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Electric Quadrupolarizability of a Source-Driven Dielectric Sphere

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Abstract—Since both metamaterials composed of artificial molecules (inclusions in a host material) and natural molecular materials at optical and greater frequencies can exhibit significant electric quadrupolarization as well as electric and magnetic dipolarization, we determine the passive, causal electric quadrupolarizability for a spherically symmetric molecule, namely a dielectric sphere subject to source-driven applied fields. For source-driven excitations, it is found that two electric quadrupolarizability constants are generally required to characterize the electric quadrupolar response of the sphere, with one of the constants multiplying the divergence of the applied electric field. For source-free fields, such as the fields of the eigenmodes of an electric quadrupolar array, the local electric field illuminating each inclusion is solenoidal, the constitutive relation is characterized by just one quadrupolarizability constant, and the electric quadrupolarization becomes traceless. It is also found that the electric quadrupolarization becomes very large and effectively traceless near the resonant frequencies of electrically small plasmonic spheres with negative permittivity and for somewhat larger spheres with positive permittivity.

1. INTRODUCTION

To explain the purpose of this paper, begin by considering Maxwell's space-time differential curl equations (Faraday's and Ampère's laws) for a continuum containing electric quadrupoles as well as electric and magnetic dipoles, namely [1, Chapter 4 and Page 172], [2, 3]

$$\nabla\times\boldsymbol{\mathcal{E}}(\mathbf{r},t)+\frac{\partial\boldsymbol{\mathcal{B}}(\mathbf{r},t)}{\partial t}=0 \tag{1}$$

$$\frac{1}{\mu_0}\nabla\times\boldsymbol{\mathcal{B}}(\mathbf{r},t)-\epsilon_0\frac{\partial\boldsymbol{\mathcal{E}}(\mathbf{r},t)}{\partial t}=\frac{\partial\boldsymbol{\mathcal{P}}(\mathbf{r},t)}{\partial t}+\nabla\times\boldsymbol{\mathcal{M}}(\mathbf{r},t)-\frac{1}{2}\nabla\cdot\frac{\partial\bar{\boldsymbol{\mathcal{Q}}}(\mathbf{r},t)}{\partial t}+\boldsymbol{\mathcal{J}}_a(\mathbf{r},t) \tag{2}$$

where $\boldsymbol{\mathcal{E}}$ and $\boldsymbol{\mathcal{B}}$ are the primary electric and magnetic fields; $\boldsymbol{\mathcal{P}}$ and $\boldsymbol{\mathcal{M}}$ are the electric and magnetic dipolarization densities; $\bar{\boldsymbol{\mathcal{Q}}}$ is the electric dyadic quadrupolarization density; ϵ_0 and μ_0 are the permittivity and permeability of free space; $\boldsymbol{\mathcal{J}}_a$ is the applied electric current density. For an enforced plane-wave applied electric current density $\boldsymbol{\mathcal{J}}_a(\mathbf{r},t)=\mathbf{J}_a(\boldsymbol{\beta},\omega)e^{i(\boldsymbol{\beta}\cdot\mathbf{r}-\omega t)}$, with $\boldsymbol{\beta}=\beta_x\hat{\mathbf{x}}+\beta_y\hat{\mathbf{y}}+\beta_z\hat{\mathbf{z}}$, these Maxwellian equations in (1) and (2) become

$$i\boldsymbol{\beta}\times\mathbf{E}(\boldsymbol{\beta},\omega)-i\omega\mathbf{B}(\boldsymbol{\beta},\omega)=0 \tag{3}$$

$$\frac{1}{\mu_0}i\boldsymbol{\beta}\times\mathbf{B}(\boldsymbol{\beta},\omega)+i\omega\epsilon_0\mathbf{E}(\boldsymbol{\beta},\omega)=-i\omega\mathbf{P}(\boldsymbol{\beta},\omega)+i\boldsymbol{\beta}\times\mathbf{M}(\boldsymbol{\beta},\omega)-\frac{1}{2}\omega\boldsymbol{\beta}\cdot\bar{\mathbf{Q}}(\boldsymbol{\beta},\omega)+\mathbf{J}_a(\boldsymbol{\beta},\omega) \tag{4}$$

where the plane-wave factor $e^{i(\boldsymbol{\beta}\cdot\mathbf{r}-\omega t)}$ multiplying each of the spectral vectors has been suppressed. (Multiplying these two spectral equations by the $e^{i(\boldsymbol{\beta}\cdot\mathbf{r}-\omega t)}$ factor and integrating over (β_x, β_y, β_z, ω)

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from $-\infty$ to $+\infty$, that is, taking the four-fold Fourier transform, returns them to the space-time equations in (1) and (2)).

Equation (4) reveals that the electric quadrupolarization density $\bar{\mathbf{Q}}$ is multiplied by the product $\omega\boldsymbol{\beta}$, whereas the dipolarization densities $\mathbf{P}$ and $\mathbf{M}$ are only multiplied by ω and $\boldsymbol{\beta}$, respectively. This means that for ω and $\boldsymbol{\beta}$ sufficiently small, the contribution of the electric quadrupolarization density $\bar{\mathbf{Q}}$ to the primary fields $\mathbf{E}$ and $\mathbf{B}$ is negligible compared to that of the electric and magnetic dipolarization densities $\mathbf{P}$ and $\mathbf{M}$ (assuming $\mathbf{P}$ or $\mathbf{M}$ approach a nonzero value for small values of the spatial and temporal frequencies $\boldsymbol{\beta}$ and ω) [2]. Consequently, the electric quadrupolarization density in most natural materials is negligible below optical frequencies [4, Page 111]. However, for natural materials used at optical or greater frequencies, and for metamaterial arrays made with artificial molecules (inclusions), the electric quadrupolarization can contribute significantly to the fields [5–7].

To theoretically investigate the propagation and scattering of fields in these electric quadrupolar materials, it is useful to determine the electric quadrupolarizabilities of their constituent molecules or inclusions. Toward this end, we determine in this paper the causal electric quadrupolarizability of a dielectric sphere of radius a in an enforced plane-wave field produced by an applied current density $\mathbf{J}_a(\boldsymbol{\beta},\omega)e^{i(\boldsymbol{\beta}\cdot\mathbf{r}-\omega t)}$ for $|\boldsymbol{\beta}a| \ll 1$. Since this enforced plane wave is source driven with $\pm|\boldsymbol{\beta}|$ not generally equal to the free-space propagation constant, $k_0 = \omega\sqrt{\mu_0\epsilon_0}$, we cannot simply use the Mie solution [8, Sections 9.25–9.27] to the dielectric sphere illuminated by a plane wave in free space to derive the source-driven electric quadrupolarizability of the sphere. Recent progress in the homogenization of artificial materials has shown the effectiveness and rigor enabled by approaches based on a source-driven excitation and Floquet modal analysis [2, 9–11]. The analysis and results of the present paper provide an important step toward a quantitative inclusion of electric quadrupolar effects in these rigorous approaches to the modeling of complex materials.

2. DERIVATION OF THE ELECTRIC QUADRUPOLARIZABILITY

Consider the dyadic electric quadrupole moment of a dielectric sphere of radius a and relative complex permittivity $\epsilon_{\rm r}$ illuminated by an applied, current-driven electric field. This electric quadrupole moment is produced by equal and opposite electric dipole moments in opposite hemispheres of the sphere [12, Section 7.10.2]. To a first approximation, these opposite electric dipole moments are induced by the first spatial derivatives of the incident electric field applied to the sphere. In particular, if we consider a time-harmonic ($e^{-i\omega t}$) applied electric field in the $\hat{\mathbf{z}}$ direction equal to $\boldsymbol{\mathcal{E}}_a = E_{az}e^{i(\beta_z z-\omega t)}\hat{\mathbf{z}}$, then the electric quadrupole moment $\bar{\mathbf{q}}$ of the sphere is given by

$$\bar{\mathbf{q}} = [\alpha_1\hat{\mathbf{z}}\hat{\mathbf{z}} + \alpha_2\left(\hat{\mathbf{x}}\hat{\mathbf{x}} + \hat{\mathbf{y}}\hat{\mathbf{y}}\right)]\,\frac{\partial\mathcal{E}_{az}}{\partial z} \tag{5}$$

where $\mathcal{E}_{az} = E_{az}e^{i\beta_z z}$ (with the time-harmonic dependence suppressed) and $\partial\mathcal{E}_{az}/\partial z = i\beta_z\mathcal{E}_{az} \approx i\beta_z E_{az}$ for a sphere that is small with respect to the enforced spatial wavelength (that is, $|\beta_z|a \ll 1$) but with an arbitrary real value of the temporal frequency ω. The α_1 term represents the $\hat{\mathbf{z}}\hat{\mathbf{z}}$ component of the electric quadrupole moment induced directly by $\partial\mathcal{E}_{az}/\partial z$. The α_2 term represents the orthogonal components of the electric quadrupole moment produced by the equal and opposite electric fields induced in the sphere by the $\hat{\mathbf{z}}\hat{\mathbf{z}}$ component of the electric quadrupole moment. Because of the spherical symmetry, the same α_2 multiplies the $\hat{\mathbf{x}}\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}\hat{\mathbf{y}}$ components. This same argument actually applies successively to each of the components of the electric quadrupole moment to give an infinite series whose sum results in the constants α_1 and α_2.

Another way to understand (5) is to note that, with the applied electric field $\boldsymbol{\mathcal{E}}_a = E_{az}e^{i(\beta_z z-\omega t)}\hat{\mathbf{z}}$, the applied and scattered fields remain unchanged with the insertion of perfectly magnetically conducting infinite planes at $x = 0$ and $y = 0$. Then it becomes apparent that the scattered $\mathcal{E}_x$ and $\mathcal{E}_y$ fields, and, thus the $\mathcal{P}_x$ and $\mathcal{P}_y$ components of polarization have odd symmetry with respect to x and y. This odd symmetry produces opposing electric dipoles in both the x and y directions that generate the q_{xx} and q_{yy} components of electric quadrupole moment in (5).

If the applied electric field also has $\partial\mathcal{E}_{ax}/\partial x$ and $\partial\mathcal{E}_{ay}/\partial y$ variations, the expression in (5) generalizes to

$$\bar{\mathbf{q}} = (\alpha_1 - \alpha_2)\left(\frac{\partial\mathcal{E}_{ax}}{\partial x}\hat{\mathbf{x}}\hat{\mathbf{x}} + \frac{\partial\mathcal{E}_{ay}}{\partial y}\hat{\mathbf{y}}\hat{\mathbf{y}} + \frac{\partial\mathcal{E}_{az}}{\partial z}\hat{\mathbf{z}}\hat{\mathbf{z}}\right) + \alpha_2(\nabla\cdot\boldsymbol{\mathcal{E}}_a)\bar{\mathbf{I}}. \tag{6}$$

Moreover, as explained in the footnote of the next section, the diagonal terms in (6) are not excited by the transverse derivatives of $\boldsymbol{\mathcal{E}}_a$. The constants α_1 and α_2 are independent of $\boldsymbol{\mathcal{E}}_a$ and the orientation of the xyz rectangular coordinate system. Thus, they do not change under a rotation of the coordinates. Also, $\nabla\cdot\boldsymbol{\mathcal{E}}_a$ is an invariant under a rotation of coordinates. However, the dyadic $(\partial\mathcal{E}_{ax}/\partial x\,\hat{\mathbf{x}}\hat{\mathbf{x}}+\partial\mathcal{E}_{ay}/\partial y\,\hat{\mathbf{y}}\hat{\mathbf{y}}+\partial\mathcal{E}_{az}/\partial z\,\hat{\mathbf{z}}\hat{\mathbf{z}})$, which was derived under the assumption that all the other first derivatives of $\boldsymbol{\mathcal{E}}_a$ are zero, transforms to $(\nabla\boldsymbol{\mathcal{E}}_a+\boldsymbol{\mathcal{E}}_a\nabla)/2$ through a Eulerian-angle rotation of coordinates (invoking spherical symmetry and the invariance of $\mathbf{r}$, $\boldsymbol{\mathcal{E}}_a$, and $\nabla\boldsymbol{\mathcal{E}}_a$ under coordinate rotations) and, thus, the full generalization of the expression in (6) for a sphere with $\boldsymbol{\mathcal{E}}_a=\mathbf{E}_a e^{i(\boldsymbol{\beta}\cdot\mathbf{r}-\omega t)}$, $|\boldsymbol{\beta}a|\ll 1$, is

$$\bar{\mathbf{q}}=(\alpha_1-\alpha_2)\frac{1}{2}(\nabla\boldsymbol{\mathcal{E}}_a+\boldsymbol{\mathcal{E}}_a\nabla)+\alpha_2(\nabla\cdot\boldsymbol{\mathcal{E}}_a)\bar{\mathbf{I}} \tag{7}$$

where $\boldsymbol{\mathcal{E}}_a\nabla$ denotes the transpose of the dyadic $\nabla\boldsymbol{\mathcal{E}}_a$.

2.1. Determination of the Constants $\alpha_1(\omega)$ and $\alpha_2(\omega)$

The frequency dependent constants $\alpha_1(\omega)$ and $\alpha_2(\omega)$ in (7) can be found by returning to the simpler Equation (5) and applying a $\partial\mathcal{E}_{az}/\partial z$ field to the dielectric sphere. This applied electric field can be produced by an applied electric current density $\boldsymbol{\mathcal{J}}_a=J_a\hat{\mathbf{z}}e^{i\beta z}$ in which we have omitted the subscripts z on β and J_a, and again suppressed the $e^{-i\omega t}$ harmonic time dependence. Maxwell's equations for the applied fields in free space show that

$$\boldsymbol{\mathcal{E}}_a=E_a\hat{\mathbf{z}}e^{i\beta z} \tag{8}$$

with $E_a=J_a/(i\omega\epsilon_0)$.

In view of (8), the total electric field of the dielectric sphere that produces the electric quadrupole moment satisfies the Maxwellian equations

$$\nabla\times\mathbf{e}-i\omega\mathbf{b}=0 \tag{9}$$

$$\frac{1}{\mu_0}\nabla\times\mathbf{b}+i\omega\epsilon_0\begin{Bmatrix}\epsilon_\mathrm{r}\\1\end{Bmatrix}\mathbf{e}=i\omega\epsilon_0E_ae^{i\beta z}\hat{\mathbf{z}} \tag{10}$$

where the relative dielectric constant equals ϵ_r inside the sphere $(r<a)$ and 1 outside the sphere $(r>a)$. The electric and magnetic fields are denoted by lower case letters in (9) and (10) because they are the microscopic fields of the dielectric sphere.

Substitution of $\mathbf{b}$ from (9) into (10) gives the following equation for the electric field†

$$\nabla\times\nabla\times\mathbf{e}-k_0^2\begin{Bmatrix}\epsilon_\mathrm{r}\\1\end{Bmatrix}\mathbf{e}=-k_0^2E_a(1+i\beta z+\ldots)\hat{\mathbf{z}}. \tag{11}$$

The exponential on the right-hand side of (10) has been replaced by its power series in (11). The first term in the power series induces an electric dipole field in the sphere, the second term produces an electric quadrupole field in the sphere, and $|\beta a|$ for the sphere is assumed small enough that the contribution of the higher-order terms in the power series is negligible. Thus, for the sake of determining the electric quadrupole fields, we can ignore all the terms in the power series except the second and rewrite (11) as

$$\nabla\times\nabla\times\mathbf{e}-k_0^2\begin{Bmatrix}\epsilon_\mathrm{r}\\1\end{Bmatrix}\mathbf{e}=-i\beta k_0^2E_az\hat{\mathbf{z}}. \tag{12}$$

A particular solution $\mathbf{e}_p$ to (12) is

$$\mathbf{e}_p=i\beta E_az\hat{\mathbf{z}}\begin{Bmatrix}\epsilon_\mathrm{r}^{-1}\\1\end{Bmatrix} \tag{13}$$

and the corresponding magnetic field is found from (9) as

$$\mathbf{b}_p=\frac{1}{i\omega}\nabla\times\mathbf{e}_p=0. \tag{14}$$

† It can be proven that if there were a transverse applied electric field on the right-hand side of (11), for example, $\boldsymbol{\mathcal{E}}_a=E_a\hat{\mathbf{x}}e^{i(\boldsymbol{\beta}\cdot\mathbf{r}-\omega t)}$, it would excite only a y-directed magnetic dipole and a $q_{xz}=q_{zx}$ electric quadrupole, thereby confirming in (6) and (7) that the diagonal terms are produced only by the longitudinal derivatives of the applied electric field.

The remaining homogeneous solution satisfies the equations

$$\nabla\times\nabla\times\mathbf{e}_h - k_0^2\begin{Bmatrix}\epsilon_\mathrm{r}\\1\end{Bmatrix}\mathbf{e}_h = 0 \tag{15}$$

$$\mathbf{b}_h = \frac{1}{i\omega}\nabla\times\mathbf{e}_h \tag{16}$$

with continuous total tangential $\mathbf{e}$ and $\mathbf{b}$ across the surface $r = a$ of the sphere, where

$$\mathbf{e} = \mathbf{e}_p + \mathbf{e}_h \tag{17}$$

$$\mathbf{b} = \mathbf{b}_p + \mathbf{b}_h. \tag{18}$$

The function $z\hat{\mathbf{z}}$ can be written in spherical coordinates $(r,\,\theta,\,\phi)$ as

$$z\hat{\mathbf{z}} = r\left(\cos^2\theta\,\hat{\mathbf{r}} - \frac{1}{2}\sin 2\theta\,\hat{\boldsymbol{\theta}}\right) \tag{19}$$

whose tangential component (the $\hat{\boldsymbol{\theta}}$ term) is proportional to Stratton's $\mathbf{n}_{02}$ even electric quadrupolar vector spherical mode. Therefore, the solution to (15)–(16) can be obtained from Pages 416 and 608 of Stratton [8] as

$$\mathbf{e}_h = A\mathbf{n}_{02} = \frac{3A}{2Kr}\left\{f_2(Kr)(3\cos 2\theta + 1)\,\hat{\mathbf{r}} - [Krf_2(Kr)]'\sin 2\theta\,\hat{\boldsymbol{\theta}}\right\} \tag{20}$$

$$\mathbf{b}_h = \frac{1}{i\omega}\nabla\times\mathbf{e}_h = \frac{KA}{i\omega}\mathbf{m}_{02} = \frac{3KA}{2i\omega}f_2(Kr)\sin 2\theta\,\hat{\boldsymbol{\phi}} \tag{21}$$

where

$$K = \begin{Bmatrix}k\\k_0\end{Bmatrix},\quad A = \begin{Bmatrix}A_1\\A_2\end{Bmatrix},\quad f_2(Kr) = \begin{Bmatrix}j_2(kr)\\h_2^{(1)}(k_0r)\end{Bmatrix}\quad \begin{matrix}r<a\\r>a\end{matrix} \tag{22}$$

with $k = k_0\sqrt{\epsilon_\mathrm{r}}$. The j_2 is the second order spherical Bessel function and $h_2^{(1)}$ is the second order spherical Hankel function of the first kind. The prime denotes differentiation with respect to the dimensionless product Kr, the Stratton function $\mathbf{m}_{02} = \nabla\times\mathbf{n}_{02}/K$, and the constants, A_1 and A_2, are found, as follows, from the boundary conditions of continuous tangential $\mathbf{e}$ and $\mathbf{b}$ fields across $r = a$.

Equating across $r = a$ the tangential components of $\mathbf{e}$ and $\mathbf{b}$ obtained by adding the particular fields in (13)–(14) to the homogeneous fields in (20)–(21), we find the two equations

$$\frac{3A_1}{ka}[kaj_2(ka)]' + \frac{i\beta aE_a}{\epsilon_r} = \frac{3A_2}{k_0a}[k_0ah_2^{(1)}(k_0a)]' + i\beta aE_a \tag{23}$$

$$A_2 = \sqrt{\epsilon_\mathrm{r}}\,\frac{j_2(ka)}{h_2^{(1)}(k_0a)}A_1. \tag{24}$$

Solving these two equations for A_1 yields

$$A_0 = \frac{kA_1}{i\beta E_a} = \frac{(k_0a)^2(1-\epsilon_\mathrm{r}^{-1})h_2^{(1)}(k_0a)}{3\left\{\epsilon_\mathrm{r}^{-1}h_2^{(1)}(k_0a)[kaj_2(ka)]' - j_2(ka)[k_0ah_2^{(1)}(k_0a)]'\right\}} \tag{25}$$

with A_2 given by (24) in terms of A_1.

The total electric field inside the sphere can be found by inserting A_1 from (25) into (20) for the homogeneous electric field and adding the particular electric field in (13) to get

$$\mathbf{e}(r<a,\theta) = i\beta E_a\left\{\left[\frac{9A_0j_2(kr)}{k^2r} + \frac{r}{\epsilon_\mathrm{r}}\right]\cos^2\theta - \frac{3A_0j_2(kr)}{k^2r}\right\}\hat{\mathbf{r}} - \frac{1}{2}\left\{\frac{3A_0[krj_2(kr)]'}{k^2r} + \frac{r}{\epsilon_\mathrm{r}}\right\}\sin 2\theta\,\hat{\boldsymbol{\theta}}. \tag{26}$$

The electric polarization density of the sphere is given by

$$\mathbf{p} = (\epsilon_\mathrm{r} - 1)\epsilon_0\mathbf{e}(r<a,\theta) \tag{27}$$

which is used in the next section to evaluate the electric quadrupole moment of the sphere.

2.1.1. Evaluation of the Electric Quadrupole Moment

Since $-i\omega\mathbf{p}$ in (27) is the equivalent electric current density of the dielectric sphere, the electric quadrupole moment of the sphere can be expressed as [1, Page 83]

$$\bar{\mathbf{q}} = \int\limits_V (\mathbf{pr} + \mathbf{rp}) dV \tag{28}$$

where the integration spans the volume V of the dielectric sphere. Carrying out this integration over the sphere by substituting $\mathbf{p}$ and $\mathbf{e}$ from (27) and (26) into (28), then expressing the spherical unit vectors ($\hat{\mathbf{r}}$, $\hat{\boldsymbol{\theta}}$) in terms of the rectangular unit vectors ($\hat{\mathbf{x}}$, $\hat{\mathbf{y}}$, $\hat{\mathbf{z}}$), we find

$$\bar{\mathbf{q}} = \frac{8\pi a^5(\epsilon_\mathrm{r} - 1)\epsilon_0}{15}\left[\left(\frac{1}{\epsilon_\mathrm{r}} + \frac{27A_0A_3}{(ka)^5}\right)\hat{\mathbf{z}}\hat{\mathbf{z}} - \frac{9A_0A_3}{(ka)^5}\bar{\mathbf{I}}\right]\frac{\partial\mathcal{E}_{az}}{\partial z} \tag{29}$$

in which $\partial\mathcal{E}_{az}/\partial z$ has replaced $i\beta E_a$ and

$$A_3 = [1 - (ka)^2/3]\sin ka - ka\cos ka. \tag{30}$$

Comparing (5) with (29), one obtains the constants α_1 and α_2 as

$$\alpha_1(\omega) - \alpha_2(\omega) = \frac{8\pi a^5(\epsilon_\mathrm{r} - 1)\epsilon_0}{15}\left(\frac{1}{\epsilon_\mathrm{r}} + \frac{27A_0A_3}{(ka)^5}\right) \tag{31}$$

$$\alpha_2(\omega) = -\frac{24\pi a^5(\epsilon_\mathrm{r} - 1)\epsilon_0}{5}\frac{A_0A_3}{(ka)^5}. \tag{32}$$

For an arbitrary applied electric field, the full generalization of (29), as given in (7), is

$$\bar{\mathbf{q}} = \frac{8\pi a^5(\epsilon_\mathrm{r}-1)\epsilon_0}{15}\left[\left(\frac{1}{\epsilon_\mathrm{r}} + \frac{27A_0A_3}{(ka)^5}\right)\frac{(\nabla\boldsymbol{\mathcal{E}}_a + \boldsymbol{\mathcal{E}}_a\nabla)}{2} - \frac{9A_0A_3}{(ka)^5}(\nabla\cdot\boldsymbol{\mathcal{E}}_a)\bar{\mathbf{I}}\right] \tag{33}$$

or

$$\bar{\mathbf{q}} = \alpha_{q1}\epsilon_0\frac{(\nabla\boldsymbol{\mathcal{E}}_a + \boldsymbol{\mathcal{E}}_a\nabla)}{2} + \alpha_{q2}\epsilon_0(\nabla\cdot\boldsymbol{\mathcal{E}}_a)\bar{\mathbf{I}} \tag{34}$$

where

$$\alpha_{q1}(\omega) = \frac{8\pi a^5(\epsilon_\mathrm{r} - 1)}{15}\left(\frac{1}{\epsilon_\mathrm{r}} + \frac{27A_0A_3}{(ka)^5}\right) \tag{35}$$

and

$$\alpha_{q2}(\omega) = -\frac{24\pi a^5(\epsilon_\mathrm{r} - 1)}{5}\frac{A_0A_3}{(ka)^5}. \tag{36}$$

Recall that $\nabla\cdot\boldsymbol{\mathcal{E}}_a$ is not necessarily zero because we are assuming a general applied electric field $\boldsymbol{\mathcal{E}}_a$ that is produced by an applied electric current density that (theoretically) permeates the sphere. (Theoretical electric current density with $e^{i\boldsymbol{\beta}\cdot\mathbf{r}}$ spatial dependence that permeates all space becomes confined to a finite volume upon a three-fold $\boldsymbol{\beta} = \beta_y\hat{\mathbf{y}} + \beta_x\hat{\mathbf{x}} + \beta_z\hat{\mathbf{z}}$ Fourier transformation.) Nonetheless, it is shown in Section 4 that the electric-quadrupolarizability expression in (34) is compatible with the Mie solution to the dielectric sphere for which $\nabla\cdot\boldsymbol{\mathcal{E}}_a = 0$.

3. LOW-FREQUENCY APPROXIMATION FOR THE ELECTRIC QUADRUPOLARIZABILITY

The electric quadrupolarizability expression in (34) for the dielectric sphere in a source-driven applied electric field holds for $|\boldsymbol{\beta}a| \ll 1$ and for all real frequencies ω or, equivalently for all k_0a. At low frequencies, $k_0a \to 0$ and the small-argument approximations to A_0 in (25) and A_3 in (30) obtained from

$$h_2^{(1)}(u) \overset{u\to 0}{\sim} -\frac{3i}{u^3}, \quad j_2(u) \overset{u\to 0}{\sim} \frac{u^2}{15} \tag{37}$$

and

$$\sin(u) \overset{u\to 0}{\sim} u - \frac{u^3}{6} + \frac{u^5}{120}, \quad \cos(u) \overset{u\to 0}{\sim} 1 - \frac{u^2}{2} + \frac{u^4}{24} \tag{38}$$

give

$$A_0 \overset{k_0a\to 0}{\sim} \frac{5(\epsilon_\mathrm{r}-1)}{\epsilon_\mathrm{r}(2\epsilon_\mathrm{r}+3)}. \tag{39}$$

$$A_3 \overset{k_0a\to 0}{\sim} \frac{(ka)^5}{45} \tag{40}$$

which reduce (34)–(36) to

$$\bar{\mathbf{q}} \overset{k_0a\to 0}{\sim} \frac{8\pi(\epsilon_\mathrm{r}-1)a^5}{3(2\epsilon_\mathrm{r}+3)}\epsilon_0 \left[\left(\frac{\nabla\boldsymbol{\mathcal{E}}_a + \boldsymbol{\mathcal{E}}_a\nabla}{2}\right) - \frac{(\epsilon_\mathrm{r}-1)}{5\epsilon_\mathrm{r}}(\nabla\cdot\boldsymbol{\mathcal{E}}_a)\bar{\mathbf{I}}\right]. \tag{41}$$

An interesting feature of the quasi-static sphere electric quadrupolarizability in (41) is its resonance as the value of the relative dielectric constant ϵ_r approaches $-3/2$ [13, Eq. (12.1)], [14]. For the general-frequency electric quadrupolarizability factors α_{q1} and α_{q2} in (34)–(36), a large resonance occurs for electrically small spheres ($0 < k_0a < 0.5$) as the relative permittivity varies between the values ($-1.5 < \epsilon_\mathrm{r} < -1.6$), as demonstrated in Figure 1 for the lossless $\epsilon_\mathrm{r} = -1.55$. This large resonance suggests that electrically small metallic spheres at plasmonic frequencies may be used to design metamaterials with a strong electric quadrupolar response. The loss tangents in the permittivity of metals at plasmonic frequencies are higher than 0.1 [15] and, for fully metallic spheres, this amount of loss essentially eliminates the resonance in the electric quadrupolar response, as indicated in Figure 2

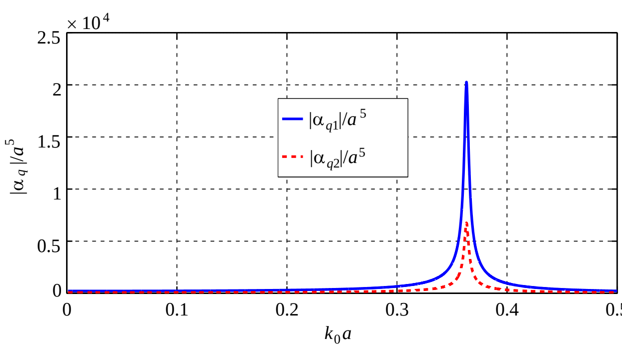

Figure 1. Absolute value of the quadrupolarizability factors versus k_0a for lossless $\epsilon_\mathrm{r} = -1.55$.

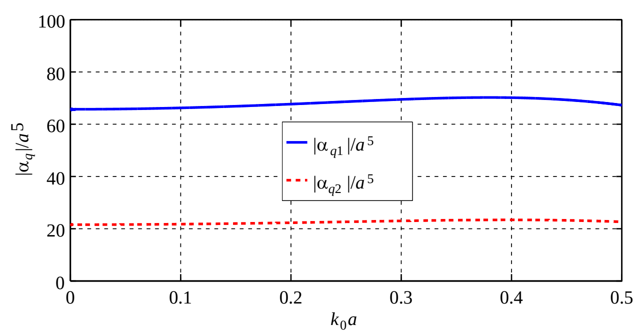

Figure 2. Absolute value of the quadrupolarizability factors versus k_0a for lossy $\epsilon_\mathrm{r} = -1.55(1 - 0.1i)$.

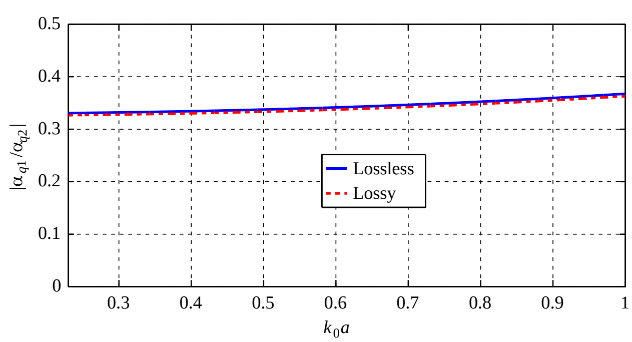

Figure 3. Absolute value of the ratio of the quadrupolarizability factors versus k_0a for the lossless $\epsilon_\mathrm{r} = -1.55$ and the lossy $\epsilon_\mathrm{r} = -1.55(1 - 0.2i)$.

for the lossy $\epsilon_r = -1.55(1 - 0.1i)$ [16]. However, for metallic shells covering ordinary dielectric spheres, the losses can be kept low enough to retain significant electric quadrupolar plasmonic resonances [17]. And, of course, if the fully plasmonic spheres could be enhanced with gainy material, this gainy material could compensate for the reduction in the size of the resonances produced by the passive loss tangents. For spatially dispersive metamaterials used in applications that can tolerate resonances at larger values of k_0a, ordinary dielectric constants with positive real parts can be used to obtain electric quadrupolar resonances. For example, a sphere with $\epsilon_r = 10(1+0.1i)$ has a significant electric quadrupolar resonance at $k_0a \approx 1.7$ despite the fairly high loss tangent of 0.1.

A property of the electric quadrupolarizability relation in (34) near the plasmonic resonance is that the ratio of α_{q2} to α_{q1} is approximately equal to $-1/3$, as shown in Figure 3. In other words, near the plasmonic resonance, the trace of $\bar{\mathbf{q}}/\alpha_{q1}$ is approximately zero even though $\nabla \cdot \boldsymbol{\mathcal{E}}_a \neq 0$. Moreover, this zero-trace ratio of approximately $-1/3$ is maintained if there are losses in the sphere as long as the loss tangent in the permittivity is appreciably less than 1; see Figure 3.

4. COMPATIBILITY WITH THE MIE SOLUTION FOR THE DIELECTRIC SPHERE

The Mie solution for the dielectric sphere has the applied electric field equal to the incident plane wave given by [8, Section 9.25]

$$\boldsymbol{\mathcal{E}}_a = E_0 e^{ik_0 z}\hat{\mathbf{x}} \tag{42}$$

which satisfies the solenoidal condition $\nabla \cdot \boldsymbol{\mathcal{E}}_a = 0$. Such a plane wave will excite only the $q_{xz} = q_{zx}$ components of the electric quadrupole moment of the electrically small dielectric sphere. From [1, Page 96] the far scattered electric field of these components of the electric quadrupole moment is given by

$$\mathbf{E} \overset{r\to\infty}{\sim} -\frac{ik_0^3}{8\pi\epsilon_0} q_{xz}\left(\cos 2\theta \cos\phi\, \hat{\boldsymbol{\theta}} - \cos\theta \sin\phi\, \hat{\boldsymbol{\phi}}\right)\frac{e^{ik_0 r}}{r}. \tag{43}$$

The angular dependence in (43) corresponds to that of Stratton's even $\mathbf{n}_{12}$ vector spherical mode. Therefore, $\mathbf{E}$ in (43) can also be written in terms of the Mie solution in [8, Eq. (5), Page 565], namely

$$\mathbf{E} \overset{r\to\infty}{\sim} -\frac{5i}{2k_0} b_2^r E_0\left(\cos 2\theta \cos\phi\, \hat{\boldsymbol{\theta}} - \cos\theta \sin\phi\, \hat{\boldsymbol{\phi}}\right)\frac{e^{ik_0 r}}{r} \tag{44}$$

where b_2^r is Stratton's notation for the second-order electric-quadrupole scattering coefficient in the Mie solution. Equating the electric far fields in (43) and (44) gives q_{xz} in terms of b_2^r

$$q_{xz} = q_{zx} = \frac{20\pi\epsilon_0}{k_0^4} E_0 b_2^r. \tag{45}$$

The low-frequency ($k_0a \to 0$) expression for $\bar{\mathbf{q}}$ that we derived in (41) predicts that q_{xz} should also be given by

$$q_{xz} = \hat{\mathbf{x}} \cdot \bar{\mathbf{q}} \cdot \hat{\mathbf{z}} = \frac{1}{2}\alpha_{q1}\epsilon_0 \frac{\partial \mathcal{E}_x}{\partial z} = \frac{1}{2} ik_0 \alpha_{q1}\epsilon_0 E_0 \tag{46}$$

with

$$\alpha_{q1} \overset{k_0a\to 0}{\sim} \frac{8\pi(\epsilon_r - 1)a^5}{3(2\epsilon_r + 3)}. \tag{47}$$

With q_{xz} from (46) inserted into (45), one finds that the Stratton Mie scattering coefficient b_2^r should be given approximately by

$$b_2^r \overset{k_0a\to 0}{\sim} i\frac{(k_0a)^5(\epsilon_r - 1)}{15(2\epsilon_r + 3)}. \tag{48}$$

This small k_0a approximation to b_2^r agrees with Stratton's asymptotic expansion of b_2^r in [8, Eq. (40), Page 571] (except for a sign error in Stratton's formula for b_2^r) and with the quasi-static electric quadrupolarizability given by Alù and Engheta [14].

5. PASSIVITY CONDITIONS FOR THE ELECTRIC QUADRUPOLARIZABILITY FACTORS

The time-average power P delivered by an externally applied time-harmonic electric field $\boldsymbol{\mathcal{E}}_a(\mathbf{r})$ to a distribution of time-harmonic electric current density $\boldsymbol{\mathcal{J}}(\mathbf{r})$ in a volume V is given by

$$P = \frac{1}{2}\mathrm{Re}\int_V \boldsymbol{\mathcal{J}}(\mathbf{r}) \cdot \boldsymbol{\mathcal{E}}_a^*(\mathbf{r}) dV \tag{49}$$

where the superscript $*$ denotes the complex conjugate. From the Maxwellian equation in (2), one sees that the equivalent electric current density for electric quadrupolarization density $\bar{\boldsymbol{\mathcal{Q}}}(\mathbf{r})$ is equal to $i\omega\nabla \cdot \bar{\boldsymbol{\mathcal{Q}}}(\mathbf{r})/2$. Thus, the time-average power P_q delivered by an externally applied electric field $\boldsymbol{\mathcal{E}}_a(\mathbf{r})$ to the electric quadrupole moment $\bar{\mathbf{q}}$ at the point $\mathbf{r} = \mathbf{r}_0$, for which $\bar{\boldsymbol{\mathcal{Q}}}(\mathbf{r}) = \bar{\mathbf{q}}\delta(\mathbf{r} - \mathbf{r}_0)$, is given by

$$P_q = \mathrm{Re}\left\{\frac{i\omega}{4}\int_V \nabla \cdot [\bar{\mathbf{q}}\delta(\mathbf{r} - \mathbf{r}_0)] \cdot \boldsymbol{\mathcal{E}}_a^*(\mathbf{r}) dV\right\}. \tag{50}$$

With the help of the vector-dyadic identities

$$\nabla \cdot [\bar{\mathbf{q}}\delta(\mathbf{r} - \mathbf{r}_0)] \cdot \boldsymbol{\mathcal{E}}_a^*(\mathbf{r}) = \nabla\delta(\mathbf{r} - \mathbf{r}_0) \cdot \bar{\mathbf{q}} \cdot \boldsymbol{\mathcal{E}}_a^*(\mathbf{r}) = \nabla \cdot [\delta(\mathbf{r} - \mathbf{r}_0)\bar{\mathbf{q}} \cdot \boldsymbol{\mathcal{E}}_a^*(\mathbf{r})] - \delta(\mathbf{r} - \mathbf{r}_0)\nabla \cdot [\bar{\mathbf{q}} \cdot \boldsymbol{\mathcal{E}}_a^*(\mathbf{r})] \tag{51}$$

(50) integrates to

$$P_q = -\mathrm{Re}\left\{\frac{i\omega}{4}\nabla_0 \cdot [\bar{\mathbf{q}} \cdot \boldsymbol{\mathcal{E}}_a^*(\mathbf{r}_0)]\right\} = -\mathrm{Re}\left\{\frac{i\omega}{4}\mathrm{Tr}[\bar{\mathbf{q}} \cdot \nabla_0\boldsymbol{\mathcal{E}}_a^*(\mathbf{r}_0)]\right\}. \tag{52}$$

With $\boldsymbol{\mathcal{E}}_a = \mathbf{E}_a e^{i\boldsymbol{\beta}\cdot\mathbf{r}}$, we have $\nabla_0\boldsymbol{\mathcal{E}}_a^*(\mathbf{r}_0) = -i\boldsymbol{\beta}\mathbf{E}_a^* e^{-i\boldsymbol{\beta}\cdot\mathbf{r}_0}$ and the electric quadrupole moment $\bar{\mathbf{q}}$ can be written from (34) in terms of $\mathbf{E}_a$ as

$$\bar{\mathbf{q}} = i\left[\alpha_{q1}\epsilon_0\frac{(\boldsymbol{\beta}\mathbf{E}_a + \mathbf{E}_a\boldsymbol{\beta})}{2} + \alpha_{q2}\epsilon_0(\boldsymbol{\beta} \cdot \mathbf{E}_a)\bar{\mathbf{I}}\right] e^{i\boldsymbol{\beta}\cdot\mathbf{r}_0} \tag{53}$$

which converts (52) to

$$P_q = -\mathrm{Re}\left\{\frac{i\omega\epsilon_0}{4}\mathrm{Tr}\left[\frac{\alpha_{q1}}{2}|\boldsymbol{\beta}|^2\mathbf{E}_a\mathbf{E}_a^* + \left(\frac{\alpha_{q1}}{2} + \alpha_{q2}\right)(\boldsymbol{\beta} \cdot \mathbf{E}_a)\boldsymbol{\beta}\mathbf{E}_a^*\right]\right\} \tag{54}$$

or

$$P_q = \frac{\omega\epsilon_0}{4}\mathrm{Im}\left[\frac{\alpha_{q1}}{2}|\boldsymbol{\beta}|^2|\mathbf{E}_a|^2 + \left(\frac{\alpha_{q1}}{2} + \alpha_{q2}\right)|\boldsymbol{\beta} \cdot \mathbf{E}_a|^2\right] \geq 0. \tag{55}$$

The inequality in (55) holds for passive electric quadrupole moments because passivity requires that power cannot be extracted from the quadrupoles and thus the power delivered to the quadrupole moment by the externally applied field must be equal to or greater than zero.

Since $\mathbf{E}_a$ can be chosen arbitrarily, first let $\boldsymbol{\beta} \cdot \mathbf{E}_a = 0$, which implies from (55) the electric quadrupolar passivity condition

$$\omega\mathrm{Im}[\alpha_{q1}] \geq 0 \tag{56}$$

provided, of course, that the microscopic permittivity ϵ_r of the dielectric sphere satisfies the passivity relation $\omega\mathrm{Im}[\epsilon_\mathrm{r}] \geq 0$. As an example of this quadrupolar passivity condition, the real and imaginary parts of α_{q1} are plotted over the domain $0 < k_0 a < 10$ in Figure 4 for the lossy relative microscopic permittivity $\epsilon_\mathrm{r} = -1.55(1 - 0.1i)$ used in Figure 2. Although the real part of α_{q1} in Figure 4 ranges over positive and negative values, its imaginary part remains positive over the entire range.

If next one chooses $\mathbf{E}_a = E_a\hat{\mathbf{z}}$ with $\boldsymbol{\beta} = \beta\hat{\mathbf{z}}$, the inequality in (55) implies the second electric quadrupolar passivity condition

$$\omega\mathrm{Im}[(\alpha_{q1} + \alpha_{q2})] \geq 0 \tag{57}$$

assuming the microscopic permittivity ϵ_r of the dielectric sphere satisfies the passivity relation $\omega\mathrm{Im}[\epsilon_\mathrm{r}] \geq 0$. The real and imaginary parts of $(\alpha_{q1} + \alpha_{q2})$ are plotted over the domain $0 < k_0 a < 10$ in Figure 5 for the lossy relative microscopic permittivity $\epsilon_\mathrm{r} = -1.55(1 - 0.1i)$ used in Figure 2. Again, although the real part of $(\alpha_{q1} + \alpha_{q2})$ in Figure 5 ranges over positive and negative values, its imaginary part remains positive over the entire range.

Lastly, we note that one can prove by inserting A_0 and A_3 from (25) and (30) directly into (36) that $\omega \mathrm{Im}[\alpha_{q2}] \leq 0$. However, it appears that this result cannot be proven from the general passivity condition in (55).

6. CAUSALITY OF THE ELECTRIC QUADRUPOLARIZABILITY FACTORS

One of the distinct advantages of the source-driven constitutive relations is that the constitutive parameters are causal functions of the temporal frequency ω at each fixed value of the real spatial frequency $\boldsymbol{\beta}$ [2]. (This stands in contrast to the noncausal constitutive parameters defined for source-free incident fields [18].) The basic reason for this is that none of the material in the inclusions is subject to incident fields before the time at which the source current turns on (usually taken to be the time $t = 0$) and the polarizations are defined directly in terms of integrals of the sources as in (28). The electric quadrupolarizability constitutive relation for a fixed $\boldsymbol{\beta}$ is given in (53) for a source-driven incident $\mathbf{E}_a$ electric field. This constitutive relation implies that $i\beta\alpha_{q1}(\omega)$ and $i\beta\alpha_{q2}(\omega)$ should be causal functions of ω at a fixed $\boldsymbol{\beta} = \beta\hat{\boldsymbol{\beta}}$. Specifically, in terms of $k_0 a = \omega a/c$, where $c = 1/\sqrt{\mu_0\epsilon_0}$ is the free-space speed of light, these causality relations can be written as

$$\underline{\alpha}_{q1}(\tau) = 2\mathrm{Re}\int_0^{+\infty} [\alpha_{q1}(k_0 a) - \alpha_{q1}(\infty)] e^{-ik_0 a\tau} d(k_0 a) = 0 \tag{58}$$

$$\underline{\alpha}_{q2}(\tau) = 2\mathrm{Re}\int_0^{+\infty} \alpha_{q2}(k_0 a) e^{-ik_0 a\tau} d(k_0 a) = 0 \tag{59}$$

for $\tau = ct/a < 0$, where the sources turn on at the time $t = 0$. (Note that the driving field in (53) is $i\beta\mathbf{E}_a$, whose ω Fourier transform is an imaginary field since the Fourier transform of $\mathbf{E}_a$ must be a real field. Also, $\alpha_{q2}(\infty) = 0$.) The Fourier transforms can be written as in (58) and (59) because of the reality conditions satisfied by $i\beta\alpha_{q1}$ and $i\beta\alpha_{q2}$, namely [2]

$$i\beta\alpha_{q1}(\omega) = [i(-\beta)\alpha_{q1}(-\omega)]^* \quad \Rightarrow \quad \alpha_{q1}(\omega) = \alpha_{q1}^*(-\omega) \tag{60}$$

$$i\beta\alpha_{q2}(\omega) = [i(-\beta)\alpha_{q2}^*(-\omega)]^* \quad \Rightarrow \quad \alpha_{q2}(\omega) = \alpha_{q2}^*(-\omega). \tag{61}$$

It is assumed that the integrands in (58) and (59) approach zero as $k_0 a \to \infty$ and that the frequency dependence of ϵ_r is chosen such that the integrals exist. The causality of the electric quadrupolarizability factors is confirmed, as shown in Figures 6 and 7, by computing the integrals in (58) and (59) for a causal, lossy $\epsilon_\mathrm{r} = 5 + i/(k_0 a)$ inserted into the expressions for α_{q1} and α_{q2} in (35) and (36). (The slight imperfections in the causality curves are caused by the finite increments as well as finite upper limits chosen for the integrations, and the finite time intervals in τ used to plot the curves.) There are discrete jumps in the time-domain electric quadrupolarizability functions $\underline{\alpha}_{q1}(\tau)$ and $\underline{\alpha}_{q2}(\tau)$ separated

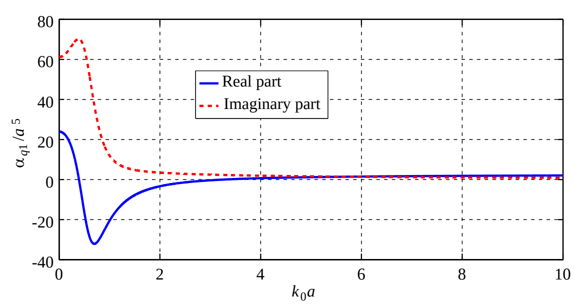

Figure 4. Real and imaginary parts of the quadrupolarizability factor α_{q1} versus $k_0 a$ for the lossy $\epsilon_\mathrm{r} = -1.55(1 - 0.1i)$.

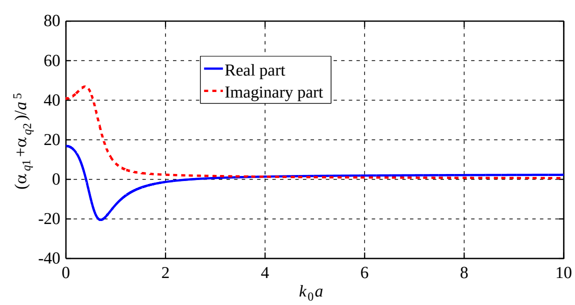

Figure 5. Real and imaginary parts of the quadrupolarizability factor $\alpha_{q1} + \alpha_{q2}$ versus $k_0 a$ for the lossy $\epsilon_\mathrm{r} = -1.55(1 - 0.1i)$.

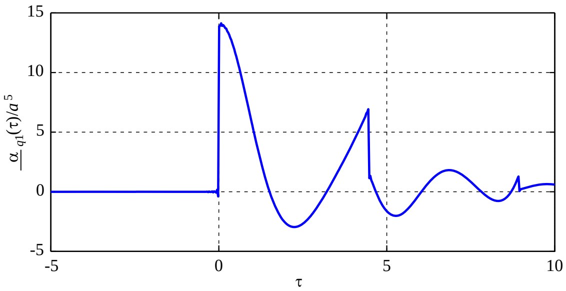

Figure 6. Causality of the quadrupolarizability factor α_{q1} for lossy $\epsilon_r = 5 + i/(k_0a)$; $\tau = ct/a$.

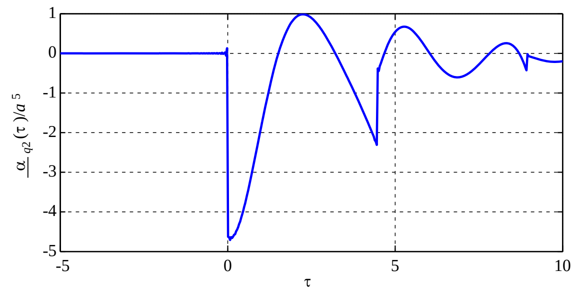

Figure 7. Causality of the quadrupolarizability factor α_{q2} for lossy $\epsilon_r = 5 + i/(k_0a)$; $\tau = ct/a$.

by $\Delta\tau \approx 2\sqrt{\epsilon_{rr}} \approx 4.5$ ($\epsilon_{rr} = 5$ is the real part of ϵ_r) because for $k_0a \gg 1$

$$\alpha_{q1}(k_0a) - \alpha_{q1}(\infty) \sim \frac{8\pi(\epsilon_r - 1)^2}{5\epsilon_r\sqrt{\epsilon_r}} \frac{a^5 \sin(ka)}{k_0a[\cos(ka) - i\sqrt{\epsilon_r}\sin(ka)]} \tag{62}$$

$$\alpha_{q2}(k_0a) \sim -\frac{8\pi(\epsilon_r - 1)^2}{15\epsilon_r\sqrt{\epsilon_r}} \frac{a^5 \sin(ka)}{k_0a[\cos(ka) - i\sqrt{\epsilon_r}\sin(ka)]} \tag{63}$$

and for $k_0a \gg 1$

$$\frac{\sin(ka)}{k_0a[\cos(ka) - i\sqrt{\epsilon_r}\sin(ka)]} \sim \frac{1}{ik_0a}\sum_{n=0}^{\infty} A_n e^{2ink_0\sqrt{\epsilon_{rr}}\,a}, \quad A_n \text{ real} \tag{64}$$

whose Fourier transform is the unit step-function series

$$-2\pi\sum_{n=0}^{\infty} A_n u(\tau - 2n\sqrt{\epsilon_{rr}}\,). \tag{65}$$

The terms in the series of (64) represent successive internal reflections of the fields across the dielectric sphere. A Fourier transform of the electric-dipole polarizability found in equation (13.107) of the first reference in [2] shows that it exhibits similar successive internal reflections in the frequency domain and step-function series in the time domain. These internal reflections are analogous to those found in the scattered field produced by a plane wave incident on a dielectric slab. For the sphere, which is entirely illuminated in the time domain by an applied-current field with delta function ($\delta(t)$) time dependence, the boundary at $r = a$ produces sources of a spherical wave beginning at $t = 0$ that not only radiates outwardly but also inwardly through the center of the sphere and back out to the boundary at $r = a$ where it reflects and transmits another spherical wave. It does this ad infinitum to form the unit step-function time series in (65).

Because $\alpha_{q1}(\omega)$ and $\alpha_{q2}(\omega)$ are causal functions, they also satisfy the Kramers-Kronig causality relations, which can be found by taking the real and imaginary parts of their compact complex version given as [19, Page 98]

$$\alpha_{q1}(\omega) - \alpha_{q1}(\infty) = \frac{i}{\pi} \fint_{-\infty}^{+\infty} \frac{\alpha_{q1}(\nu) - \alpha_{q1}(\infty)}{\omega - \nu} d\nu \tag{66}$$

$$\alpha_{q2}(\omega) = \frac{i}{\pi} \fint_{-\infty}^{+\infty} \frac{\alpha_{q2}(\nu)}{\omega - \nu} d\nu \tag{67}$$

where the lines through the integrals denote Cauchy principal values. It is assumed in (66) that $\alpha_{q1}(\infty)$ is real. A sufficient condition for the principal value integrals to be well defined for all real ω is that $\alpha_{q1}(\omega) - \alpha_{q1}(\infty)$ and $\alpha_{q2}(\omega)$ be Hölder continuous [20, Chapter 1].

7. CONCLUDING DISCUSSION

The expression in (34) for the electric quadrupolarizability of a dielectric sphere of radius a, illuminated by an applied, source-driven electric field $\boldsymbol{\mathcal{E}}_a = \mathbf{E}_a e^{i(\boldsymbol{\beta}\cdot\mathbf{r} - \omega t)}$ with $|\boldsymbol{\beta} a| \ll 1$ and $\nabla \cdot \boldsymbol{\mathcal{E}}_a$ not necessarily equal to zero, holds for all real temporal frequencies ω and for all passive, causal complex dielectric constants ϵ_r. If the incident electric field has no sources over the volume of the sphere, then $\nabla \cdot \boldsymbol{\mathcal{E}}_a = 0$ and (34) reduces to

$$\bar{\mathbf{q}} = \alpha_{q1}\epsilon_0 \left(\frac{\nabla \boldsymbol{\mathcal{E}}_a + \boldsymbol{\mathcal{E}}_a \nabla}{2} \right). \tag{68}$$

The trace of $\bar{\mathbf{q}}$ in (68) is zero since

$$\mathrm{Tr}(\bar{\mathbf{q}}) = \alpha_{q1}\epsilon_0 \left(\nabla \cdot \boldsymbol{\mathcal{E}}_a \right) = 0. \tag{69}$$

We observe from (41) that, even if $\nabla \cdot \boldsymbol{\mathcal{E}}_a \neq 0$, (68) applies approximately to electrically small spheres ($|k_0 a| \ll 1$) for weak dielectrics with ϵ_r close to the free space value, that is, for $\epsilon_r \approx 1$. As $k_0 a \to 0$, the electric quadrupole moment $\bar{\mathbf{q}}$ becomes resonant with an unbounded value as $\epsilon_r \to -3/2$. This unbounded resonance as $k_0 a \to 0$ becomes bounded if either $k_0 a \neq 0$ or ϵ_r has a small loss, that is, if $\omega\epsilon_r$ has a small positive imaginary part.

Lastly, consider a random three-dimensional distribution of inclusions with the electric quadrupolarizability of each of the inclusions given in (34). Assuming the temporal and spatial frequencies of the sources and fields are low enough that the ensemble behaves as an isotropic continuum, the *source-free* modes in the continuum will have a macroscopic electric quadrupole density that is traceless because the electric quadrupole moment of each of the inclusions is traceless in the microscopic, solenoidal, source-free, electric field that illuminates each inclusion. In other words, the source-free, macroscopic, time-harmonic electric quadrupolarization density $\bar{\mathbf{Q}}$ will satisfy the traceless isotropic constitutive relation [3]

$$\bar{\mathbf{Q}} = \alpha_Q \epsilon_0 \left[\frac{1}{2}(\nabla \mathbf{E} + \mathbf{E}\nabla) - \frac{1}{3}(\nabla \cdot \mathbf{E})\bar{\mathbf{I}} \right] \tag{70}$$

where $\mathbf{E}$ is the macroscopic continuum electric field, which is not necessarily solenoidal because

$$\nabla \cdot \mathbf{E} = \frac{1}{2\epsilon_0} \nabla \cdot \left(\nabla \cdot \bar{\mathbf{Q}} \right) \tag{71}$$

even though $\mathrm{Tr}(\bar{\mathbf{Q}}) = 0$. The α_Q in (70) is a bulk electric quadrupolarizability constant.

Even for *source-driven* macroscopic fields, the trace of the electric quadrupolarization density $\bar{\mathbf{Q}}$ does not affect the magnetic field. However, the trace of $\bar{\mathbf{Q}}$ does, in general, change the electric field and thus, in general, it is not permissible to redefine $\bar{\mathbf{Q}}$ to make its trace equal to zero. To prove this, express $\mathbf{B}$ in terms of its vector potential $\mathbf{A}$; specifically

$$\mathbf{B}(\mathbf{r}) = \nabla \times \mathbf{A}(\mathbf{r}) = \frac{i\omega\mu_0}{2} \nabla \times \int_V \bar{\mathbf{Q}}(\mathbf{r}') \cdot \nabla G(\mathbf{r}, \mathbf{r}') dV' \tag{72}$$

with $G(\mathbf{r}, \mathbf{r}') = \exp(ik_0|\mathbf{r} - \mathbf{r}'|)/(4\pi|\mathbf{r} - \mathbf{r}'|)$. Next, let

$$\bar{\mathbf{Q}}_0(\mathbf{r}) = \bar{\mathbf{Q}}(\mathbf{r}) - \mathrm{Tr}[\bar{\mathbf{Q}}(\mathbf{r})]\bar{\mathbf{I}}/3 = \bar{\mathbf{Q}}(\mathbf{r}) - C(\mathbf{r})\bar{\mathbf{I}} \tag{73}$$

so that $\bar{\mathbf{Q}}_0(\mathbf{r})$ has zero trace for all $\mathbf{r}$. Then

$$\mathbf{B}(\mathbf{r}) = \frac{i\omega\mu_0}{2} \nabla \times \int_V \bar{\mathbf{Q}}_0(\mathbf{r}') \cdot \nabla G dV' + \frac{i\omega\mu_0}{2} \nabla \times \int_V C(\mathbf{r}') \nabla G dV' = \frac{i\omega\mu_0}{2} \nabla \times \int_V \bar{\mathbf{Q}}_0(\mathbf{r}') \cdot \nabla G(\mathbf{r}, \mathbf{r}') dV' \tag{74}$$

because $\nabla \times \nabla G = 0$. Thus, $\mathbf{B}(\mathbf{r})$ has not been changed by making the trace of $\bar{\mathbf{Q}}(\mathbf{r})$ zero, and it can be rewritten in terms of the traceless $\bar{\mathbf{Q}}_0(\mathbf{r})$.

The electric field $\mathbf{E}(\mathbf{r})$ can be expressed in terms of the magnetic field and electric quadrupolar density $\bar{\mathbf{Q}}(\mathbf{r})$ along with the applied current density $\mathbf{J}_a(\mathbf{r})$ from Maxwell's second equation as

$$\mathbf{E}(\mathbf{r}) = -\frac{1}{i\omega\mu_0\epsilon_0} \nabla \times \mathbf{B}(\mathbf{r}) + \frac{1}{2\epsilon_0} \nabla \cdot \bar{\mathbf{Q}}(\mathbf{r}) + \frac{\mathbf{J}_a(\mathbf{r})}{i\omega\epsilon_0}. \tag{75}$$

This equation reveals that even though making the trace of $\bar{\mathbf{Q}}(\mathbf{r})$ zero, that is, replacing it by $\bar{\mathbf{Q}}_0(\mathbf{r})$, does not change $\mathbf{B}(\mathbf{r})$ or $\mathbf{J}_a(\mathbf{r})$, it would change $\nabla \cdot \bar{\mathbf{Q}}(\mathbf{r})$ by an amount equal to $\nabla C(\mathbf{r})$, and therefore $\mathbf{E}(\mathbf{r})$ would change unless $\nabla C(\mathbf{r}) = 0$. Consequently, for source-driven macroscopic fields, the electric quadrupolarization density $\bar{\mathbf{Q}}(\mathbf{r})$ cannot generally be redefined to reduce its trace to zero.

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REFERENCES

1. Papas, C. H., *Theory of Electromagnetic Wave Propagation*, McGraw-Hill, New York, 1965; and Dover, New York, 1988.
2. Yaghjian, A. D., A. Alù, and M. G. Silveirinha, "Anisotropic representation for spatially dispersive periodic metamaterial arrays," *Transformation Electromagnetics and Metamaterials*, Chapter 13, Springer, 2014, also "Homogenization of spatially dispersive metamaterial arrays in terms of generalized electric and magnetic polarizations," *Photonics and Nanostructures — Fundamentals and Applications*, 374–396, Nov. 2013.
3. Yaghjian, A. D., "Boundary conditions for electric quadrupolar continua," *Radio Science*, Vol. 49, 1289–1299, Dec. 2014.
4. Scott, W. T., *The Physics of Electricity and Magnetism*, Robert E. Krieger, Huntington, NY, 1977.
5. Raab, R. E. and O. L. de Lange, *Multipole Theory in Electromagnetism*, Clarendon Press, Oxford NY, 2005.
6. Cho, D. J., F. Wang, X. Zhang, and Y. R. Shen, "Contribution of the electric quadrupole resonance in optical metamaterials," *Phys. Rev. B*, Vol. 78, 121101(1–4), 2008.
7. Silveirinha, M. G., "Boundary conditions for electric quadrupolar metamaterials," *New Journal of Physics*, Vol. 16, 083042(1–30), 2014.
8. Stratton, J. A., *Electromagnetic Theory*, McGraw-Hill, New York, 1941.
9. Agranovich, V. M. and V. L. Ginzburg, *Spatial Dispersion in Crystal Optics and the Theory of Excitons*, Wiley-Interscience, New York, 1966; also see 2nd Edition, Springer, New York, 1984.
10. Silveirinha, M. G., "Nonlocal homogenization theory of structured materials," *Metamaterials Handbook: Theory and Phenomena of Metamaterials*, Chapter 13, F. Capolino (ed.), CRC Press, Boca Raton, 2009,
11. Chebykin, A. V., A. A. Orlov, A. V. Vozianova, S. I. Maslovski, Y. S. Kivshar, and P. A. Belov, "Nonlocal effective medium model for multilayered metal-dielectric metamaterials," *Phys. Rev. B*, Vol. 84, 115438(1–9), 2011.
12. Van Bladel, J. G., *Electromagnetic Fields*, 2nd Edition, IEEE/Wiley, Piscataway, NJ, 2007.
13. Bohren, C. F. and D. R. Huffman, *Absorption and Scattering of Light by Small Particles*, John Wiley, New York, 1983.
14. Alù, A. and N. Engheta, "Guided propagation along quadrupolar chains of plasmonic nanoparticles," *Phys. Rev. B*, Vol. 79, 235412(1–12), 2009.
15. Naik, G. V., V. M. Shalaev, and A. Boltasseva, "Alternative plasmonic materials: beyond gold and silver," *Adv. Mater.*, Vol. 25, 3264–3294, 2013.
16. Alù, A. and N. Engheta, "Enhanced directivity from subwavelength infrared/optical nano-antennas loaded with plasmonic materials or metamaterials," *IEEE Trans. Antennas Propagat.*, Vol. 55, 3027–3029, Nov. 2007.
17. Oldenburg, S. J., G. D. Hale, J. B. Jackson, and N. J. Halas, "Light scattering from dipole and quadrupole nanoshell antennas," *Applied Phys. Letts.*, Vol. 75, 1063–1065, Aug. 1999.
18. Alù, A., A. D. Yaghjian, R. A. Shore, and M. G. Silveirinha, "Causality relations in the homogenization of metamaterials," *Phys. Rev. B*, Vol. 84, 054305(1–16), Aug. 2011.
19. Hansen, T. B. and A. D. Yaghjian, *Plane-wave Theory of Time-domain Fields*, IEEE/Wiley, New York, 1999.
20. Lu, J. K., *Boundary Value Problems for Analytic Functions*, World Scientific, New York, 1993.