)$$

in addition to  $\triangle D_n, \triangle B_n = 0$ . (Notations:  $\hat{\mathbf{n}}$  is the interface normal;  $\triangle A \equiv A_{\text{left}} - A_{\text{right}}$ ;  $A_n$  and  $\mathbf{A}_t$  are the normal and perpendicular components of  $\mathbf{A}, A_n \equiv \mathbf{A} \cdot \hat{\mathbf{n}}$ with  $\hat{\mathbf{n}}$  pointing to the right, say along  $\hat{\mathbf{z}}$ .) The first two of Eqs. (10) and the first of Eqs. (11) belong to the first type of boundary conditions and state the continuity of fluxes. ( $\triangle \mathbf{H}_{t}^{M} = 0$  is not valid at the CFI because D, the corresponding variable, is not independent in the conductor.) The rest are three Onsager relations, with  $\zeta_1, \zeta_2, \zeta_3 > 0$  and all quantities referring to the ferrofluid. The upper sign holds if the ferrofluid is on the right; the lower one does if it is on the left. The last of Eqs. (10) is derived by inserting  $\Delta \mathbf{H}_t^M, \Delta \mathbf{E}_t^M = 0$  in the expression for the total energy flux, to obtain  $\triangle Q_n = T \triangle f_n \mp c[\mathbf{H}^{\mathbf{D}} \cdot (E^D \times \hat{\mathbf{n}})] + \ldots = 0$ , with  $f_n$  the entropy flux. Identifying  $-T \triangle f_n$  as  $R_s$ , the positive, singular entropy production of the surface,  $\mathbf{H}_{t}^{\mathbf{D}}$  and  $\mp (\mathbf{E}_{t}^{\mathbf{D}} \times \hat{\mathbf{n}})$  are shown to be a thermodynamic force-flux pair, proportional to each other. Isotropy of the interface then allows only the scalar Onsager coefficient  $\zeta_1$ . (For the sake of simpler display, no cross coefficients have been included.) The two Onsager relations of Eqs. (11) are derived in a similar manner, the only difference being the lack of  $\Delta \mathbf{H}_t^M = 0$ . The entropy production  $R_s$  therefore contains two terms instead of one, yielding two Onsager relations. Note that  $\triangle H = 0$  is retrieved if  $E^M$ , a nonequilibrium quantity in conductors, vanishes.

Finally, we turn to discussing the experiments mentioned in the introduction: A slab of dielectric ferrofluid is placed between two plates and exposed to an oscillating external field, electric or magnetic, of quasistationary frequency  $\omega \ll \overline{c}/L$ . The plates are vacuumlike or conducting in the above sense, oriented perpendicular to  $\hat{z}$  and located at z < 0 and z > L, respectively. The fields in the plates are  $\mathbf{E} = E_{\text{ex}}(t)\hat{\mathbf{x}}$  and  $\mathbf{H} = H_{\text{ex}}(t)\hat{\mathbf{y}}$  if they are nonconducting, with  $E_{\text{ex}}$ ,  $H_{\text{ex}}$  uniform (and  $\hat{\mathbf{x}}$ ,  $\hat{\mathbf{y}}$ ,  $\hat{\mathbf{z}}$  denoting unit vectors). In the conducting plates, with  $\omega \ll \sigma$  and a correspondingly large penetration depth, the magnetic field acquires a constant gradient:  $\mathbf{H} = (H_{\text{ex}}^L - \sigma E_{\text{ex}} z/c)\hat{\mathbf{y}}$  for z < 0 and  $\mathbf{H} = [H_{\text{ex}}^R - \sigma E_{\text{ex}}(z - L)/c]\hat{\mathbf{y}}$  for z > L, though the dependent electric field remains uniform,  $\mathbf{E} = E_{\text{ex}}\hat{\mathbf{x}}$ . Inside the ferrofluid,  $0 \le z \le L$ , the field can be taken as

$$\mathbf{E} = [E_0 + \mathcal{E}_+ \exp^{(z-L)/\lambda} + \mathcal{E}_- \exp^{-z/\lambda}] \mathbf{\hat{x}}, \quad (12)$$
$$\mathbf{H} = \left[ H_0 + \sqrt{\alpha/\beta} \left( \mathcal{E}_+ \exp^{(z-L)/\lambda} - \mathcal{E}_- \exp^{-z/\lambda} \right) \right] \mathbf{\hat{y}}. \quad (13)$$

The task now is to determine the unknown amplitudes  $E_0$ ,  $H_0$ ,  $\mathcal{E}_+$ , and  $\mathcal{E}_-$  from the external fields,  $E_{\text{ex}}$  and  $H_{\text{ex}}$ . Conventionally, we expect  $E_0 = E_{\text{ex}}(t)$ ,  $H_0 = H_{\text{ex}}^L(t) = H_{\text{ex}}^R(t)$ , from  $\Delta E$ ,  $\Delta H = 0$ ; and  $\mathcal{E}_{\pm} = 0$ , since the linear response theory does not contain the sq-mode. The hydrodynamic results are obtained employing the boundary conditions, Eqs. (10) and (11), for z = 0 and z = L.

First, the case of the vacuumlike plates: The first two of Eq. (10) yield  $E_0 + \gamma \nabla_x T = E_{\text{ex}}$  and  $H_0 = H_{\text{ex}}$ , while the third determines the sq-amplitude. If the width of the ferrofluid slab is much larger than the sq-decay length,  $L \gg \lambda$  (possible if the magnetic particles are small, the polarization relaxes quickly, or the substance is macromolecular rather than ferrofluid), then the sq-modes are well formed, with the amplitudes

$$\mathcal{I}_{\pm} = (\beta \dot{D} + \gamma \nabla_x T \pm \zeta_1 \alpha \dot{B}) / A_1^{(+)}, \qquad (14)$$

where

$$A_k^{(\pm)} = \zeta_k \sqrt{\alpha/\beta} \pm 1.$$
 (15)

Approximating  $\dot{D} \approx \overline{\epsilon} \dot{E}_{ex}$ ,  $\dot{B} \approx \overline{\mu} \dot{H}_{ex}$  for  $\gamma \longrightarrow 0$  (because  $\mathcal{E}_{\pm}/E_0$ ,  $\mathcal{E}_{\pm}/H_0$  are then of order  $\omega \beta \overline{\epsilon}$ ,  $\omega \alpha \overline{\mu} \ll 1$ ), we conclude that the sq-mode can always be excited, irrespective of the value of  $\zeta_1$ : via  $\dot{H}_{ex}$  for  $\zeta_1 \longrightarrow \infty$ , and via  $\dot{E}_{ex}$  if  $\zeta_1 \longrightarrow 0$ . If  $\gamma$  is sufficiently large,  $\dot{E}_{ex}$  can be substituted by a transverse temperature gradient.

In the opposite limit, for  $L \ll \lambda$ , the sq-amplitudes are  $\mathcal{E}_{\pm} = \pm \frac{1}{2} \sqrt{\alpha \beta} \dot{B} + (\beta \dot{D} + \gamma \nabla_x T) / [2 + A_1^{(-)} L / \lambda].$ (16)

The approximations  $\dot{D} \approx \overline{\epsilon} \dot{E}_{ex}$  and  $\dot{B} \approx \overline{\mu} \dot{H}_{ex}$  (with the additional, possibly stricter condition  $A_1^{(\pm)} L/\lambda \ll 1$  for the electric case), then lead to the phase lags

$$D = \overline{\epsilon} E_{\text{ex}}(t) \exp(-i\omega\beta\overline{\epsilon}), \qquad (17)$$

$$B = \overline{\mu} H_{\text{ex}}(t) \exp(-i\omega \,\alpha \,\overline{\mu}) \,. \tag{18}$$

Compare these to the linear response theory: Inserting Eqs. (6) into  $D = \epsilon(\omega)E_{\text{ex}}$  and  $B = \mu(\omega)H_{\text{ex}}$ , the respective phase lag has the same magnitude, but a different sign. Using these formulas to interpret the experimental outcome would therefore lead to the false conclusion that  $\alpha \sim \mu''$  and  $\beta \sim \epsilon''$  were negative.

Next we consider the conducting plates. The first two of Eqs. (11) yield  $E_0 = E_{\text{ex}}$  and  $H_0 = \frac{1}{2}(H_{\text{ex}}^L - H_{\text{ex}}^R)$ , the third again determines the sq-amplitude,

$$\mathcal{E}_{\pm} = E_{\rm ex} / (e^{-L/\lambda} A_2^{(-)} - A_2^{(+)}).$$
(19)

In the narrow slab limit,  $L \ll \lambda$ , and assuming that  $\zeta_2$  is such that  $A_2^{(\pm)}L/\lambda \ll 1$ , we have

$$\frac{E}{E_{\rm ex}} = \frac{\zeta_2 L}{2\lambda} \sqrt{\frac{\alpha}{\beta}}, \qquad \frac{H_0 - H}{E_{\rm ex}} = \frac{z - L/2}{\lambda} \sqrt{\frac{\alpha}{\beta}}.$$
(20)

The *E* field vanishes in the limit  $L/\lambda \rightarrow 0$  because the constant term  $\sim (L/\lambda)^0$  in  $\mathcal{I}_{\pm}$  exactly cancels  $E_0$  in Eq. (12). With L = 1 cm and  $\lambda = 30$  m, the quotient  $L/\lambda$ is indeed small in the envisioned experiment. So although  $\zeta_2$  is unknown, unless it has some symmetry reasons to diverge consistently, we may still conclude  $E \ll E_{\text{ex}}$ .

A static and uniform field, electric or magnetic and normal to the surface, modifies the above considerations in two aspects: (i) If the field is strong enough, the system is no longer isotropic. This leads to many more possible couplings, with a proliferation of unknown Onsager coefficients. (ii) More immediately, fields normal to the interface give rise to ponderomotive surface forces,  $\mathbf{f}_m^{\mathbf{D}} = \mathbf{B} \times (\nabla \times \mathbf{H}^{\mathbf{D}})$ ,  $\mathbf{f}_e^{\mathbf{D}} = \mathbf{D} \times (\nabla \times \mathbf{E}^{\mathbf{D}})$ , which have been shown to account for the peculiar spin-up behavior of ferrofluids under a rotating external field [3]. Given the presence of an sq-mode, these forces penetrate into the bulk and need to be compensated by the viscous stress tensor there,  $\eta v_t'' + f_m^{D} + f_e^{D} = 0$ . As a result, the transverse velocity participates in the exponential excursion of the sq-mode.

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