PHYSICAL REVIEW E VOLUME 59, NUMBER 3 MARCH 1999

Electromagnetic fields in ferrofluids

Mario Liu*
Institut für Theoretische Physik, Universität Hannover, 30167 Hannover, Germany

(Received 29 September 1998)

The behavior of electromagnetic fields in dielectric ferrofluids is considered in detail by employing two independent methods: linear response and hydrodynamic Maxwell equations. The respective results differ in an experimentally relevant way. As the calculation takes place where the ranges of validity of both theories overlap, weak fields and small frequencies, this discrepancy calls for an understanding. The conclusion drawn here is that the linear response theory is the culprit, and its results are faulty. This paper contains especially the algebraic details that were left out of a previous, brief publication [Mario Liu, Phys. Rev. Lett., **80**, 2937 (1998)]. [S1063-651X(99)07603-5]

PACS number(s): 75.50.Mm, 41.20.−q

I. INTRODUCTION

Linear response theory applies impeccable logic to derive the properties of the permittivities ϵ and μ. Without explicit reference, atomic scale distribution of charges and currents — both present and in the past — are accounted for. The real parts of ϵ and μ are even functions of the frequency ω; 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 ω; 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 not always correct even in isotropic media.

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 thermodynamic 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 thermodynamic and hydrodynamic considerations.

In 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 some magnetic systems such as dielectric ferrofluids [2], where it is around 30 m. Clearly, one is hard pressed to find any bulk region here.

In what follows, in Sec. II, we shall first compare two ways to close the macroscopic Maxwell equations, via linear response or the hydrodynamic theory. Then, in Sec. III, a one-dimensional, static, and dissipative solution is presented that is contained only in the hydrodynamic theory. In Sec. IV, the appropriate boundary conditions needed to calculate the associated amplitudes are derived. In the last two sections, two experiments are discussed in which this static solution is important.

Although this paper is the long version of a previously published short communication [3], only part of the discussion there is repeated here. So the reader is advised also to consult Ref. [3].

All formulas in this paper are in the MKSA system of units, for easy comparison to experiments. The original formulas [3] were in the Heaviside-Lorentz units. It is easy to go from one to the other system at any point in this paper by employing the following formulas, in which the fields in MKSA are denoted with hats, such as $\hat{E}$, $\hat{H}$, etc.:

$$\hat{H}=H/\sqrt{\mu_o}, \quad \hat{B}=B\sqrt{\mu_o}, \tag{1}$$

$$\hat{E}=E/\sqrt{\epsilon_o}, \quad \hat{D}=D\sqrt{\epsilon_o}, \tag{2}$$

$$\hat{\varrho}_e=\varrho_e\sqrt{\epsilon_o}, \quad \hat{j}_e=j_e\sqrt{\epsilon_o}, \quad \hat{\sigma}=\sigma\epsilon_o. \tag{3}$$

II. MAXWELL EQUATIONS

The macroscopic Maxwell equations of a stationary, dielectric, magnetizable, and polarizable substance are

$$\dot{\mathbf{D}}=\nabla\times\mathbf{H}^M, \quad \nabla\cdot\mathbf{D}=0, \tag{4}$$

$$\dot{\mathbf{B}}=-\nabla\times\mathbf{E}^M, \quad \nabla\cdot\mathbf{B}=0. \tag{5}$$

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

As such, these four equations contain little useful information (aside from general principles such as Lorentz covariance and charge conservation). Their considerable predictive power only comes into play when they are closed — frequently by employing the linear response theory. Alternatively, we may employ the hydrodynamic Maxwell theory that was derived only recently by including electromagnetic dissipation systematically [3,4].

A. Linear response theory

The linear response theory starts from the diagonal constitutive relations

$$\mathbf{D}=\epsilon(\omega)\mathbf{E}^M, \quad \mathbf{B}=\mu(\omega)\mathbf{H}^M. \tag{6}$$

Since we are only interested in the low frequency behavior, we shall exclude, in the inverse permeabilities $1/\epsilon(\omega)$ and $1/\mu(\omega)$, terms higher than linear in the frequency ω. So, for $\partial_t \leftrightarrow -i\omega$, we have, quite generally,

$$\epsilon(\omega)=\frac{\bar{\epsilon}}{1-i\omega\beta\bar{\epsilon}/\epsilon_0}, \quad \mu(\omega)=\frac{\bar{\mu}}{1-i\omega\alpha\bar{\mu}/\mu_0}. \tag{7}$$

The static permeabilities are $\bar{\epsilon}=\epsilon(\omega\to 0), \bar{\mu}=\mu(\omega\to 0)$, while α and β parametrize the imaginary parts of $\epsilon(\omega)$ and $\mu(\omega)$. All four parameters are real, positive, frequency independent, and chosen to coincide with the hydrodynamic ones below.

B. Hydrodynamic theory

The Maxwell equations (4) and (5) remain, but the constitutive relations are given as [3,4]

$$\mathbf{H}^M=\mathbf{H}+\mathbf{H}^D, \quad \mathbf{E}^M=\mathbf{E}+\mathbf{E}^D, \tag{8}$$

where

$$\mathbf{H}\equiv \partial u/\partial \mathbf{B}, \quad \mathbf{E}\equiv \partial u/\partial \mathbf{D} \tag{9}$$

are thermodynamic derivatives, u being the energy density. They contain only equilibrium information, and are functions of all the thermodynamic variables such as temperature and pressure.

(Note an important step here: Taking **D** and **B** as the variables, and the two temporal of the Maxwell equations as the associated equations of motion, the hydrodynamic theory distinguishes between $\mathbf{E},\mathbf{H}$ and $\mathbf{E}^M,\mathbf{H}^M$: The first are the thermodynamic conjugate variables, the latter the respective fluxes of $\dot{\mathbf{D}},\dot{\mathbf{B}}$, defined solely by the Maxwell equations — they are therefore given the superscript ^{M}, and referred to as the Maxwell fields below. Usually, this differentiation is not made, leading necessarily to the caveat that the basic thermodynamic form

$$\mathrm{d}u=\mathbf{H}\cdot \mathrm{d}\mathbf{B}+\mathbf{E}\cdot \mathrm{d}\mathbf{D}$$

is only valid for nondissipative systems [1].)

Returning to the linear, weak field, limit, the thermodynamic, constitutive relations reduce to

$$\mathbf{H}=\mathbf{B}/\bar{\mu}, \quad \mathbf{E}=\mathbf{D}/\bar{\epsilon}, \tag{10}$$

where $\bar{\mu},\bar{\epsilon}>0$ depend on temperature and density, but not the frequency. The dissipative fields $\mathbf{H}^D$ and $\mathbf{E}^D$ are (for the simplest case)

$$\mathbf{H}^D=-(\alpha/\mu_0)\nabla\times\mathbf{E}, \quad \mathbf{E}^D=(\beta/\epsilon_0)\nabla\times\mathbf{H}, \tag{11}$$

where $\alpha,\beta>0$ represent Onsager (or transport) coefficients, similar to the viscosity or a diffusion coefficient.

If either α or β vanishes, the hydrodynamic theory is identical to the linear response theory: If (say) $\alpha=0$, then $H^D=0$; therefore $\dot{D}=\nabla\times H$, hence $E^D=(\beta/\epsilon_0)\dot{D}$, so $E^M=D/\bar{\epsilon}+(\beta/\epsilon_0)\dot{D}$, or $\mathbf{D}/\bar{\epsilon}=\mathbf{E}^M/(1-i\omega\beta\bar{\epsilon}/\epsilon_0)$, as in Eqs. (6) and (7).

However, there are quite a number of systems in which α and β are both finite. Then the linear response and hydrodynamic theory are not equivalent. The purpose of this paper is to work out the difference, and find an experimentally relevant situation in which this difference is large.

Strictly speaking, the significance of α and β have already been introduced above: They parametrize the imaginary parts of the permeabilities in the linear response theory, and are transport coefficients in the hydrodynamic theory. However, it is frequently useful to have a more intuitive understanding: In a simple, relaxative, model, α and β are closely related to τ_M and τ_P, the relaxation time of magnetization and polarization, respectively:

$$\alpha=\tau_M(\bar{\mu}-\mu_0)/\bar{\mu}, \quad \beta=\tau_P(\bar{\epsilon}-\epsilon_0)/\bar{\epsilon}. \tag{12}$$

(See the Appendix for a derivation of these formulas.)

Hydrodynamic theories are only valid for small frequencies, and the one considered here is valid for

$$\omega\tau_M\ll 1, \quad \omega\tau_P\ll 1. \tag{13}$$

[Aiming for an accuracy of, say 10%, the limit is $\omega\tau_M\approx 0.3$, as the neglected terms are $\sim(\omega\tau_M)^2$, in the real part of μ.] In the notation of this paper, Eqs. (12) and (13) imply

$$\beta\omega\ll 1, \quad \alpha\omega\ll 1. \tag{14}$$

Two also rather frequent combinations are $\bar{\epsilon}\beta\omega$, and $\bar{\mu}\alpha\omega$ which, depending on system properties, need not be small compared to 1.

III. A DISSIPATIVE STATIC SOLUTION

The static solutions of the linear response theory are those every student of physics is familiar with, from introductory lectures in electromagnetism. Setting $\omega\to 0$ in Eqs. (7), the Maxwell equations (4) and (5) only contain solutions that are either electric or magnetic, given, respectively, by

$$\nabla\times\mathbf{E}=0, \quad \nabla\cdot(\bar{\epsilon}\mathbf{E})=0, \tag{15}$$

$$\nabla\times\mathbf{H}=0, \quad \nabla\cdot(\bar{\mu}\mathbf{B})=0. \tag{16}$$

In one-dimensional geometries — of width L — these are simply constant fields. If the boundary conditions change with time slowly, oscillating with a frequency in the quasistatic regime, $c/\omega\gg L$, the internal field will also oscillate, but remain constant in space.

If either α or β vanishes, the hydrodynamic theory is, as we saw, identical to the linear response theory; if both α and β are finite, it contains more structure. Setting $\dot{D}$, $\dot{B}=0$ in Eqs. (4) and (5), the set of equations to be solved are

$$\nabla\times[\mathbf{H}-(\alpha/\mu_0)\nabla\times\mathbf{E}]=0,\quad \nabla\cdot(\overline{\mu}\mathbf{B})=0, \tag{17}$$

$$\nabla\times[\mathbf{E}-(\beta/\epsilon_0)\nabla\times\mathbf{H}]=0,\quad \nabla\cdot(\overline{\epsilon}\mathbf{E})=0. \tag{18}$$

These are satisfied by the one-dimensional, exponential solution, in which both the electric and magnetic field participate,

$$\mathbf{E}=E_0\hat{\mathbf{x}}+\ (1/\sqrt{\epsilon_0})\ [\mathcal{E}_+e^{(z-L)/\lambda}+\mathcal{E}_-e^{-z/\lambda}]\hat{\mathbf{x}}, \tag{19}$$

$$\mathbf{H}=H_0\hat{\mathbf{y}}+\sqrt{\alpha/\beta\mu_0}[\mathcal{E}_+e^{(z-L)/\lambda}-\mathcal{E}_-e^{-z/\lambda}]\hat{\mathbf{y}}, \tag{20}$$

where

$$\lambda=c\sqrt{\alpha\beta}. \tag{21}$$

Being essentially the relaxation time of magnetization and polarization, cf. Eq. (12), the coefficients α and β vary greatly, from $\beta\approx10^{-15}$ s for transparent dielectrics, to $\alpha\approx10^{-5}$ s for ferrofluids; while water, with a permanent molecular dipole moment, is in the middle range, $\beta\simeq10^{-9}$ s. So a water-based ferrofluid should have a colossal $\lambda\simeq3\times10^3$ cm. Clearly, if we can identify situations in which the amplitudes of the exponential decay $\mathcal{E}_\pm\neq0$, linear response theory is proven wrong — for weak fields and linear constitutive relations.

The above calculation assumes a dielectric medium with zero conductivity. This is of course an idealized concept, as the conductivity σ is never truly zero. In fact, strictly speaking, any system becomes conducting in the limit $\omega\to0$, for which case the above calculation needs to be generalized, yielding the decay length

$$\lambda^2=\alpha\beta c^2/(1+\sigma\beta/\epsilon_0). \tag{22}$$

It remains unchanged from Eq. (21) if $\sigma\beta/\epsilon_0\ll1$, i.e., if the relaxation time τ_P of the polarization is much smaller than the charge relaxation time ϵ_0/σ.

IV. BOUNDARY CONDITIONS

The boundary conditions of the linear response theory are

$$\Delta\mathbf{E}_t^M=0,\quad \Delta\mathbf{H}_t^M=0 \tag{23}$$

for the components tangential to the interface, and

$$\Delta D_n=0,\quad \Delta B_n=0 \tag{24}$$

for the normal ones. They are obtained by integrating the Maxwell equations over an infinitesimally narrow slab across the boundary. (Notations, here and especially below: $\hat{\mathbf{n}}$ is the interface normal; $\triangle A\equiv A_{\rm left}-A_{\rm right}$; and 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 $\hat{\mathbf{z}}$.)

Because the associated differential equations (17) and (18) are an order higher in spatial derivative, the hydrodynamic Maxwell theory possesses more boundary conditions. (This comes in handy, of course, since the amplitudes $\mathcal{E}_\pm$ need definite values for any solution to be uniquely determined.) As in the case of the linear response, these boundary conditions are derived from the equations of motion themselves. (Therefore, they should not be considered as independent and additional information, which is the usual point of view in mathematics.)

There is a useful formalism for doing this, developed for hydrodynamic theories of systems with spontaneously broken symmetries [5]. It yields boundary conditions that are rather general, valid where the hydrodynamic theory is. Both depend on the same input: 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 within the given frame. As any transport coefficient, they need to be determined either experimentally, for a given pair of substances 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, as in Eqs. (23) and (24). 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 (which contains both the material and field contributions [4]).

We study two types 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 α and β so small that λ is microscopic, and E^D, and H^D are negligible for the given frequency. It behaves like a vacuum if nonconducting, and possesses both E and H as independent variables. If it is conducting, $\sigma/\epsilon_0\gg\omega$, only the magnetic field is retained as an independent variable, with the electric one given by $\mathbf{E}=\nabla\times\mathbf{H}/\sigma$. (Since the condition $\sigma/\epsilon_0\gg\omega$ can always be satisfied by lowering the frequency, the ''conductor'' may be taken as the low frequency limit.) Finally, ''ferrofluid'' stands for any system (i) that is nonconducting (for the given frequency), (ii) is strongly polarizable and magnetizable, and (iii) in which both E^D and H^D are important. The boundary conditions for the VFI are

$$\triangle\mathbf{H}_t^M=0,\quad \triangle\mathbf{E}_t^M=0, \tag{25}$$

$$\zeta_1\mathbf{H}_t^D=\mp\sqrt{\epsilon_0/\mu_0}\quad \mathbf{E}_t^D\times\hat{\mathbf{n}}. \tag{26}$$

Those for the CFI are

$$\triangle\mathbf{E}_t^M=0, \tag{27}$$

$$\sqrt{\epsilon_0/\mu_0}\quad \mathbf{E}^M=\zeta_2\triangle\mathbf{H}\times\hat{\mathbf{n}}, \tag{28}$$

$$\sqrt{\epsilon_0/\mu_0}\quad \mathbf{E}=\mp\zeta_3\mathbf{H}^D\times\hat{\mathbf{n}}; \tag{29}$$

in addition to $\triangle D_n=0$ and $\triangle B_n=0$.

Equations (25) and (27) belong to the first type of boundary conditions, and state the continuity of fluxes. [$\triangle\mathbf{H}_t^M=0$ is not a generally valid boundary condition at the CFI, as the corresponding variable D is not an independent one on the conductor side. So instead we have $\triangle \mathbf{H}_t^M \sim \mathbf{E}^M$, $\mathbf{E}^D$, cf. Eq. (30).]

The rest are Onsager relations, with three dimensionless coefficients $\zeta_1, \zeta_2, \zeta_3 > 0$, 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. Equation (26) is derived by starting from the expression for the total energy flux [4],

$$\mathbf{Q} = T\mathbf{f} + \mathbf{E}^M \times \mathbf{H}^M - \mathbf{E}^D \times \mathbf{H}^D + \cdots, \tag{30}$$

with $\mathbf{f}$ the entropy flux and T the temperature. Inserting (i) $\triangle \mathbf{H}_t^M, \triangle \mathbf{E}_t^M = 0$, and (ii) $\mathbf{E}^D$, $\mathbf{H}^D = 0$ on the vacuum side, we obtain

$$\triangle Q_n = T \triangle f_n + f_n \triangle T \mp [\mathbf{H}^\mathbf{D} \cdot (\mathbf{E}^\mathbf{D} \times \hat{\mathbf{n}})] + \cdots, \tag{31}$$

where $\triangle Q_n = 0$ because of energy conservation. Identifying $-T\triangle f_n$ as the positive, singular entropy production of the surface R_s, the two terms $\mathbf{H}_\mathbf{t}^\mathbf{D}$ and $\mp(\mathbf{E}_\mathbf{t}^\mathbf{D} \times \hat{\mathbf{n}})$ are shown to be a thermodynamic force-flux pair which, (similar to f_n and $\triangle T$ giving rise to the Kapitza resistance,) are proportional to each other. Isotropy of the interface then only allows the scalar Onsager coefficient ζ_1. The factor $\sqrt{\epsilon_0/\mu_0}$ was included to render ζ_1 dimensionless.

The two Onsager relations, Eqs. (28) and (29), are derived in a similar manner, the only difference being the lack of $\triangle \mathbf{H}_t^M = 0$. The entropy production $R_s \equiv -T\triangle f_n$, therefore, contains two terms instead of one,

$$-T\triangle f_n = f_n \triangle T + [\mathbf{E}^M \times \triangle \mathbf{H} \mp \mathbf{E} \times \mathbf{H}^D] \cdot \hat{\mathbf{n}} + \cdots \tag{32}$$

yielding two Onsager relations, Eqs. (28) and (29). Note that $\triangle H = 0$ is retrieved from Eq. (28) if E^M, a nonequilibrium quantity in conductors, vanishes. (For the sake of simpler display, no cross coefficients have been included.)

V. FIRST EXPERIMENT, DIELECTRIC BOUNDARIES

A. Linear response prediction

Consider a $\hat{z}$ axis with three regions: $z<0$ is region 1, $0<z<L$ is region 2, and $z>L$ is region 3. Only region 2 (with a width say of $L=1$ cm) contains ferrofluid; regions 1 and 3 consist of a dielectric substance with negligible field dissipation for the given frequencies: $H^D, E^D = 0$. (It is the ''vacuum'' in the above sense.) Applying tangential fields $\perp \hat{z}$, with a finite frequency ω,

$$E^3 \hat{\mathbf{x}} = E^1 \hat{\mathbf{x}} = E^{\mathrm{ex}} \hat{\mathbf{x}}, \quad H^3 \hat{\mathbf{y}} = H^1 \hat{\mathbf{y}} = H^{\mathrm{ex}} \hat{\mathbf{y}}, \tag{33}$$

we have in region 2, because of Eq. (23),

$$\mathbf{E}^M = E^{\mathrm{ex}} \hat{\mathbf{x}}, \quad \mathbf{H}_x^M = H^{\mathrm{ex}} \hat{\mathbf{y}}. \tag{34}$$

All fields are uniform. The field D and B are, to linear order in the frequency,

$$\mathbf{D}/\bar{\epsilon} = (1 + i\omega\beta\bar{\epsilon}/\epsilon_0) E^{\mathrm{ex}} \hat{\mathbf{x}}, \tag{35}$$

$$\mathbf{B}/\bar{\mu} = (1 + i\omega\alpha\bar{\mu}/\mu_0) H^{\mathrm{ex}} \hat{\mathbf{y}}. \tag{36}$$

They can be measured in a small air gap (with its long dimension along $\hat{z}$) inside the ferrofluid — because the normal component of D and B are continuous, cf. Eq. (24).

B. Hydrodynamic prediction

The hydrodynamic Maxwell equations contain solutions (termed the sq modes [4]) that the linear response theory does not possess. For the given geometry, they assume the form given in Eqs. (19) and (20).

It is useful to first understand a technical point: Although this solution remains valid for finite frequencies, as long as $L\omega/c \ll 1$, we may not treat E_0 and H_0 as strictly uniform when calculating E^D and H^D via Eq. (11) and Eqs. (19) and (20). This is because E_0 and H_0 contain terms $\sim \exp i\omega(x/c \pm t)$, and the spatial derivative in Eq. (11) leads to terms $\sim \omega$, ie terms $\sim \dot{D}, \dot{B}$, which are relevant in the present context. To include these terms, we rewrite $\mathbf{H}^D = -(\alpha/\mu_0)\nabla \times \mathbf{E} = (\alpha/\mu_0)(\dot{\mathbf{B}} + \nabla \times \mathbf{E}^D)$, or

$$\mathbf{H}^D = (\alpha/\mu_0)\dot{\mathbf{B}} + \lambda^2 \nabla \times \nabla \times \mathbf{H}. \tag{37}$$

Similarly,

$$\mathbf{E}^D = (\beta/\epsilon_0)\dot{\mathbf{D}} + \lambda^2 \nabla \times \nabla \times \mathbf{E}. \tag{38}$$

Now, when calculating the terms $\sim \lambda^2$, we may indeed take E_0 and H_0 as spatially constant, as the terms $\sim \omega^2$ are of higher than the considered order. The results are

$$\mathbf{E}^D = \frac{\beta}{\epsilon_0}\dot{D}\hat{\mathbf{x}} - \frac{1}{\sqrt{\epsilon_0}}[\mathcal{E}_+ e^{(z-L)/\lambda} + \mathcal{E}_- e^{-z/\lambda}]\hat{\mathbf{x}}, \tag{39}$$

$$\mathbf{H}^D = \frac{\alpha}{\mu_0}\dot{B}\hat{\mathbf{y}} - \sqrt{\frac{\alpha}{\beta\mu_0}}\,[\mathcal{E}_+ e^{(z-L)/\lambda} - \mathcal{E}_- e^{-z/\lambda}]\hat{\mathbf{y}}. \tag{40}$$

Together with Eqs. (19) and (20), the Maxwell fields are given as

$$\mathbf{H}^M = [H_0 + (\alpha/\mu_0)\dot{B}]\hat{\mathbf{y}} = H^{\mathrm{ex}}\hat{\mathbf{y}}, \tag{41}$$

$$\mathbf{E}^M = [E_0 + (\beta/\epsilon_0)\dot{D}]\hat{\mathbf{x}} = E^{\mathrm{ex}}\hat{\mathbf{x}}. \tag{42}$$

We now calculate $\mathcal{E}_\pm$ by inserting Eqs. (39) and (40) into Eq. (26):

$$(\zeta_1 \alpha/\sqrt{\mu_0})\dot{B} - (\beta/\sqrt{\epsilon_0})\dot{D} = A_- \mathcal{E}_+ - A_+ \mathcal{E}_-, \tag{43}$$

$$(\zeta_1 \alpha/\sqrt{\mu_0})\dot{B} + (\beta/\sqrt{\epsilon_0})\dot{D} = A_+ \mathcal{E}_+ - A_- \mathcal{E}_-, \tag{44}$$

where

$$A_+ \equiv (\zeta_1 \sqrt{\alpha/\beta} + 1), \quad A_- \equiv (\zeta_1 \sqrt{\alpha/\beta} - 1)e^{-L/\lambda}. \tag{45}$$

The resolved amplitudes are

$$\mathcal{E}_+ = \frac{\beta\dot{D}/\sqrt{\epsilon_0}}{(A_+ - A_-)} + \frac{\zeta_1 \alpha \dot{B}/\sqrt{\mu_0}}{(A_+ + A_-)}, \tag{46}$$

$$\mathcal{E}_- = \frac{\beta \dot{D}/\sqrt{\epsilon_0}}{(A_+ - A_-)} - \frac{\zeta_1 \alpha \dot{B}/\sqrt{\mu_0}}{(A_+ + A_-)}, \tag{47}$$

Combining the results of Eqs. (41) and (42) for E_0 and H_0 with that of Eqs. (46) and (47) for $\mathcal{E}_\pm$, we finally find the expression for the field [Eqs. (19) and (20)].

If $\mathcal{E}_\pm$ were zero, the fields in region 2 would be

$$\mathbf{E} = E_0 \hat{\mathbf{x}} = [E^{\text{ex}} - (\beta/\epsilon_0)\dot{D}]\hat{\mathbf{x}}, \tag{48}$$

$$\mathbf{H} = H_0 \hat{\mathbf{y}} = [H^{\text{ex}} - (\alpha/\mu_0)\dot{B}]\hat{\mathbf{y}}. \tag{49}$$

(Note the discontinuity $\sim \dot{D}, \dot{B}$, in the fields E and H — though this is not usually apparent, as we only deal with E^M and H^M in the linear response theory. Note also that these two terms were erroneously omitted in Ref. [3].) Taking the respective second term as a small perturbation, cf. Eqs. (14), we may approximate

$$\dot{D} = \overline{\epsilon}\dot{E} \approx \overline{\epsilon}\dot{E}^{\text{ex}}, \tag{50}$$

$$\dot{B} = \overline{\mu}\dot{H} \approx \overline{\mu}\dot{H}^{\text{ex}} \tag{51}$$

to obtain

$$\mathbf{D}/\overline{\epsilon} \equiv \mathbf{E} = (1 + i\omega\beta\overline{\epsilon}/\epsilon_0)E^{\text{ex}}\hat{\mathbf{x}}, \tag{52}$$

$$\mathbf{B}/\overline{\mu} \equiv \mathbf{H} = (1 + i\omega\alpha\overline{\mu}/\mu_0)H^{\text{ex}}\hat{\mathbf{y}}, \tag{53}$$

which agree with the linear response results [Eq. (35) and (36)]. The point is, of course, irrespective of the value of ζ_1, the sq amplitudes $\mathcal{E}_\pm$ are always finite. And the results from the linear response theory are not valid.

If $\lambda \ll L$, we may set $A_- = 0$ and take the field contribution $\sim \mathcal{E}_+$ to be well separated from that $\sim \mathcal{E}_-$. Close to $z = 0$, only the term $\sim \mathcal{E}_-$ contributes,

$$\mathbf{E} = \left[E^{\text{ex}} - \frac{\beta}{\epsilon_0}\dot{D} + \frac{e^{-z/\lambda}}{A_+\sqrt{\epsilon_0}} \left(\frac{\beta}{\sqrt{\epsilon_0}}\dot{D} - \frac{\zeta_1 \alpha}{\sqrt{\mu_0}}\dot{B} \right) \right] \hat{\mathbf{x}}, \tag{54}$$

partly eliminating the discontinuities in the field of the linear response theory: If $\zeta = 0$, the electric field is continuous, and if $\zeta_1 = \infty$, the magnetic field is. So a more gradual change to the bulk values of Eqs. (48) and (49) is achieved as a result of the sq modes.

For $\lambda \gg L$ and $(\zeta_1\sqrt{\alpha/\beta} - 1)L/\lambda \ll 1$, (i.e., assuming that ζ_1 is not large enough to compensate for the smallness of $L/\lambda \approx 1$ cm/30 m$= 3\times10^{-4}$,) the especially simple and time-independent results emerge,

$$\mathbf{E} = E^{\text{ex}}\hat{\mathbf{x}}, \quad \mathbf{H} = H^{\text{ex}}\hat{\mathbf{y}}, \tag{55}$$

valid to $O(L/\lambda)^0$. Finally, it is useful to remind ourselves that an E field normal to the interface does not couple to the sq mode, and will not result in finite amplitudes $\mathcal{E}_\pm$.

VI. SECOND EXPERIMENT, CONDUCTING BOUNDARIES

As we shall see, subject to a plausible assumption, the difference between linear response and hydrodynamic theory is, with conducting walls, much more pronounced.

A. Linear response prediction

We retain the geometry of Sec. V, and again consider a $\hat{z}$ axis with the three regions. Region 2 still contains ferrofluid; regions 1 and 3 now consists of a conducting substance, where $H^D, E^D = 0$ still holds. Applying tangential fields along $\hat{x}$, and $\hat{y}$, of vanishing frequency $\omega \to 0$ (though high enough to ensure that the ferrofluid remains dielectric), we keep

$$E^3\hat{\mathbf{x}} = E^1\hat{\mathbf{x}} = E^{\text{ex}}\hat{\mathbf{x}}, \tag{56}$$

while the magnetic field acquires a constant gradient,

$$\mathbf{H}^1 = [H^L - \sigma E^{\text{ex}} z]\hat{\mathbf{y}}, \tag{57}$$

$$\mathbf{H}^3 = [H^R - \sigma E^{\text{ex}}(z - L)]\hat{\mathbf{y}}. \tag{58}$$

(With $E^{\text{ex}} = \nabla \times \mathbf{H}^{1,3}/\sigma$, the electric field is clearly dependent.) The boundary conditions [Eqs. (23)] render the fields within region 2 constant and the same as outside:

$$E^2 = E^{\text{ex}}, \quad H^2 = H^L = H^R. \tag{59}$$

B. Hydrodynamic prediction

The linear response solution holds in regions 1 and 3; but in region 2, the fields are predicted to behave differently. The boundary conditions [Eqs. (27) and (28)] yield

$$\mathbf{E}_0 = E^{\text{ex}}\hat{\mathbf{x}}, \tag{60}$$

$$H_0 = \tfrac{1}{2}(H^L - H^R). \tag{61}$$

More precisely, the symmetry of Eq. (61) only expresses the fact that essentially the same boundary condition is valid on each sides of region 2. The actual value of $H^L - H^R$ depends on ζ_2, i.e., H^R is not independent of H^L.

The third boundary condition [Eq. (29)] determines the sq amplitudes $\mathcal{E}_\pm$. Applying it twice, for both boundaries, and employing formulas (39)–(42), we first find

$$\mathcal{E} = \mathcal{E}_- = \mathcal{E}_+, \tag{62}$$

leading to an E field symmetric at $z = L/2$, and an associated, antisymmetric H field:

$$E = E_0 + \sqrt{1/\epsilon_0}\,\mathcal{E}(e^{-z/\lambda} + e^{(z-L)/\lambda})\hat{\mathbf{x}}, \tag{63}$$

$$H = H_0 - \sqrt{\alpha/(\beta\mu_0)}\,\mathcal{E}(e^{-z/\lambda} - e^{(z-L)/\lambda})\hat{\mathbf{y}}. \tag{64}$$

Furthermore, the amplitude is given as

$$\mathcal{E} = E_0(\hat{A}_- - \hat{A}_+)\sqrt{\epsilon_0}, \tag{65}$$

where

$$\hat{A}_+ \equiv (\zeta_3\sqrt{\alpha/\beta} + 1), \tag{66}$$

$$\hat{A}_- \equiv (\zeta_3\sqrt{\alpha/\beta} - 1)e^{-L/\lambda},$$

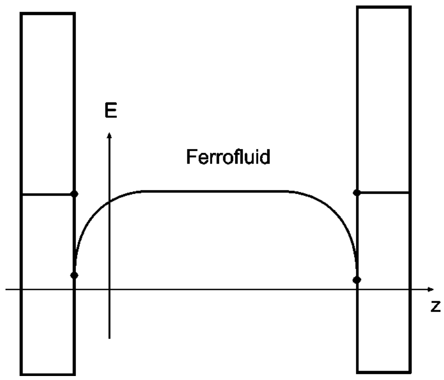

FIG. 1. The electric field for $L \gg \lambda$.

cf. Figs. 1 and 2.

As discussed, the value of the surface Onsager coefficient ζ_3 is unfortunately (as yet) unknown. However, since it has been chosen as a dimensionless quantity, it does not have any cause to stray far from unity — unless there is a hidden symmetry reason forcing it to vanish or diverge consistently, for any CFI. If it is indeed infinite, then Eq. (29) forces H^D to vanish. With Eq. (64), this implies $\mathcal{E} \sim H^D = 0$, and there is no difference from the linear response results for the experimental circumstances under consideration. (This is a crucial difference from the results of Sec. V, where the discrepancy persists irrespective of the value of ζ_1.)

If, on the other hand, ζ_3 remains finite, more precisely if

$$\zeta_3 L/\lambda \ll 1, \tag{67}$$

the above results may be expanded to become

$$\frac{E}{E_{\rm ex}} = \frac{\zeta_3 L}{2\lambda} \sqrt{\frac{\alpha}{\beta}}, \tag{68}$$

$$\frac{H_0 - H}{E_{\rm ex}} = \frac{z - L/2}{\lambda} \sqrt{\frac{\alpha \epsilon_0}{\beta \mu_0}},$$

The E field vanishes in the limit $L/\lambda \to 0$ because the constant term $\sim (L/\lambda)^0$ in $\mathcal{E}_\pm$ exactly cancels E_0 in Eq. (19). With $L = 1$ cm and $\lambda = 30$ m, the quotient L/λ is indeed small in the envisioned experiment, so we may conclude that $E \ll E_{\rm ex}$ (see Figs. 3 and 4).

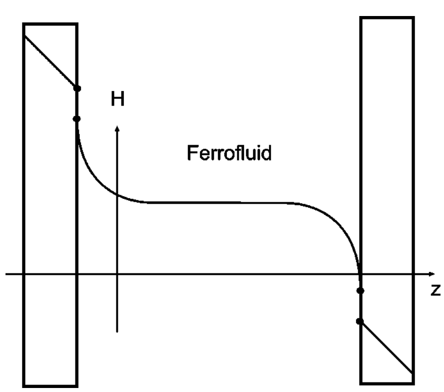

FIG. 2. The magnetic field for $L \gg \lambda$.

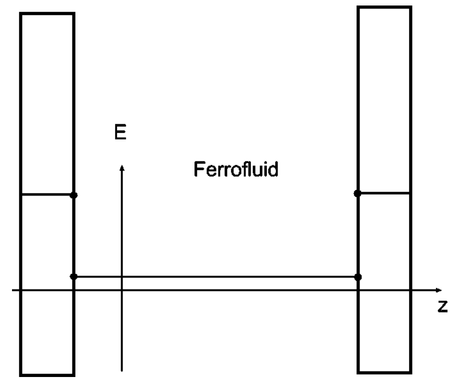

FIG. 3. The electric field for $L \ll \lambda$.

VII. SUMMARY

Studying macroscopic electrodynamics for low frequencies and small fields, such that linear constitutive relations and terms linear in the frequency suffice, discrepancies were found between linear response and the hydrodynamic theory. Careful deliberation shows that the routine neglect, in the linear response theory, of cross constitutive terms and spatial dispersions to be the reason for this. In particular, a stationary, dissipative field configuration, in which both the electric and the magnetic field participate, is shown to be erroneously ruled out by this omission. In ferrofluids, this field configuration should be a dominating effect. Two experiments, with boundaries of different electromagnetic behaviors, are suggested that should verify these results.

APPENDIX

We start with

$$\dot{\mathbf{D}} = c \nabla \times \mathbf{H}^M, \quad \dot{\mathbf{B}} = -c \nabla \times \mathbf{E}^M, \tag{A1}$$

and the relaxation equation for the magnetization,

$$\dot{\mathbf{M}} = -(\mathbf{M} - \mathbf{M}_{\rm eq})/\tau_M, \tag{A2}$$

where

$$\mathbf{M} = \mathbf{B} - \mathbf{H}^M. \tag{A3}$$

Electric dissipation, from the dynamics of a polarization, is neglected,

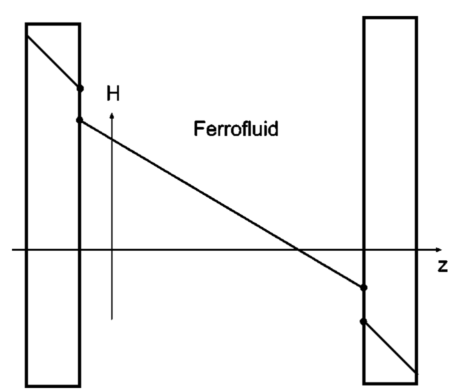

FIG. 4. The magnetic field for $L \ll \lambda$.

$$\mathbf{E}^M = \mathbf{E} = \mathbf{D}. \tag{A4}$$

The equilibrium value of the magnetization is

$$\mathbf{M}_{\rm eq} = \chi_M \mathbf{H} = (\chi_M / \bar{\mu}) \mathbf{B}, \quad \chi_M = \bar{\mu} - 1. \tag{A5}$$

Taking δ to denote the deviation from the respective static value, we rewrite Eq. (A2) as

$$-i\omega\tau_M \delta\mathbf{M} = -\delta\mathbf{M} + (\chi_M/\bar{\mu})\delta\mathbf{B}, \tag{A6}$$

or

$$\delta\mathbf{M} \approx (1 + i\omega\tau_M)(\chi_M/\bar{\mu})\delta\mathbf{B}. \tag{A7}$$

Inserting this into

$$\dot{\mathbf{D}} = c\nabla \times (\delta\mathbf{B} - \delta\mathbf{M}), \tag{A8}$$

we have

$$\dot{\mathbf{D}} = c\nabla \times \delta\mathbf{H} - (\tau_M \chi_M/\bar{\mu})\dot{\mathbf{B}}, \tag{A9}$$

or

$$\dot{\mathbf{D}} = c\nabla \times \delta\mathbf{H} - (\tau_M \chi_M/\bar{\mu}) c\nabla \times \mathbf{E}. \tag{A10}$$

Identifying the second term as $-\alpha c \nabla \times \mathbf{E}$, we obtain

$$\alpha = \tau_M \chi_M/\bar{\mu} = \tau_M(\bar{\mu} - 1)/\bar{\mu}. \tag{A11}$$

Finally, we switch from the Heaviside-Lorentz units to the MKSA units by multiplying both the denominator and numerator with μ_0, while noticing that $\bar{\mu}$ in the MKSA is $\bar{\mu}\mu_0$,

$$\alpha = \tau_M \chi_M/\bar{\mu} = \tau_M(\bar{\mu} - \mu_0)/\bar{\mu}. \tag{A12}$$

The formula for β may be obtained analogously.

[1] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, Oxford, 1984), especially Sec. 10, p. 77.

[2] R. E. Rosensweig, *Ferrohydrodynamics* (Cambridge University Press, Cambridge, 1985); M. I. Schliomis, Usp. Fiz. Nauk **112**, 427 (1974) [Sov. Phys. Usp. **17**, 153 (1974)].

[3] Mario Liu, Phys. Rev. Lett. **80**, 2937 (1998).

[4] Mario Liu, Phys. Rev. Lett. **70**, 3580 (1993); **74**, 1884 (1995); **74**, 4535 (1995); **77**, 1043 (1996).

[5] B. Castaing and P. Nozières, J. Phys. (Paris) **41**, 701 (1980); Y. Sun, P. Wölfe, and S. Yip, Phys. Rev. Lett. **63**, 1613 (1989); M. Grabinski and M. Liu, *ibid.* **65**, 2666 (1990); P. Panzer and M. Liu, *ibid.* **69**, 3658 (1992); Peter Kostädt and Mario Liu, *ibid.* **71**, 3513 (1993); Europhys. Lett. **28**, 409 (1994); Jie Qin and Mario Liu, *ibid.* **24**, 47 (1993).