Traceless multipole moment densities and transformations in macroscopic electromagnetism

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We consider whether use of traceless multipole moment densities in macroscopic electromagnetism can yield physically acceptable results. For harmonic plane wave fields it is shown that a traceless electric quadrupole density yields linear constitutive relations for which the dynamical material constants (permittivity and magnetoelectric coefficients) and response fields are unphysical. We further show that, within multipole theory, these constitutive relations cannot be transformed into physically acceptable relations. Specifically, the transformed response field **D** is unphysical for all orders beyond the electric dipole. This contrasts with use of primitive (traced) moment densities, for which unphysical constitutive relations have been successfully transformed up to electric octopole-magnetic quadrupole order, thereby providing also the leading contribution to the ac permeability.

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I. INTRODUCTION

There have been numerous discussions of the manner in which the macroscopic Maxwell equations

$$\nabla \cdot \mathbf{D} = \rho_f, \tag{1}$$

$$\nabla \times \mathbf{H} = \mathbf{J}_f + \dot{\mathbf{D}} \tag{2}$$

can be deduced from the microscopic equations of electrodynamics. Essentially, one performs suitable averages of series expansions for the microscopic charge and current densities (or for the dynamic scalar and vector potentials). This introduces the macroscopic multipole moment densities. Detailed accounts have been given by Russakoff [1], Robinson [2], Scaife [3], and Jackson [4], where references to earlier work can be found. For consistency in these multipole expansions, it is necessary to pair successive electric and magnetic multipole orders beyond the electric dipole in a particular way, namely, electric quadrupole with magnetic dipole, electric octopole with magnetic quadrupole, and so on [5,6].

To electric quadrupole-magnetic dipole order the response fields are given in terms of the macroscopic electric and magnetic fields and moment densities by [1–6]

$$D_i = \epsilon_0 E_i + P_i - \frac{1}{2}\nabla_j Q_{ij} \tag{3}$$

$$H_i = \mu_0^{-1} B_i - M_i, \tag{4}$$

where P_i and M_i are the electric and magnetic dipole moment densities, and Q_{ij} is the electric quadrupole moment density. These densities are obtained from (weighted) spatial averages of the relevant multipole moments of "molecules" in the medium [1–4]. Thus Q_{ij} involves an average of the electric quadrupole moment

$$q_{ij} = \sum q r_i r_j, \tag{5}$$

where **r** is the position vector of charge q relative to an origin inside a molecule, and the summation is over all charges in a molecule. We refer to (5) as a primitive (or traced) moment to distinguish it from the corresponding traceless moment (33). Traceless multipole moments (and hence traceless densities) can be constructed for all orders beyond the electric dipole.

It is well known that in the electrostatics of a charge distribution in vacuum the traces of successive primitive electric multipole moments (q_{kk}, q_{ikk}, etc.) do not contribute to the potential or the field; this is a consequence of Laplace's equation [7,8]. There has been discussion in the literature on whether this property applies also in macroscopic electrodynamics; that is, whether one can replace (3), for example, by [9]

$$D_i = \epsilon_0 E_i + P_i - \frac{1}{3}\nabla_j \Theta_{ij}, \tag{6}$$

where

$$\Theta_{ij} = \frac{1}{2}(3Q_{ij} - Q_{kk}\delta_{ij}) \tag{7}$$

is the traceless electric quadrupole moment density. It has been shown that with **D** given by Eq. (6), the Maxwell equations (1) and (2) lose their translational invariance [5], and certain macroscopic observables in transmission phenomena [10] and electrostatics [11] are unphysical because they depend on the choice of origin for the laboratory system of coordinates. There are exceptions, where the trace Q_{kk} does not contribute, namely, in the theory of optical activity for light propagating along the symmetry axis of a fluid of aligned molecules [9,10], and the theory of field-gradient-induced birefringence [12,13]. However, it is clear that in general it is not valid to neglect Q_{kk}. Even in the simple example of multipole radiation, the trace of the electric quadrupole moment makes a contribution in media where the radiated fields have longitudinal components [14].

An alternative way of bringing the macroscopic theory into conformity with electrostatics has been presented [4]. Instead of omitting the trace Q_{kk} in Eq. (3), one incorporates

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it into the free source densities in such a way that the Maxwell equations (1) and (2) are unaltered. Thus, for **D** given by Eq. (6), one makes the replacements

$$\rho_f \to \rho_f + \frac{1}{6}\nabla^2 Q_{kk}, \tag{8}$$

$$J_{fi} \to J_{fi} - \frac{1}{6}\nabla_i \dot{Q}_{kk} \tag{9}$$

in Eqs. (1) and (2). Higher-order multipole contributions to **D** and **H** can be redefined in a similar way, and they result in complicated modifications to ρ_f and $\mathbf{J}_f$. The purpose of this paper is to discuss the physical implications of using the traceless formalism of Eqs. (6)–(9), and to compare these with the use of Eq. (3), which is based on the primitive density Q_{ij}.

Note that transmission phenomena do not allow one to distinguish between the primitive and traceless formalisms because the wave equation [6,15] obtained from Eq. (2) is the same whether one uses Eq. (3) or Eqs. (6)–(9). Instead, we consider linear constitutive relations which express the response fields **D** and **H** in terms of the electric and magnetic fields **E** and **B**. In addition to providing macroscopic observables (material constants), these relations are required for field matching at a crystal surface [6,16], and for our purposes they provide a more stringent test of the theory than do transmission effects.

The microscopic starting points of both formulations involve expectation values of appropriate multipole moment operators. It is well known that these expectation values are the same for the two formulations, at least to electric quadrupole-magnetic dipole order (Sec. V). It is therefore of interest to enquire what differences, if any, arise as one proceeds to the macroscopic theory and its transformations.

In Sec. II we give a brief account of recent work on the multipole theory of constitutive relations based on primitive moment densities. Although these constitutive relations obtained directly from multipole theory are unphysical, they can be transformed into physically acceptable relations. In Sec. III we obtain constitutive relations based on the traceless quadrupole moment density (7), and show that these are also unphysical. In Sec. IV it is shown that, within multipole theory, the latter constitutive relations cannot be successfully transformed.

II. CONSTITUTIVE RELATIONS FROM PRIMITIVE QUADRUPOLE MOMENT DENSITIES

We consider the electromagnetic fields of harmonic plane waves

$$\mathbf{E} = \mathbf{E}_0 e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)}, \tag{10}$$

and similarly for **B**. The multipole moment densities in Eqs. (3) and (4) that are induced in a magnetic medium by the fields are, to electric quadrupole-magnetic dipole order [15,17],

$$P_i = \alpha_{ij}E_j + \omega^{-1}\alpha'_{ij}\dot{E}_j + \frac{1}{2}a_{ijk}\nabla_k E_j + \frac{1}{2}\omega^{-1}a'_{ijk}\nabla_k \dot{E}_j + G_{ij}B_j + \omega^{-1}G'_{ij}\dot{B}_j, \tag{11}$$

$$Q_{ij} = a_{kij}E_k - \omega^{-1}a'_{kij}\dot{E}_k, \tag{12}$$

$$M_i = G_{ji}E_j - \omega^{-1}G'_{ji}\dot{E}_j. \tag{13}$$

Here α_{ij} (a_{ijk} and G'_{ij}) are polarizability densities of electric dipole (electric quadrupole-magnetic dipole) order for nonmagnetics, and α'_{ij} a'_{ijk}, and G_{ij} are their counterparts for magnetics.

Quantum-mechanical expressions for these polarizability densities are given in Ref. [16]. For our purposes the relevant features are their intrinsic symmetries [9,16,17]

$$\alpha_{ij} = \alpha_{ji}, \quad \alpha'_{ij} = -\alpha'_{ji}, \tag{14}$$

$$a_{ijk} = a_{ikj}, \quad a'_{ijk} = a'_{ikj} \tag{15}$$

and origin dependences [9,16]

$$\Delta\alpha_{ij} = 0, \quad \Delta\alpha'_{ij} = 0, \tag{16}$$

$$\Delta a_{ijk} = -R_k\alpha_{ij} - R_j\alpha_{ik}, \quad \Delta a'_{ijk} = -R_k\alpha'_{ij} - R_j\alpha'_{ik}, \tag{17}$$

$$\Delta G_{ij} = -\frac{1}{2}\omega\epsilon_{jkl}R_k\alpha'_{il}, \quad \Delta G'_{ij} = \frac{1}{2}\omega\epsilon_{jkl}R_k\alpha_{il}. \tag{18}$$

Here ϵ_{ijk} is the Levi-Cività tensor, and Δ denotes a change due to an arbitrary shift $\mathbf{R}=(R_x,R_y,R_z)$ of the coordinate origin of the laboratory system. Equations (10)–(18) apply to both nondissipativeand dissipative media.

For a homogeneous medium, Eqs. (3), (4), and (10)–(13) yield linear constitutive relations of the form

$$D_i = A_{ij}E_j + T_{ij}B_j, \tag{19}$$

$$H_i = U_{ij}E_j + X_{ij}B_j. \tag{20}$$

The complex material constants represent the permittivity (A_{ij}), magnetoelectric coefficients (T_{ij} and U_{ij}), and inverse permeability (X_{ij}), and they are given by

$$A^M_{ij} = \epsilon_0\delta_{ij} + \alpha_{ij} - i\alpha'_{ij} + \frac{1}{2}(ia_{ijk} - ia_{jik} + a'_{ijk} + a'_{jik})k_k, \tag{21}$$

$$T^M_{ij} = G_{ij} - iG'_{ij}, \tag{22}$$

$$U^M_{ij} = -G_{ji} - iG'_{ji}, \tag{23}$$

$$X^M_{ij} = \mu_0^{-1}\delta_{ij}. \tag{24}$$

The superscript M denotes material constants obtained directly from multipole theory, to distinguish them from the transformed material constants referred to below. We see that terms of electric quadrupole-magnetic dipole order provide the first-order terms in the magnetoelectric coefficients, and the second-order contribution to the permittivity, while they do not contribute to the permeability. (To obtain the first-order contribution to the permeability, one must work to electric octopole-magnetic quadrupole order [18].)

There are three conditions that must be satisfied by the material constants in Eqs. (19) and (20).

(1) Symmetries. For nondissipative media the following symmetries apply [19–22]:

$$A_{ij} = A^*_{ji}, \quad T_{ij} = -U^*_{ji}, \quad X_{ij} = X^*_{ji}. \tag{25}$$

Using Eqs. (14) and (15), and because the wave vector **k** and the polarizability densities are real for nondissipative media, we see that Eqs. (21)–(24) possess these symmetries.

(ii) Translational invariance. Polarizability densities beyond electric dipole order are origin-dependent quantities [see Eqs. (16)–(18) and Ref. [16]]. Thus when multipole theory constructs macroscopic observables out of polarizability densities, it should combine these densities in such a way that the overall expressions are origin independent. This it does successfully in the wave equation for transmission phenomena, but not for the material constants (21)–(23) [6]. For example, according to Eqs. (18) and (22),

$$\Delta T^M_{ij} = -\frac{i}{2}\omega\epsilon_{jkl}R_k(\alpha_{il} - i\alpha'_{il}), \tag{26}$$

which, for ac effects, is not zero in general.

(iii) The Post constraint. This constraint [20,23] requires equality of the traces of the magnetoelectric tensors

$$T_{ii} = U_{ii}. \tag{27}$$

It is violated by Eqs. (22) and (23).

Thus the material constants (21)–(23), obtained directly from multipole theory, fail to satisfy two of the above conditions. This failure of the theory occurs also at electric octopole-magnetic quadrupole order [6].

A way round this difficulty is provided by a transformation theory based on the nonuniqueness of **D** and **H** in Maxwell's equations [6,24]. This theory transforms the unphysical material constants (21)–(23) into [6,24]

$$A_{ij} = \epsilon_0\delta_{ij} + \alpha_{ij} - i\alpha'_{ij} + \frac{1}{3}(a'_{ijk} + a'_{jki} + a'_{kij})k_k, \tag{28}$$

$$T_{ij} = -i\left(G'_{ij} - \frac{1}{2}\omega\epsilon_{jkl}a_{kli}\right) + G_{ij} - \frac{1}{3}G_{ll}\delta_{ij} - \frac{1}{6}\omega\epsilon_{jkl}a'_{kli}, \tag{29}$$

$$U_{ij} = -i\left(G'_{ji} - \frac{1}{2}\omega\epsilon_{ikl}a_{klj}\right) - G_{ji} + \frac{1}{3}G_{ll}\delta_{ij} + \frac{1}{6}\omega\epsilon_{ikl}a'_{klj}, \tag{30}$$

while it leaves Eq. (24) unchanged. These transformed expressions are unique, origin independent, and satisfy the symmetries (25) and the constraint (27). They are therefore physically acceptable expressions for the material constants of a magnetic medium to electric quadrupole-magnetic dipole order.

III. CONSTITUTIVE RELATIONS FROM TRACELESS QUADRUPOLE MOMENT DENSITIES

We now come to the main part of this paper, namely, what are the properties of constitutive relations based on traceless electric quadrupole moment densities, and, if necessary, can they be transformed?

We first consider how Eqs. (11)–(13) are modified in the traceless formalism. The only polarizability densities in Eqs. (11)–(13) that involve the electric quadrupole moment operator are the quadrupole polarizability densities a_{ijk} and a'_{ijk} [16]. Thus Eq. (13) is unchanged, while Eqs. (11) and (12) become [25]

$$P_i = \alpha_{ij}E_j + \omega^{-1}\alpha'_{ij}\dot{E}_j + \frac{1}{3}A_{ijk}\nabla_k E_j + \frac{1}{3}\omega^{-1}A'_{ijk}\nabla_k\dot{E}_j + G_{ij}B_j + \omega^{-1}G'_{ij}\dot{B}_j, \tag{31}$$

$$\Theta_{ij} = A_{kij}E_k - \omega^{-1}A'_{kij}\dot{E}_j. \tag{32}$$

We refer to A_{ijk} and A'_{ijk} as traceless quadrupole polarizability densities to distinguish them from the corresponding primitive densities a_{ijk} and a'_{ijk}. The relationships between these tensors can be written down by noting that the primitive densities involve matrix elements of the primitive quadrupole moment q_{ij} in Eq. (5) [16], whereas the traceless densities involve matrix elements of the traceless moment

$$\frac{1}{2}(3q_{ij} - q_{kk}\delta_{ij}). \tag{33}$$

Replacing q_{ij} by (33) in Eqs. (B8) and (B9) of Ref. [16], we have

$$A_{ijk} = \frac{3}{2}a_{ijk} - \frac{1}{2}a_{ill}\delta_{jk}, \tag{34}$$

$$A'_{ijk} = \frac{3}{2}a'_{ijk} - \frac{1}{2}a'_{ill}\delta_{jk}. \tag{35}$$

These tensors have the following properties. The traces A_{ijj} and A'_{ijj} are zero, and therefore Θ_{ij} in Eq. (32) is traceless. Also, from Eqs. (34), (35), (15), and (17) we have the intrinsic symmetries

$$A_{ijk} = A_{ikj}, \quad A'_{ijk} = A'_{ikj}, \tag{36}$$

and the origin dependences

$$\Delta A_{ijk} = -\frac{3}{2}R_k\alpha_{ij} - \frac{3}{2}R_j\alpha_{ik} + R_l\alpha_{il}\delta_{jk}, \tag{37}$$

$$\Delta A'_{ijk} = -\frac{3}{2}R_k\alpha'_{ij} - \frac{3}{2}R_j\alpha'_{ik} + R_l\alpha'_{il}\delta_{jk}. \tag{38}$$

Constitutive relations follow from Eqs. (4), (6), (10), (13), (31), and (32). They have the form (19) and (20) with

$$A_{ij}^M = \epsilon_0\delta_{ij} + \alpha_{ij} - i\alpha'_{ij} + \frac{1}{3}(iA_{ijk} - iA_{jik} + A'_{ijk} + A'_{jik})k_k \tag{39}$$

and T_{ij}^M, U_{ij}^M, and X_{ij}^M given by Eqs. (22)–(24). The permittivity tensor (39) satisfies the symmetry in (25), but it is not origin independent according to (37) and (38). Thus the material constants obtained directly from multipole theory using the traceless formalism have the same unphysical properties as those of the primitive formalism in Sec. II: three of them are origin dependent and the Post constraint is violated. We therefore consider whether these material constants can be transformed into physically acceptable quantities.

IV. TRANSFORMATIONS

The response fields in Eqs. (19) and (20) are represented by complex harmonic plane waves like (10). For such fields, the Maxwell equations (1) and (2) are invariant under the transformations [24]

$$A_{ij} \to A_{ij}^M - \frac{1}{\omega}\epsilon_{ikl}k_k U_{lj}^G + \frac{1}{\omega}\epsilon_{jkl}k_k Z_{il}^F, \tag{40}$$

$$T_{ij} \to T_{ij}^M - \frac{1}{\omega}\epsilon_{ikl}k_k X_{lj}^G + Z_{ij}^F, \tag{41}$$

$$U_{ij} \to U_{ij}^M + U_{ij}^G + \frac{1}{\omega}\epsilon_{jkl}k_k Y_{il}^F, \tag{42}$$

$$X_{ij} \to X_{ij}^M + X_{ij}^G + Y_{ij}^F \tag{43}$$

of the direct multipole material constants. The superscript F denotes a Faraday transformation, while G denotes a gauge transformation [24].

The tensors U_{ij}^G, X_{ij}^G, Y_{ij}^F, and Z_{ij}^F should satisfy the following criteria.

(i) For a given multipole order, they are constructed from the polarizability densities of that order, and δ_{ij} and ϵ_{ijk}; see (44). They are linear in the polarizability densities and, at electric quadrupole–magnetic dipole order, are independent of the wave vector $\mathbf{k}$.

(ii) The transformations should be consistent with the requirements of space inversion and time reversal, and this means that U_{ij}^G and Z_{ij}^F are time-odd axial tensors, while X_{ij}^G and Y_{ij}^F are time-even polar tensors.

(iii) The transformations should preserve the symmetries (25) and they should impose origin independence on the material constants.

(iv) A Faraday transformation cannot change the response fields **D** and **H**.

We now apply this transformation theory to the traceless formalism at electric quadupole–magnetic dipole order. Consider first a nonmagnetic medium. Then the nonzero polarizability densities are G'_{ij} and A_{ijk}. The available building blocks for constructing U_{ij}^G, X_{ij}^G, Y_{ij}^F, and Z_{ij}^F are the second-rank tensors

$$G_{ij}, \quad G'_{ji}, \quad G'_{ll}\delta_{ij}, \quad \epsilon_{ikl}A_{klj}, \quad \epsilon_{jkl}A_{kli}, \quad \epsilon_{ijk}A_{llk}. \tag{44}$$

[Here we have taken account of the property $A_{kll}=0$, and the symmetry in Eq. (36) for A_{ijk}.] The tensors in (44) are all axial and therefore so are linear combinations constructed from them. Because X_{ij}^G and Y_{ij}^F are necessarily polar (see above), it follows that

$$X_{ij}^G = Y_{ij}^F = 0. \tag{45}$$

Using (44) we write

$$U_{ij}^G = \beta_1 G'_{ij} + \beta_2 G'_{ji} + \beta_3 G'_{ll}\delta_{ij} + \beta_4\epsilon_{ikl}A_{klj} + \beta_5\epsilon_{jkl}A_{kli} + \beta_6\epsilon_{ijk}A_{llk}, \tag{46}$$

$$Z_{ij}^F = \gamma_1 G'_{ij} + \gamma_2 G'_{ji} + \gamma_3 G'_{ll}\delta_{ij} + \gamma_4\epsilon_{ikl}A_{klj} + \gamma_5\epsilon_{jkl}A_{kli} + \gamma_6\epsilon_{ijk}A_{llk}, \tag{47}$$

where the 12 coefficients β_i and γ_i are to be determined. The tensors on the right-hand sides of Eqs. (46) and (47) are all time even, while U_{ij}^G and Z_{ij}^F are necessarily time odd (see above). Therefore the β_i and γ_i are either imaginary or zero.

Next, we make a convenient manipulation. From Eqs. (40), (46), and (47), and Faraday's law for a plane wave, we see that $A_{ij}E_j$ contains the terms

$$-\frac{1}{\omega}(\epsilon_{ikl}\beta_3\delta_{lj} - \epsilon_{jkl}\gamma_3\delta_{il})G'_{mm}k_kE_j = -(\beta_3+\gamma_3)G'_{mm}\delta_{ij}B_j. \tag{48}$$

Incorporating Eq. (48) in $T_{ij}B_j$ in Eq. (19), and using the direct multipole results of the previous section, we obtain from Eqs. (40)–(43) and (45)–(47)

$$\begin{aligned}A_{ij} = {}& \epsilon_0\delta_{ij} + \alpha_{ij} + \frac{1}{3}ik_k(A_{ijk} - A_{jik}) - \frac{1}{\omega}\epsilon_{ikl}k_k(\beta_1 G'_{lj} \\ & + \beta_2 G'_{jl} + \beta_4\epsilon_{lmn}A_{mnj} + \beta_5\epsilon_{jmn}A_{mnl} + \beta_6\epsilon_{ljm}A_{nnm}) \\ & + \frac{1}{\omega}\epsilon_{jkl}k_k(\gamma_1 G'_{il} + \gamma_2 G'_{li} + \gamma_4\epsilon_{imn}A_{mnl} + \gamma_5\epsilon_{lmn}A_{mni} \\ & + \gamma_6\epsilon_{ilm}A_{nnm}),\end{aligned} \tag{49}$$

$$T_{ij} = -iG'_{ij} + \gamma_1 G'_{ij} + \gamma_2 G'_{ji} + \gamma_4\epsilon_{ikl}A_{klj} + \gamma_5\epsilon_{jkl}A_{kli} + \gamma_6\epsilon_{ijk}A_{llk} - \beta_3 G'_{ll}\delta_{ij}, \tag{50}$$

$$U_{ij} = -iG'_{ji} + \beta_1 G'_{ij} + \beta_2 G'_{ji} + \beta_3 G'_{ll}\delta_{ij} + \beta_4\epsilon_{ikl}A_{klj} + \beta_5\epsilon_{jkl}A_{kli} + \beta_6\epsilon_{ijk}A_{llk}, \tag{51}$$

$$X_{ij} = \mu_0^{-1}\delta_{ij}. \tag{52}$$

Now impose origin independence on T_{ij} and U_{ij}. For a shift of origin $\mathbf{R}=(0,0,R_z)$ one finds from Eqs. (51), (37), and (18)

$$\Delta U_{xy} = R_z\left[\alpha_{xx}\left(\frac{1}{2}\omega\beta_1 + \frac{3}{2}\beta_5 - \frac{3}{2}\beta_6\right) + \alpha_{yy}\left(\frac{1}{2}i\omega - \frac{1}{2}\omega\beta_2 - \frac{3}{2}\beta_4 - \frac{3}{2}\beta_6\right) + \alpha_{zz}(-\beta_4 + \beta_5 - 2\beta_6)\right]. \tag{53}$$

Since $\alpha_{xx}, \alpha_{yy}, \alpha_{zz}$ are independent components of α_{ij}, the invariance $\Delta U_{xy}=0$ requires

$$\omega\beta_1 + 3\beta_5 - 3\beta_6 = 0, \tag{54}$$

$$i\omega - \omega\beta_2 - 3\beta_4 - 3\beta_6 = 0, \tag{55}$$

$$-\beta_4 + \beta_5 - 2\beta_6 = 0. \tag{56}$$

Invariance of the other components of U_{ij} does not provide additional equations for the β_i. From Eqs. (54)–(56) we have

$$\beta_2 = i + \beta_1, \quad \beta_5 = -\frac{2}{3}\omega\beta_1 - \beta_4, \quad \beta_6 = -\frac{1}{3}\omega\beta_1 - \beta_4. \tag{57}$$

With Eq. (57) in Eq. (51), a short calculation shows that the terms in β_4 cancel, and

$$U_{ij} = \beta_1\left(G'_{ij} + G'_{ji} - \frac{2}{3}\omega\epsilon_{ikl}A_{klj} + \frac{1}{3}\omega\epsilon_{ijk}A_{llk}\right) + \beta_3 G'_{ll}\delta_{ij}. \tag{58}$$

A similar calculation based on Eq. (50) and $\Delta T_{xy}=0$ yields

$$\gamma_2 = -i + \gamma_1, \quad \gamma_5 = -\frac{2}{3}\omega\gamma_2 - \gamma_4, \quad \gamma_6 = -\frac{1}{3}\omega\gamma_2 - \gamma_4. \tag{59}$$

Then Eq. (50) becomes

$$T_{ij} = \gamma_2\left(G'_{ij} + G'_{ji} - \frac{2}{3}\omega\epsilon_{jkl}A_{kli} - \frac{1}{3}\omega\epsilon_{ijk}A_{llk}\right) - \beta_3 G'_{ll}\delta_{ij}. \tag{60}$$

The magnetoelectric coefficients (58) and (60) are origin independent for all values of the coefficients β_1, β_3, and γ_2. Note that it was only necessary to impose origin independence on the *xy* components of T_{ij} and U_{ij}; invariance of all other components followed.

We now impose the symmetry $T_{ij}=-U^*_{ji}$ of Eq. (25). Because the β_i and γ_i are imaginary or zero in Eqs. (58) and (60), this means

$$\beta_1 = \gamma_2, \quad \beta_3 = 0.$$

Thus U_{ij} in Eq. (58) is proportional to γ_2 and the response field **H** in Eq. (20) depends on γ_2, which is a coefficient in a Faraday transformation; see Eq. (47). This violates the restriction mentioned above, that a Faraday transformation cannot change a response field [24]. Thus $\gamma_2=0$, and the magnetoelectric coefficients of a nonmagnetic medium are transformed to zero,

$$U_{ij} = T_{ij} = 0. \tag{61}$$

The transformed permittivity (40), or (49), is now fixed because from Eqs. (41), (42), (45), and (61):

$$U^G_{ij} = -U^M_{ij} = iG'_{ji} \tag{62}$$

$$Z^F_{ij} = -T^M_{ij} = iG'_{ij}. \tag{63}$$

From Eqs. (39), (40), (62), and (63) we have

$$A_{ij} = \epsilon_0\delta_{ij} + \alpha_{ij} + i\left[\frac{1}{3}(A_{ijk} - A_{jik}) - \frac{1}{\omega}\epsilon_{ikl}G'_{jl} + \frac{1}{\omega}\epsilon_{jkl}G'_{il}\right]k_k. \tag{64}$$

This is the transformed permittivity for a nonmagnetic medium according to the traceless theory. It is not origin independent because from Eqs. (16), (18), and (37),

$$\Delta A_{ij} = \frac{1}{3}ik_kR_l(\alpha_{il}\delta_{jk} - \alpha_{jl}\delta_{ik}), \tag{65}$$

which is not zero in general.

If the medium is magnetic then there are additional contributions to the material constants: that in α'_{ij} [see Eq. (21)] and the transformed ones associated with the polarizability densities G_{ij} and A'_{ijk}. These contributions must be added to Eqs. (61), (64), and (52). The details are similar to the above and we simply quote the final results:

$$A_{ij} = \epsilon_0\delta_{ij} + \alpha_{ij} - i\alpha'_{ij} + \left[\frac{i}{3}(A_{ijk} - A_{jik}) - \frac{i}{\omega}\epsilon_{ikl}G'_{jl} + \frac{i}{\omega}\epsilon_{jkl}G'_{il} + \frac{2}{9}(A'_{ijk} + A'_{jik} + A'_{kij}) - \frac{2}{45}(2A'_{llk}\delta_{ij} - A'_{lli}\delta_{jk} - A'_{llj}\delta_{ki})\right]k_k, \tag{66}$$

$$T_{ij} = G_{ij} - \frac{1}{3}G_{ll}\delta_{ij} + \frac{2}{45}\omega\epsilon_{ijk}A'_{llk} - \frac{1}{9}\omega\epsilon_{jkl}A'_{kli}, \tag{67}$$

$$U_{ij} = -G_{ji} + \frac{1}{3}G_{ll}\delta_{ij} + \frac{2}{45}\omega\epsilon_{ijk}A'_{llk} + \frac{1}{9}\omega\epsilon_{ikl}A'_{klj}, \tag{68}$$

$$X_{ij} = \mu_0^{-1}\delta_{ij}. \tag{69}$$

These are the total transformed material constants, to electric quadrupole–magnetic dipole order, of a magnetic medium in the traceless theory. They satisfy the symmetries (25), and T_{ij} and U_{ij} are traceless and origin independent. However, the permittivity A_{ij} is origin dependent because, according to Eqs. (16), (18), (37), and (38),

$$\Delta A_{ij} = \frac{1}{3}ik_kR_l[(\alpha_{il} - i\alpha'_{il})\delta_{jk} - (\alpha_{jl} + i\alpha'_{jl})\delta_{ik}]. \tag{70}$$

Such origin dependence for a macroscopic observable is, of course, unphysical. For example, if $\mathbf{D}_0$ denotes the amplitude of a harmonic response field **D** then, from Eqs. (19) and (67),

$$\Delta D_{0i} = (\Delta A_{ij}) E_{0j}. \tag{71}$$

According to Eqs. (70) and (71), the amplitude of the transformed response field **D** is origin dependent in the traceless formalism.

V. DISCUSSION

(i) The two main results obtained in this paper are, first, that constitutive relations obtained directly from multipole theory in the traceless formalism are unphysical (Sec. III), and, second, that they cannot be transformed into physically acceptable relations (Sec. IV). This contrasts with the primitive formalism where unphysical constitutive relations can be successfully transformed (Sec. II).

(ii) Because the polarizability densities G_{ij}, G'_{ij}, A_{ijk}, and A'_{ijk} of electric quadrupole–magnetic dipole order do not occur again at higher multipole orders, the response field **D** associated with the transformed permittivity (66) of the traceless theory will be unphysical to all multipole orders.

(iii) Comparison of Eqs. (28)–(30) and (66)–(68) shows that the transformed material constants A_{ij}, T_{ij}, and U_{ij} in the primitive and traceless versions of the theory differ markedly in their dependence on the polarizabilities G'_{ij}, A_{ijk}, and A'_{ijk}. For example, in Eqs. (66)–(68) the density G'_{ij} has been transformed out of the magnetoelectric coefficients and into the permittivity.

(iv) It is interesting to consider where the differences in these two versions of the theory arise. The starting points of both versions are the quantum-mechanical relations on which Eqs. (11)–(13), (31), and (32) are based; that is, on relations between expectation values of multipole moment operators (electric dipole and quadrupole, magnetic dipole) of a charge distribution in vacuum and the microscopic electromagnetic fields [6,17]. These relations are independent of whether one uses a primitive or a traceless quadrupole moment operator. This is due to the transverse nature of the microscopic fields, as is clear from the treatment of Barron [26]. Thus the microscopic starting points of the primitive and traceless theories are the same. (This is not true at electric octopole order because the trace of the primitive electric octopole moment operator does contribute to the expectation values [27].)

(v) A difference in the two approaches first appears in the permittivity tensors obtained directly from macroscopic multipole theory. The difference of the permittivities (39) and (21) is, when expressed in terms of a_{ijk} and a'_{ijk} using Eqs. (34) and (35), equal to

$$-\frac{1}{6} i(a_{ill} - i a'_{ill}) \delta_{jk} k_k + \frac{1}{6} i(a_{jll} + i a'_{jll}) \delta_{ik} k_k. \tag{72}$$

The first term in (72) makes a contribution to **D** in Eq. (19) if the macroscopic electric field **E** is not transverse. The second term in Eq. (72) is due to the different electrodynamic expressions (3) and (6) for **D**.

(vi) The major differences between the results of the two theories occur after the unphysical material constants of each have been transformed, and then come from a seemingly small cause: namely, the different translational properties of A_{ijk} and a_{ijk} on the one hand, and A'_{ijk} and a'_{ijk} on the other; see Eqs. (17), (37), and (38). Specifically, it is the terms $R_l \alpha_{il} \delta_{jk}$ and $R_l \alpha'_{il} \delta_{jk}$ in Eqs. (37) and (38) that cause the transformation theory to produce such different results.

(vii) At electric quadrupole–magnetic dipole order there is no contribution to the permeability of the medium. This is true of both the direct and the transformed theories, in either the primitive or the traceless version (Secs. II–IV). The ac permeability is a property of electric octopole–magnetic quadrupole order and its expression in direct multipole theory is unphysical [6].

(viii) The transformation theory has been successfully applied to electric octopole–magnetic quadrupole order in the primitive formalism, thereby providing a physically acceptable expression for the lowest-order contribution to the ac permeability [18]. These calculations are considerably more difficult than those of electric quadrupole-magnetic dipole order, and they pose a challenge for multipole theory. Their successful completion encourages confidence in the primitive formulation of the theory.

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