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Key Points:
- Definitive boundary conditions are derived for electric quadrupolar media
- A required additional boundary condition is derived from the Maxwell differential equations
- Analytical and numerical solutions are found for an electric quadrupolar slab

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Additional boundary condition for electric quadrupolar continua derived from Maxwell's differential equations

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Abstract Electric quadrupolar continua satisfying a physically reasonable constitutive relation supports both an evanescent and a propagating eigenmode. Thus, three interface boundary conditions, two plus an "additional boundary condition" (ABC), are required to obtain a unique solution to a plane wave incident from free space upon an electric quadrupolar half-space. By generalizing the constitutive relation to hold within the transition layer between the free space and the quadrupolar continuum, we derive these three boundary conditions directly from Maxwell's differential equations. The three boundary conditions are used to determine the unique solution to the boundary value problem of an electric quadrupolar slab. Numerical computations show that for long wavelengths, two previous boundary conditions, derived under the assumption that the electric quadrupolarization contains negligible effective delta functions in the transition layer, produce an accurate solution by neglecting the evanescent eigenmode, that is, by assuming it decays within the transition layer. It appears that the general method used to derive the electric quadrupolar ABC can be applied to obtain the boundary conditions for any other realizable constitutive relation in a Maxwellian multipole continuum.

1. Introduction

Although the electric quadrupolarization in most natural materials is negligible below optical frequencies compared to the dipolarization, at optical and higher frequencies, and for metamaterial arrays made with artificial molecules (inclusions), the electric quadrupolarization can contribute significantly to the fields [*Raab and Lange*, 2005; *Cho et al.*, 2008; *Silveirinha*, 2014]. Large electric quadrupolar resonances of electrically small metallic spheres at plasmonic frequencies may be used to design metamaterials with a strong electric quadrupolar response [*Alù and Engheta*, 2014]. The loss tangents in the permittivity of metals at plasmonic frequencies are higher than 0.1 [*Naik et al.*, 2013] and, for fully metallic spheres, this amount of loss greatly reduces the resonance in the electric quadrupolar response [*Alù and Engheta*, 2007]. However, for metallic shells covering ordinary dielectric spheres, the losses can be kept low enough to retain significant electric quadrupolar plasmonic resonances [*Oldenburg et al.*, 1999]. And, of course, if the fully plasmonic spheres were enhanced with gainy material, this gainy material could compensate for the reduction in the size of the resonances produced by the passive loss tangents. Also, for spatially dispersive metamaterials designed for applications that can tolerate resonances at larger electrical sizes, ordinary dielectric constants with positive real parts can be used to obtain electric quadrupolar resonances. Thus, it seems appropriate for more than academic reasons to characterize the electromagnetic properties of electric quadrupolar media.

In the publications [*Yaghjian et al.*, 2014], criteria was given for the average fields of a general periodic array of inclusions separated in free space to obey Maxwell's macroscopic continuum equations. Specifically, the lattice spacing has to be a small enough fraction of both the free-space and modal wavelengths ($k_0 d$ and $|\beta d| \ll 1$) that the average fields and sources over the unit cell are approximately equal to the corresponding coefficients of the fundamental Floquet modes. In order to consider an example other than dipolar continua, we concentrated on electric quadrupolar continua and derived boundary conditions at the interface of two such continua, in particular, the boundary conditions at the interface between free space and an electric quadrupolar continuum. Two independent boundary conditions on the tangential electric and magnetic fields were revealed. Across a free-space/quadrupolar interface, these two boundary conditions can be written as

$$\mathbf{E}_s^{(2)} - \mathbf{E}_s^{(1)} = \frac{1}{2\epsilon_0}\nabla_s\left(\hat{\mathbf{n}}\cdot\overline{\mathbf{Q}}^{(2)}\cdot\hat{\mathbf{n}}\right) \tag{1}$$

$$\mathbf{B}_s^{(2)} - \mathbf{B}_s^{(1)} = -\frac{i\omega\mu_0}{2}\hat{\mathbf{n}} \times \left(\hat{\mathbf{n}} \cdot \overline{\mathbf{Q}}^{(2)}\right), \tag{2}$$

where $\mathbf{E}$ and $\mathbf{B}$ are the macroscopic time-harmonic ($e^{-i\omega t}$, $\omega > 0$) electric and magnetic fields (with ϵ_0 and μ_0 the permittivity and permeability of free space), $\overline{\mathbf{Q}}$ is the macroscopic electric quadrupolarization density in the continuum ($\overline{\mathbf{Q}}$ in free space is zero), and n and s refer to the normal and tangential directions with respect to the interface with $\hat{\mathbf{n}}$ the unit normal pointing from the free space (designated by superscript 1) into the electric quadrupolar continuum (designated by superscript 2).

The boundary conditions across the interface on the normal components of macroscopic $\mathbf{B}$ and $\mathbf{D}$ fields, namely,

$$B_n^{(2)} - B_n^{(1)} = 0 \tag{3}$$

$$D_n^{(2)} - D_n^{(1)} = \frac{1}{2}\nabla_s \cdot \left(\hat{\mathbf{n}} \cdot \overline{\mathbf{Q}}_s^{(2)}\right) \tag{4}$$

are not independent of the tangential field boundary conditions in (1) and (2) because (3) and (4) are implied by Maxwell's equations combined with (1) and (2).

Maxwell's homogeneous differential equations for the macroscopic fields in the electric quadrupolar continuum can be written as

$$\nabla \times \mathbf{E} - i\omega\mathbf{B} = 0 \tag{5}$$

$$\frac{1}{\mu_0}\nabla \times \mathbf{B} + i\omega\epsilon_0\mathbf{E} - \frac{1}{2}i\omega\nabla \cdot \overline{\mathbf{Q}} = 0 \tag{6}$$

with $\mathbf{D}$ and $\mathbf{H}$ defined by the constitutive relations

$$\mathbf{D} = \epsilon_0\mathbf{E} - \frac{1}{2}\nabla \cdot \overline{\mathbf{Q}} \tag{7}$$

$$\mathbf{H} = \frac{1}{\mu_0}\mathbf{B}, \tag{8}$$

where in (7) the quantity $-\nabla \cdot \overline{\mathbf{Q}}/2$ can be considered equal to an effective electric polarization $\mathbf{P}$. The boundary conditions in (1) and (2) were derived in *Yaghjian et al.* [2014] from an integration of Maxwell's equations in (5) and (6) using two main assumptions (i) that $\nabla \cdot \overline{\mathbf{Q}}$ can have a delta function across the interface because of the jump in $\overline{\mathbf{Q}}$ that occurs in general across the interface and (ii) that there are no delta functions in $\overline{\mathbf{Q}}$ within the transition layer in which the electromagnetic properties change from those of free space to those of the electric quadrupolar continuum. These boundary conditions in (1) and (2) were found by *Raab and Lange* [2005] (with a minor error corrected in *Raab and Lange* [2013]) using assumption (i) but without stating assumption (ii).

In reference *Yaghjian* [2014], the validity of these assumptions was investigated by first deriving the fields in a source-free, isotropic electric quadrupolar continuum satisfying the following physically reasonable electric quadrupolar constitutive relation for a random distribution of spherically symmetric electric quadrupolar particles in source-free external fields

$$\overline{\mathbf{Q}} = \alpha_Q\epsilon_0\left[\frac{1}{2}(\nabla\mathbf{E} + \mathbf{E}\nabla) - \frac{1}{3}(\nabla \cdot \mathbf{E})\bar{\mathbf{I}}\right], \tag{9}$$

where the constant α_Q is a macroscopic electric quadrupolarizability density (real valued in a lossless continuum) and $\mathbf{E}\nabla$ denotes the transpose of the $\nabla\mathbf{E}$ dyadic. The $\overline{\mathbf{Q}}$ in (9) is a symmetric dyadic (a requirement of electric quadrupolarizability) with zero trace that can be derived from the electric quadrupole moment of an electrically small dielectric-sphere inclusion in a source-free local electric field *Yaghjian et al.* [2015]. (We note that in equation (25c) of reference *Yaghjian* [2014], the D variables in (4) were mistakenly written as $\epsilon_0 E$.)

We found in *Yaghjian* [2014] that because the electric fields can be discontinuous across the interface, the electric quadrupolarization, given in the continuum by the constitutive relation in (9), can have delta functions in the transition layer, and thus, the boundary conditions in (1) and (2) may not hold for this electric quadrupolar continuum. Moreover, we found that two modes can exist in this electric quadrupolar continuum for transverse magnetic (TM) plane wave incidence. One of the modes is propagating and the other is evanescent. Therefore, even if the boundary conditions in (1) and (2) did hold, we would need an additional boundary condition to solve for the coefficients of these two modes as well as for the coefficient of the reflected plane wave for a plane wave incident from free space on a semi-infinite electric quadrupolar continuum.

Despite these obstacles to finding a solution to this half-space problem, we were able to show in *Yaghjian* [2014] that if the frequency is low enough (that is, if $k_0 d$ and $|\beta d|$ are sufficiently small, where k_0 $(= \omega\sqrt{\mu_0\epsilon_0})$ and β are the free-space and modal propagation constants, respectively, and d is the lattice spacing of the inclusions comprising the continuum), then the evanescent mode could be considered as confined to the transition layer and, moreover, the boundary conditions in (1) and (2) will still apply.

The motivational purpose of the present paper is to confirm that the boundary conditions in (1) and (2) are indeed valid at the lower frequencies for the electric quadrupolar continuum satisfying the constitutive relation in (9), by finding an exact solution to the boundary value problem of a plane wave incident from free space upon an electric quadrupolar slab. To solve this boundary value problem, we have to include the evanescent waves in the electric quadrupolar continuum and, thus, we also have to derive an additional boundary condition (ABC) that can be applied to the interfaces of the slab and free space. To obtain this particular electric-quadrupolar ABC, we devise and demonstrate a general method that appears capable of determining the boundary conditions (including ABCs) for any other realizable constitutive relation in a Maxwellian multipole continuum.

2. Derivation of Three Independent Boundary Conditions

The additional boundary condition will be determined below by combining the Maxwell differential equations in (5) and (6) with the constitutive relation in (9). This leads to the possibility of the electric quadrupolarization having delta functions and normal derivatives of delta functions as well as a unit-step function within the transition layer between the free space and the electric quadrupolar continuum. It turns out that some of these delta functions are squared in the equations and thus violate finite energy conditions unless the tangential electric field $\mathbf{E}_s$ is continuous across the transition layer and the normal component $Q_{nn} = \hat{\mathbf{n}}\cdot\overline{\mathbf{Q}}\cdot\hat{\mathbf{n}}$ of the electric quadrupolarization is zero on either side of the transition layer. The boundary condition on the tangential magnetic field $\mathbf{B}_s$ also changes from the one in (2) for transverse magnetic plane wave incidence but only by a term that becomes negligible as $k_0 d$ becomes small (that is, for a sufficiently accurate continuum). In summary, then, we will find that the constitutive relation in (9) requires through Maxwell's equations the three following independent boundary conditions at the interface between free space and an electric quadrupolar continuum

$$\mathbf{E}_s^{(2)} - \mathbf{E}_s^{(1)} = 0 \tag{10}$$

$$\mathbf{B}_s^{(2)} - \mathbf{B}_s^{(1)} = -\frac{i\omega\mu_0}{2}\hat{\mathbf{n}}\times\left(\hat{\mathbf{n}}\cdot\overline{\mathbf{Q}}^{(2)}\right) \tag{11}$$

$$\hat{\mathbf{n}}\cdot\overline{\mathbf{Q}}^{(2)}\cdot\hat{\mathbf{n}} = 0. \tag{12}$$

These boundary conditions are compatible with the general boundary conditions first derived by *Silveirinha* [2014] for electric quadrupolar metamaterials using "transverse averaging" and assuming no delta functions in the fields and electric quadrupolarization in the transition layer. The next subsection provides a detailed derivation of the three boundary conditions in (10)–(12) directly from Maxwell's differential equations in (5) and (6) and the constitutive relation in (9), without assuming that there are no delta functions in the fields or electric quadrupolarization in the transition layer.

2.1. Details of the Derivation

To derive the three boundary conditions in (10)–(12) from Maxwell's differential equations, we consider a planar interface between free space and the electric quadrupolar continuum. Let the interface be coincident with the *xy* plane of a rectangular coordinate system whose positive *z* axis is normal to the interface and points into the quadrupolar half-space, as shown in Figure 1. As mentioned above, there will be an interface transition layer across which the electromagnetic properties of the medium change from those of free space

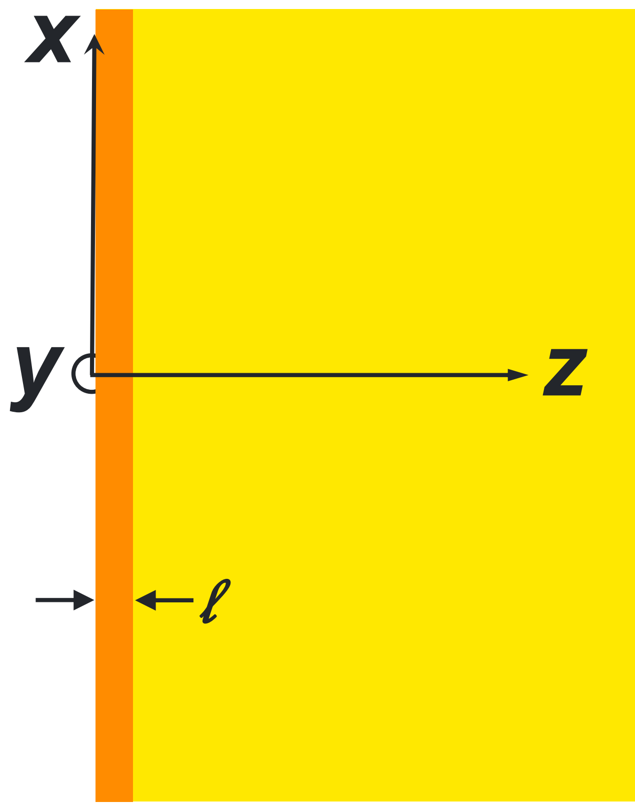

Figure 1. Geometry of interface between free space and electric quadrupolar continuum with transition layer of thickness ℓ.

to those of the electric quadrupolar continuum. We let this transition layer of thickness ℓ lie between $z = 0$ and $z = \ell$, also shown in Figure 1. (Under the conditions that dipolar arrays behave as a continua, namely, $k_0 d \ll 1$ and $|\beta d| \ll 1$, analytical and numerical results indicate that the thickness ℓ of the transition layer is on the order of the average separation distance d of the dipoles [*Simovski and Tretyakov*, 2007; *Sher and Kuester*, 2009]. The same result presumably holds for electric quadrupoles.)

In the free space to the left of the transition layer ($z < 0$), the value of the electric quadrupolarizability density in (9) is zero, and to the right of the transition layer ($z > \ell$) its value is equal to α_Q. Since the quadrupolarizability of each of the electric quadrupoles is finite, a macroscopic electric quadrupolarizability density α_0 that holds throughout all space is finite in the transition layer and can be expressed as

$$\alpha_0 = \alpha_Q u(z), \tag{13}$$

where $u(z)$ is a continuous "unit-step" function that varies from 0 (at $z = 0$) to 1 (at $z = \ell$) across the transition layer. For an ideal continuum, $\ell \to 0$ and $u(z)$ approaches the usual unit-step distribution function. With α_0 in (13) replacing α_Q in (9), the electric quadrupolarization density $\overline{\mathbf{Q}}_0$ that holds throughout all space, including the transition layer, is given by

$$\overline{\mathbf{Q}}_0 = \alpha_Q \epsilon_0 u(z) \left[\frac{1}{2}(\nabla \mathbf{E} + \mathbf{E}\nabla) - \frac{1}{3}(\nabla \cdot \mathbf{E})\bar{\mathbf{I}} \right]. \tag{14}$$

We note that $\overline{\mathbf{Q}}_0$ equals $\overline{\mathbf{Q}}$ in (9) for $z > \ell$. With the electric quadrupolarization density in (14), Maxwell's homogeneous differential macroscopic equations that hold throughout all space are given by

$$\nabla \times \mathbf{E} - i\omega \mathbf{B} = 0 \tag{15}$$

$$\frac{1}{\mu_0} \nabla \times \mathbf{B} + i\omega\epsilon_0 \mathbf{E} - \frac{1}{2} i\omega \nabla \cdot \overline{\mathbf{Q}}_0 = 0. \tag{16}$$

Since the fields incident from sources in the free-space region ($z < 0$) can be expanded in a spectrum of transverse electric (TE: **E** perpendicular to the plane of incidence) and transverse magnetic (TM: **B** perpendicular to the plane of incidence) plane waves, we can consider these two incident plane waves separately. If we choose the plane of incidence perpendicular to the y direction, then the TE and TM plane waves have their electric and magnetic fields in the y direction, respectively. For a TE incident plane wave, (16) implies that only $\overline{\mathbf{Q}}_0 \cdot \hat{\mathbf{y}}$ is nonzero and that, in particular, $Q_{0zz} = 0$ everywhere. Thus, the boundary condition in (1) predicts a continuous tangential electric field E_y. Moreover, the Maxwell equations in (5) and (6) can be used to show that there is just one mode, a propagating mode, that can exist in the electric quadrupolar continuum. Consequently, for TE incident plane waves, the two boundary conditions in (1) and (2) are sufficient for a unique solution and lead to fields consistent with the equations in (15) and (16). Moreover, these TE solutions satisfy the boundary conditions in (10)–(12) because Q_{0zz} is zero everywhere and thus the boundary conditions in (1) and (2) become identical to those in (10) and (11). Our remaining task, then, is to derive the boundary conditions in (10)–(12) for incident TM plane waves.

To do this, first substitute **B** from (15) into (16) to get a single vector wave equation for **E**, namely,

$$\nabla \times \nabla \times \mathbf{E} - k_0^2 \mathbf{E} + \frac{1}{2}\omega^2 \mu_0 \nabla \cdot \overline{\mathbf{Q}}_0 = 0 \tag{17}$$

with $\nabla \cdot \overline{\mathbf{Q}}_0$ obtained from (14) as

$$\nabla \cdot \overline{\mathbf{Q}}_0 = \alpha_Q \epsilon_0 \left\{ u(z) \nabla \cdot \left[\frac{1}{2}(\nabla \mathbf{E} + \mathbf{E}\nabla) - \frac{1}{3}(\nabla \cdot \mathbf{E})\bar{\mathbf{I}} \right] + \delta(z)\hat{\mathbf{z}} \cdot \left[\frac{1}{2}(\nabla \mathbf{E} + \mathbf{E}\nabla) - \frac{1}{3}(\nabla \cdot \mathbf{E})\bar{\mathbf{I}} \right] \right\}, \tag{18}$$

where $\delta(z) = \mathrm{d}u(z)/\mathrm{d}z$. For the TM solution with the magnetic field in the y direction, but no variation in the y direction, and $e^{ik_{0x}x}$ variation in the x direction, the E_y field is zero everywhere, and the evaluation of the x and z components of (17) and (18) gives the following two equations

$$\left(k_0^2 + \frac{\partial^2}{\partial z^2}\right) E_x - ik_{0x}\frac{\partial E_z}{\partial z} - \frac{1}{2}\omega^2 \mu_0 \left(\nabla \cdot \overline{\mathbf{Q}}_0\right)_x = 0 \tag{19}$$

$$\left(k_0^2 - k_{0x}^2\right) E_z - ik_{0x}\frac{\partial E_x}{\partial z} - \frac{1}{2}\omega^2 \mu_0 \left(\nabla \cdot \overline{\mathbf{Q}}_0\right)_z = 0, \tag{20}$$

where the x and y components of $\nabla \cdot \overline{\mathbf{Q}}_0$ are

$$\left(\nabla \cdot \overline{\mathbf{Q}}_0\right)_x = \alpha_Q \epsilon_0 \left[\frac{1}{2}\frac{\partial}{\partial z}\left(u\frac{\partial E_x}{\partial z}\right) + ik_{0x}\left(\frac{1}{6}u\frac{\partial E_z}{\partial z} + \frac{1}{2}\delta E_z\right) - \frac{2}{3}k_{0x}^2 u E_x \right] \tag{21}$$

$$\left(\nabla \cdot \overline{\mathbf{Q}}_0\right)_z = \alpha_Q \epsilon_0 \left[\frac{2}{3}\frac{\partial}{\partial z}\left(u\frac{\partial E_z}{\partial z}\right) + ik_{0x}\left(\frac{1}{6}u\frac{\partial E_x}{\partial z} - \frac{1}{3}\delta E_x\right) - \frac{1}{2}k_{0x}^2 u E_z \right] \tag{22}$$

and we have made use of the relation $\partial/\partial x = ik_{0x}$.

One cannot assume a priori that the electric field cannot contain delta functions in the transition layer. In fact, a planar multipole expansion for the continuously differentiable electric and magnetic fields outside the source plane at $z = 0$ shows that within the source plane the electric and magnetic fields can be expressed as a continuous function plus a sum of unit-step distribution functions, that is, the unit-step function and all its derivatives. Consequently, the components of the electric field everywhere, including the transition layer, can be expressed as

$$E_x(z) = E_x^r(z) + E_x^u u(z) + E_x^\delta \delta(z) + E_x^{\delta'} \delta'(z) + \ldots \tag{23}$$

$$E_z(z) = E_z^r(z) + E_z^u u(z) + E_z^\delta \delta(z) + E_z^{\delta'} \delta'(z) + \ldots \tag{24}$$

in which the $e^{ik_{0x}x}$ variation in the x direction is suppressed. The $E_x^r(z)$ and $E_z^r(z)$ are continuous ramp functions that have constant values in the transition layer, and $\partial/\partial z$ derivatives from the left and right of the transition layer that can have different values. The constant values of $E_x^r(z)$ and $E_z^r(z)$ in the transition layer are not necessarily zero. The primes on the delta functions in (23) and (24) denote differentiation with respect to z and, of course, $\delta(z) = u'(z)$. Unit-step/delta distribution theory legitimizes the use of delta functions and shows that there is no loss in generality in choosing the defining step function in (23) and (24) equal to the $u(z)$ in (13) as the thickness of the transition layer approaches zero; that is, $\ell \to 0$.

If E_x and E_z are substituted from (23) and (24) into (19) and (20), the $\left(\nabla \cdot \overline{\mathbf{Q}}_0\right)_x$ and $\left(\nabla \cdot \overline{\mathbf{Q}}_0\right)_z$ quadrupolar source terms will contain squares of δ functions and derivatives of delta functions. These squared sources would produce infinite fields everywhere, and thus, their coefficients must be zero. To find the coefficients that must be zero, first integrate $\left(\nabla \cdot \overline{\mathbf{Q}}_0\right)_x$ in (21) with respect to z over the transition layer to get

$$\frac{1}{2}\frac{\partial E_x^{(2)}}{\partial z} + \frac{1}{3} ik_{0x} \int_0^\ell \delta E_z \mathrm{d}z + \frac{1}{6} ik_{0x} \frac{\partial E_z^{(2)}}{\partial z} - \frac{2}{3} k_{0x}^2 \int_0^\ell u E_x \mathrm{d}z, \tag{25}$$

where we have used integration by parts in the second term on the right-hand side of (21). Multiplying (24) by $\delta(z)$ and using integration by parts, we find

$$\int_0^\ell \delta E_z \mathrm{d}z = \text{finite value} + \int_0^\ell \left[E_z^\delta \delta^2(z) - E_z^{\delta''} \delta'^2(z) + E_z^{\delta''''} \delta''^2(z) - \dots\right] \mathrm{d}z. \tag{26}$$

Similarly,

$$\int_0^\ell u E_x \mathrm{d}z = \text{finite value} - \int_0^\ell \left[E_x^{\delta'} \delta^2(z) - E_x^{\delta'''} \delta'^2(z) + E_x^{\delta'''''} \delta''^2(z) - \dots\right] \mathrm{d}z. \tag{27}$$

With (26) and (27) inserted into (25), each of the squared singular terms, δ^2, δ'^2, δ''^2, $\delta'''^2, \dots$, produce higher-order infinite fields everywhere and, thus, the coefficients of these terms must be zero. This gives the infinite set of equations

$$\begin{aligned} iE_z^\delta + 2k_{0x} E_x^{\delta'} &= 0 \\ iE_z^{\delta''} + 2k_{0x} E_x^{\delta'''} &= 0 \\ &\vdots \end{aligned} \tag{28}$$

Integrating $\left(\nabla \cdot \overline{\mathbf{Q}}_0\right)_z$ in (22) with respect to z over the transition layer, we obtain a similar infinite set of linear equations

$$\begin{aligned} iE_x^\delta - k_{0x} E_z^{\delta'} &= 0 \\ iE_x^{\delta''} - k_{0x} E_z^{\delta'''} &= 0 \\ &\vdots \end{aligned} \tag{29}$$

All the planar multipole moments of $\left(\nabla \cdot \overline{\mathbf{Q}}_0\right)_x$ and $\left(\nabla \cdot \overline{\mathbf{Q}}_0\right)_z$ must give finite fields outside the source region, and thus, in particular, the first moment obtained by integrating the product of z and these two electric quadrupolar source components over the transition layer must not have squared singular terms, δ^2, δ'^2, δ''^2, $\delta'''^2, \dots$. Carrying through the algebra for $\int_0^\ell z \left(\nabla \cdot \overline{\mathbf{Q}}_0\right)_x \mathrm{d}z$, using integration by parts, leads to the infinite set of equations

$$\begin{aligned} \frac{1}{2}E_x^\delta - \frac{1}{3}k_{0x}^2 E_x^{\delta''} + 0ik_{0x} E_z^{\delta'} &= 0 \\ \frac{1}{2}E_x^{\delta''} - \frac{4}{3}k_{0x}^2 E_x^{\delta''''} - \frac{1}{3}ik_{0x} E_z^{\delta'''} &= 0 \\ &\vdots \end{aligned} \tag{30}$$

Similarly, $\int_0^\ell z \left(\nabla \cdot \overline{\mathbf{Q}}_0\right)_z \mathrm{d}z$ leads to

$$\begin{aligned} \frac{2}{3}E_z^\delta - \frac{1}{4}k_{0x}^2 E_z^{\delta''} + \frac{5}{12}ik_{0x} E_x^{\delta'} &= 0 \\ \frac{2}{3}E_z^{\delta''} - k_{0x}^2 E_z^{\delta''''} - \frac{11}{12}ik_{0x} E_x^{\delta'''} &= 0 \\ &\vdots \end{aligned} \tag{31}$$

Inserting $E_z^{\delta'}, E_z^{\delta'''}, E_z^{\delta'''''}, \ldots$, from (29) into (30), then assuming there is an N such that $E_x^{\delta(\text{nth}')} = 0$ for $n > N$, we find that

$$E_x^{\delta}, E_x^{\delta''}, E_x^{\delta''''}, \ldots, = 0 \tag{32}$$

which implies from (30) that

$$E_z^{\delta'}, E_z^{\delta'''}, E_z^{\delta'''''}, \ldots, = 0. \tag{33}$$

Similarly, inserting $E_x^{\delta'}, E_x^{\delta'''}, E_x^{\delta'''''}, \ldots$, from (28) into (31), then assuming there is an N such that $E_z^{\delta(\text{nth}')} = 0$ for $n > N$, we find that

$$E_z^{\delta}, E_z^{\delta''}, E_z^{\delta''''}, \ldots, = 0 \tag{34}$$

which implies from (28) that

$$E_x^{\delta'}, E_x^{\delta'''}, E_x^{\delta'''''}, \ldots, = 0. \tag{35}$$

With (32)–(35) implying that

$$E_x^{\delta}, E_x^{\delta'}, E_x^{\delta''}, E_x^{\delta'''}, E_x^{\delta''''}, \ldots, = 0 \tag{36}$$

and

$$E_z^{\delta}, E_z^{\delta'}, E_z^{\delta''}, E_z^{\delta'''}, E_z^{\delta''''}, \ldots, = 0 \tag{37}$$

we see from (21) and (23) and (24) that

$$\begin{aligned}\left(\nabla\cdot\overline{\mathbf{Q}}_0\right)_x = \alpha_Q\epsilon_0\left\{\frac{1}{2}\frac{\partial}{\partial z}\left[u\left(\frac{\partial E_x^r}{\partial z}+\delta E_x^u\right)\right]+ik_{0x}\left[\frac{1}{6}u\left(\frac{\partial E_z^r}{\partial z}+\delta E_z^u\right)\right.\right.\\ \left.\left.+\frac{1}{2}\delta\left(E_z^r+uE_z^u\right)\right]-\frac{2}{3}k_{0x}^2u\left(E_x^r+uE_x^u\right)\right\}.\end{aligned} \tag{38}$$

If (38) is multiplied by z and integrated over the transition layer, one obtains with the aid of integration by parts

$$\int_0^{\ell}\left(\nabla\cdot\overline{\mathbf{Q}}_0\right)_x \mathrm{d}z = -\frac{1}{4}\alpha_Q\epsilon_0E_x^u. \tag{39}$$

Therefore, multiplying (19) by z and integrating over the transition layer gives in view of (36) and (37)

$$-\left(E_x^{(2)}-E_x^{(1)}\right)+\frac{1}{8}k_0^2\alpha_QE_x^u = \left(\frac{1}{8}k_0^2\alpha_Q-1\right)E_x^u = 0 \tag{40}$$

since $E_x^{(2)}-E_x^{(1)} = E_x^u$. Equation (40) implies that $E_x^u = 0$ because $k_0^2\alpha_Q$ does not generally have a value equal to 8. (For a highly accurate continuum, $k_0^2\alpha_Q \ll 1$ and, moreover, it would be impossible for $k_0^2\alpha_Q$ to realistically be equal to exactly 8 because there is always at least an infinitesimally small loss that would preclude a perfectly real value.) That is, we have $E_x^{(2)} - E_x^{(1)} = 0$ or

$$\mathbf{E}_s^{(2)} - \mathbf{E}_s^{(1)} = 0. \tag{41}$$

In like manner, integrating (20) over the transition layer with $\left(\nabla\cdot\overline{\mathbf{Q}}_0\right)_z$ substituted from (22), then using (36) and (37) and (41), and integration by parts gives

$$\frac{2}{3}\frac{\partial E_z^{(2)}}{\partial z} - \frac{1}{3}ik_{0x}E_x^{(2)} = 0. \tag{42}$$

However, $Q_{zz}^{(2)} = \alpha_Q\epsilon_0(2\partial E_z^{(2)}/\partial z - ik_{0x}E_x^{(2)})/3$, so that (42) implies that $Q_{zz}^{(2)} = 0$ or

$$\hat{\mathbf{n}}\cdot\overline{\mathbf{Q}}^{(2)}\cdot\hat{\mathbf{n}} = 0. \tag{43}$$

We note that this boundary condition of $Q_{zz}^{(2)} = 0$ does not imply continuous Q_{0zz} throughout the transition layer. In fact, with E_z having a unit-step function $u(z)E_z^u$ across the transition layer, (14) implies that Q_{0xx}, Q_{0yy}, and Q_{0zz} contain delta functions within the transition layer because each of these diagonal elements contain a term proportional to $\partial E_z/\partial z$, and thus a term proportional to $\delta(z)E_z^u$.

Lastly, we determine the boundary condition on B_y across the transition layer. To do this, first take the x component of (16) to get

$$\frac{\partial B_y}{\partial z} = i\omega\mu_0\epsilon_0 E_x - \frac{1}{2}i\omega\mu_0\left(\nabla\cdot\overline{\mathbf{Q}}_0\right)_x. \tag{44}$$

Integrating (44) over the transition layer, we find with the help of (38) and (41) that

$$B_y^{(2)} - B_y^{(1)} = -\frac{1}{2}i\omega\mu_0\epsilon_0\alpha_Q\left[\frac{1}{2}\frac{\partial E_x^{(2)}}{\partial z} + \frac{1}{2}ik_{0x}E_z^{(2)} - \frac{1}{6}ik_{0x}\left(E_z^{(2)} - E_z^{(1)}\right)\right]. \tag{45}$$

An expression for $B_y^{(2)} - B_y^{(1)}$ also follows from evaluating Maxwell's equation in (15) on either side of the transition layer, namely,

$$i\omega\left(B_y^{(2)} - B_y^{(1)}\right) = \frac{\partial E_x^{(2)}}{\partial z} - \frac{\partial E_x^{(1)}}{\partial z} - ik_{0x}\left(E_z^{(2)} - E_z^{(1)}\right). \tag{46}$$

Combining (45) and (46), one obtains

$$\frac{\partial E_x^{(2)}}{\partial z} - \frac{\partial E_x^{(1)}}{\partial z} - ik_{0x}\left(1 - k_0^2\alpha_Q/12\right)\left(E_z^{(2)} - E_z^{(1)}\right) = \frac{1}{4}k_0^2\alpha_Q\left(\frac{\partial E_x^{(2)}}{\partial z} + ik_{0x}E_z^{(2)}\right). \tag{47}$$

For a highly accurate continuum, $k_0^2\alpha_Q \ll 1$ and, thus, the $k_0^2\alpha_Q/12$ term in (47) can be neglected. Also, $Q_{xz}^{(2)} = \alpha_Q\epsilon_0(\partial E_x^{(2)}/\partial z + ik_{0x}E_z^{(2)})/2$, so that (47) yields

$$\frac{\partial E_x^{(2)}}{\partial z} - \frac{\partial E_x^{(1)}}{\partial z} - ik_{0x}\left(E_z^{(2)} - E_z^{(1)}\right) = \frac{1}{2}\omega^2\mu_0 Q_{xz}^{(2)} \tag{48}$$

and (46) becomes simply

$$\left(B_y^{(2)} - B_y^{(1)}\right) = -\frac{i\omega\mu_0}{2}Q_{xz}^{(2)} \tag{49}$$

or, in vector-dyadic notation,

$$\mathbf{B}_s^{(2)} - \mathbf{B}_s^{(1)} = -\frac{i\omega\mu_0}{2}\hat{\mathbf{n}}\times\left(\hat{\mathbf{n}}\cdot\overline{\mathbf{Q}}^{(2)}\right). \tag{50}$$

In summary, the three boundary conditions in (10)–(12) at the interface between free space and an electric quadrupolar continuum satisfying the constitutive relation in (9) (with its generalization in (14) to include the transition layer) have been derived directly from Maxwell's differential equations in (15) and (16).

3. Plane Wave Incident on an Electric Quadrupolar Slab

The three boundary conditions in (10)–(12) can be used to solve the problem of a transverse magnetic (TM) plane wave incident from free space upon an electric quadrupolar slab of thickness z_0 satisfying the constitutive relation in (9). There is a reflected wave in free space, a propagating and evanescent wave traveling in both directions in the slab, and a transmitted wave leaving the trailing interface of the slab [*Yaghjian*, 2014]. The boundary conditions in (10)–(12) are applied at both the leading and trailing interfaces of the slab. It is verified that the sum of the magnitudes squared of the reflection and transmission coefficients equals unity to within computer accuracy. It was shown in *Silveirinha* [2014] that the boundary conditions in (10)–(12) ensure the continuity of the normal component of the Poynting's vector at the interfaces, and hence this conservation of energy.

Second, the same scattering problem is solved assuming that the evanescent modes are negligible and applying the two boundary conditions in (1) and (2). Again, it is verified that the sum of the magnitudes squared of the reflection and transmission coefficients equals unity to within computer accuracy.

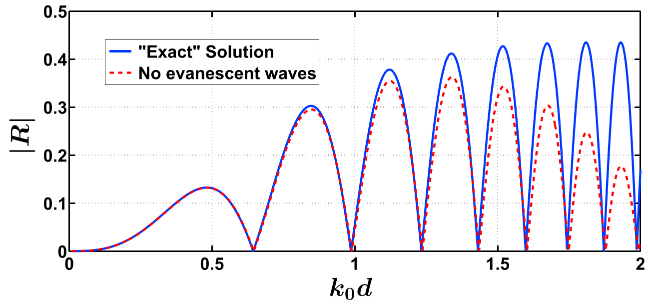

Figure 2. Magnitude of the reflection coefficient using the exact solution from the three boundary conditions (10)–(12) and the approximate solution from the two boundary conditions (1) and (2) omitting the evanescent waves.

In Figure 2, the magnitude of the reflection coefficient is plotted using the “exact” solution from the three boundary conditions (10)–(12) and the approximate solution from the two boundary conditions (1) and (2) omitting the evanescent waves. The macroscopic quadrupolarizability constant α_Q and the thickness z_0 of the slab are normalized to a hypothetical average quadrupole spacing d of the material such that $\alpha_Q = .27d^2$ and the thickness of the slab is $z_0 = 20d$. The angle that the propagation vector of the incidence TM plane wave makes with the normal to the interface is equal to 80° for the results shown in Figure 2. This large angle of oblique incidence is chosen to obtain an appreciable value of the reflection coefficient. At small values of the incident angle, the scattering from the slab is so small that the reflection coefficient is $\ll 1$ until values of k_0d are reached that are much greater than 1.

An important result revealed in Figure 2 is that the original electric quadrupolar boundary conditions in (1) and (2) with the evanescent waves neglected (or assumed to be part of the transition layer) produce accurate results for $k_0d \lesssim 1$, as predicted in the analysis of *Yaghjian* [2014]. In other words, the relative magnitude of the electric quadrupolarization of both the evanescent mode and the extra delta functions in the electric quadrupolarization that exist in the transition layer become negligible as k_0d becomes less than 1.

4. Conclusion

Using a physically reasonable constitutive relation for a continuum material composed of a random distribution of spherically symmetric electric quadrupoles (molecules or inclusions of an array), three boundary conditions at the interface between free space and the quadrupolar continuum are determined directly from Maxwell’s differential equations by modeling the electromagnetic behavior within the thin transition layer between the free space and the inside of the material where the continuum constitutive relation is satisfied. The boundary condition on the tangential magnetic field across the transition layer remains the same as the one derived previously under the assumption that the electric quadrupolarization density contained no effective delta functions in the transition layer [*Yaghjian et al.*, 2014]. However, the previously derived discontinuous boundary condition on the tangential electric field reduces to the tangential electric field being continuous across the transition layer. Moreover, an additional boundary condition (ABC) is found, namely, that the normal component of the electric quadrupolarization density has to be zero on either side of the transition layer ($\hat{\mathbf{n}} \cdot \overline{\mathbf{Q}}^{(2)} \cdot \hat{\mathbf{n}} = 0$). This ABC agrees with the result obtained in *Silveirinha* [2014] under the hypothesis that the macroscopic fields and electric quadrupolarization are piecewise continuous in the transition layer. Here we derive this ABC directly from Maxwell’s differential equations with the constitutive relation in (14), and we find that the diagonal elements of the electric quadrupolarization dyadic can contain delta functions in the transition layer.

An analytic solution to a plane wave incident on an electric quadrupolar slab in which there are two eigenmodes (a propagating and an evanescent mode) traveling in each direction confirms that three boundary conditions are required at each interface to obtain a unique solution. Moreover, a second analytic solution is obtained by applying the two previously defined boundary conditions assuming that the evanescent modes are negligible. Numerical computations of each solution shows what was predicted in a previous paper [*Yaghjian*, 2014], namely, that at the lower frequencies where $k_0d \ll 1$, the two solutions are in close agreement. A future task would be to verify that these analytically derived additional boundary conditions also predict results that compare favorably with the numerical solutions to arrays of electric quadrupolar inclusions.

From this work with electric quadrupolar continua, it appears to be generally possible to derive, directly from the Maxwell differential equations, additional boundary conditions (ABCs) for higher-order multipole media that produce a deterministic set of boundary conditions for the extra modes that may arise depending upon the particular constitutive relations. It is emphasized that the boundary conditions strongly depend on the constitutive relations that hold in the medium.

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