Polarizability and Fluctuation-Dissipation Theorem for a Point Dipole: Does Shape Matter?

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Abstract. The concept of a point dipole is potentially ambiguous due to inherent singularity of electro-magnetic fields at its location. We discuss this concept from several points of view. First, we consider a point dipole as a singular point in space whose sole ability is to be polarized due to the external electric field. We introduce the source Green's dyadic that provides a unified albeit empiric description of the contribution of the dipole to the electromagnetic properties of the whole space. We argue that this is the most complete, concise, and unambiguous definition of a point dipole and its polarizability. Next, we revisit classic expressions for absorption and emission power by integration of Poynting vector over a surface enclosing the point dipole, thereby avoiding the singularity. We also consider the energy balance between the fluctuating dipole moment and the medium (thermal bath) to derive the fluctuation-dissipation theorem in terms of fluctuating dipole moment. This solves the long-standing controversy in the literature. Second, the same results can be obtained for a very small homogeneous sphere, in which the internal field is known to be constant. This leads to unambiguous microscopic definition of the particle dipole moment and polarizability in terms of its size and refractive index. Third, and most interestingly, we generalize this microscopic description to small particles of arbitrary shape. Both bare (electrostatic) and dressed (corrected) polarizabilities are defined as double integrals of the corresponding dyadic transition operator over the particle's volume.

INTRODUCTION

The "point dipole" is a convenient abstraction for small particles [1]. However, it is a mathematical singularity and therefore should be used with caution. For instance, it is no simple question how to write and use Maxwell's equations in the point where such a dipole is located. There are phenomenological ways to describe optical properties of point dipole, such as absorption and emission, however, some expressions are contradictory or excessive. Moreover, it is not always clear how to adapt these formulae to the case of complex environment (from the free-space case). This problem becomes especially prominent when one tries to formulate the fluctuation-dissipation theorem (FDT) for the fluctuating dipole moment $\mathbf{p}_{\mathrm{fl}}$. For instance, Refs. [2,3] provided contradicting expressions for this FDT, and although more recent Ref. [4] pointed the inconsistency in Ref. [3], it didn't attributed the issue to the formulation of the FDT, thus adding to the controversy rather than solving it.

In this contribution, we aim to provide a unified and self-consistent description of the "point dipole" concept from all possible points of view in the framework of the frequency-domain electrodynamics. We start with an empiric description, based on postulating the optical properties of a point dipole using only a single quantity – its polarizability tensor $\bar{\boldsymbol{\alpha}}$. But instead of a common approach to postulate several expressions for various quantities (e.g., scattering, absorption, and emission) [1], we limit ourselves to a single one – an expression for the source Green's dyadic $\bar{\mathbf{G}}_{\mathrm{s}}(\mathbf{r},\mathbf{r}')$, which describes a response of a given environment with the point dipole to arbitrary point-source excitation [5]. We show that all other optical properties can be rigorously expressed through $\bar{\mathbf{G}}_{\mathrm{s}}(\mathbf{r},\mathbf{r}')$. Combining the latter with thermal-bath-equilibrium arguments, we even derive the FDT in terms of $\mathbf{p}_{\mathrm{fl}}$.

Next, we consider the simplest non-singular model for a point dipole – that is a limit of a small sphere. Naturally all the phenomenological properties are then rigorously derived from the electrostatic limit of the Mie theory, as is

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commonly considered in the literature [1]. Finally, we relax the requirement of particle sphericity and homogeneity keeping the main conclusion that all optical properties (at distances much larger than the particle size) are determined solely by $\bar{\boldsymbol{\alpha}}$, and derive an explicit microscopic expression for the latter. While the notion that any small particle looks like a point dipole is universally accepted [6], we are not aware of explicit prove for all scattering quantities, especially including the consideration of the FDT. Much more detailed description of these results can be found in [7].

PHENOMENOLOGICAL DESCRIPTION OF A POINT DIPOLE

Let us denote the wavelength λ, the wavenumber $k \stackrel{\text{def}}{=} 2\pi/\lambda$, the internal (characteristic) size of a dipole a, and its size parameter $x \stackrel{\text{def}}{=} ka$. The concept of a point dipole implies that it acts on the electromagnetic fields *outside*, i.e. at distances much larger than a, and that $a \ll \lambda \Leftrightarrow x \ll 1$. The most universal way to express this effect is through $\bar{\mathbf{G}}_{\mathrm{s}}(\mathbf{r},\mathbf{r}')$, which we postulate in the following form:

$$\bar{\mathbf{G}}_{\mathrm{s}}(\mathbf{r},\mathbf{r}') = \bar{\mathbf{G}}(\mathbf{r},\mathbf{r}') + \omega^2\mu_0\bar{\mathbf{G}}(\mathbf{r},\mathbf{r}_0)\cdot\bar{\boldsymbol{\alpha}}\cdot\bar{\mathbf{G}}(\mathbf{r}_0,\mathbf{r}'), \qquad \mathbf{r},\mathbf{r}' \neq \mathbf{r}_0, \tag{1}$$

where $\bar{\mathbf{G}}(\mathbf{r},\mathbf{r}')$ is the Green's dyadic for a given environment (without the dipole), $\mathbf{r}_0$ is the dipole location, and SI units are used. The environment may be arbitrarily complex, but all its components (other particles, substrate, etc.) are assumed to be separated from the dipole (much farther than a). Then

$$\bar{\mathbf{G}}(\mathbf{r},\mathbf{r}') = \bar{\mathbf{G}}_0(\mathbf{r},\mathbf{r}') + \bar{\mathbf{G}}_{\mathrm{env}}(\mathbf{r},\mathbf{r}'), \tag{2}$$

where $\bar{\mathbf{G}}_0$ is the free-space Green's dyadic [5], and $\bar{\mathbf{G}}_{\mathrm{env}}$ is smooth and finite when $\mathbf{r}$ and/or $\mathbf{r}'$ approach $\mathbf{r}_0$. Eq. (1) becomes more natural when applied to an arbitrary source $\mathbf{J}_{\mathrm{s}}(\mathbf{r}')$:

$$\mathbf{E}(\mathbf{r}) = \mathrm{i}\omega\mu_0\bar{\mathbf{G}}_{\mathrm{s}}(\mathbf{r},\mathbf{r}')\cdot\mathbf{J}_{\mathrm{s}}(\mathbf{r}') = \mathbf{E}_0(\mathbf{r}) + \omega^2\mu_0\bar{\mathbf{G}}(\mathbf{r},\mathbf{r}_0)\cdot\mathbf{p} = \mathbf{E}_0(\mathbf{r}) + \mathbf{E}_{\mathrm{sca}}(\mathbf{r}), \tag{3}$$

where $\mathbf{E}_0(\mathbf{r}) = \mathrm{i}\omega\mu_0\bar{\mathbf{G}}(\mathbf{r},\mathbf{r}')\cdot\mathbf{J}_{\mathrm{s}}(\mathbf{r}')$ is the field without the dipole, $\mathbf{p} = \bar{\boldsymbol{\alpha}}\cdot\mathbf{E}_0(\mathbf{r}_0)$ is the induced dipole moment, and $\mathbf{E}_{\mathrm{sca}}$ is the scattered field. Note that $\mathbf{E}(\mathbf{r})$ and, hence, $\bar{\mathbf{G}}_{\mathrm{s}}(\mathbf{r},\mathbf{r}')$ are potentially measurable quantities, which can then be used to obtain $\mathbf{p}$ and $\bar{\boldsymbol{\alpha}}$. Moreover, Eq. (1) implicitly considers $\bar{\boldsymbol{\alpha}}$, as relating $\mathbf{p}$ with the external fields, thus corresponding to renormalized (or *dressed*) polarizability [1].

By integrating the Poynting vector over the sphere with radius R_0 around the dipole ($a \ll R_0 \ll \lambda$), calculated from Eq. (3), we obtain power rates for extinction and scattering [5]:

$$W_{\mathrm{ext}} = \frac{\omega}{2}\Im(\mathbf{E}_0^*\cdot\mathbf{p}) = \frac{\omega}{2}\mathbf{E}_0^*\cdot\bar{\boldsymbol{\alpha}}^I\cdot\mathbf{E}_0, \qquad W_{\mathrm{sca}} = \frac{\omega^3\mu_0}{2}\mathbf{p}^*\cdot\bar{\mathbf{G}}^I(\mathbf{r}_0,\mathbf{r}_0)\cdot\mathbf{p}, \tag{4}$$

where $\Im$ denotes the imaginary part, $\mathbf{E}_0$ without an argument is the same as $\mathbf{E}_0(\mathbf{r}_0)$, $\bar{\boldsymbol{\alpha}}^I \stackrel{\text{def}}{=} (\bar{\boldsymbol{\alpha}} - \bar{\boldsymbol{\alpha}}^H)/(2\mathrm{i})$ is (–i) times the skew-Hermitian part of $\bar{\boldsymbol{\alpha}}$ (H denotes the Hermitian transpose), and

$$\bar{\mathbf{G}}^I(\mathbf{r},\mathbf{r}') \stackrel{\text{def}}{=} \frac{1}{2\mathrm{i}}[\bar{\mathbf{G}}(\mathbf{r},\mathbf{r}') - \bar{\mathbf{G}}^H(\mathbf{r}',\mathbf{r})] \tag{5}$$

is the kernel of self-adjoint emission operator [5]. Note that $\bar{\boldsymbol{\alpha}}^I = \Im(\bar{\boldsymbol{\alpha}})$ for any symmetric polarizability (as is commonly the case), and $\bar{\mathbf{G}}^I(\mathbf{r},\mathbf{r}') = \Im[\bar{\mathbf{G}}(\mathbf{r},\mathbf{r}')]$ in any reciprocal environment. In particular, in free space $\bar{\mathbf{G}}_0^I(\mathbf{r}_0,\mathbf{r}_0) = k\bar{\mathbf{I}}/(6\pi)$, leading to the classical result $W_{\mathrm{sca}} = k\omega^3\mu_0|\mathbf{p}|^2/(12\pi)$. The absorption power rate is

$$W_{\mathrm{abs}} = W_{\mathrm{ext}} - W_{\mathrm{sca}} = \frac{\omega}{2}\mathbf{E}_0^*\cdot\bar{\boldsymbol{\beta}}\cdot\mathbf{E}_0, \qquad \bar{\boldsymbol{\beta}} \stackrel{\text{def}}{=} \bar{\boldsymbol{\alpha}}^I - \omega^2\mu_0\bar{\boldsymbol{\alpha}}^H\cdot\bar{\mathbf{G}}^I(\mathbf{r}_0,\mathbf{r}_0)\cdot\bar{\boldsymbol{\alpha}}. \tag{6}$$

Note that $\bar{\boldsymbol{\beta}}$ is always Hermitian. For a scalar polarizability $\bar{\boldsymbol{\alpha}} = \alpha\bar{\mathbf{I}}$ ($\bar{\mathbf{I}}$ is the unit dyadic) the definition of $\bar{\boldsymbol{\beta}}$ reduces to the one, known previously [2,8]. Moreover, in many cases the second term in this definition can be neglected, i.e. $W_{\mathrm{abs}} \approx W_{\mathrm{ext}}$. One can even argue that this assumption is always correct for a truly point dipole, unless $W_{\mathrm{abs}} = 0$.

The above expressions for basic optical properties are well-known, but typically they (or part of them) are postulated. Here we rigorously derived them from a single Eq. (1). Moreover, these expressions do not include the electrostatic (*bare*) polarizability $\bar{\boldsymbol{\chi}}$, which is commonly defined by excluding the "self-action" of the dipole, calculated through the regularized Green's tensor [1]

$$\bar{\boldsymbol{\chi}}^{-1} \stackrel{\text{def}}{=} \bar{\boldsymbol{\alpha}}^{-1} + \omega^2\mu_0\bar{\mathbf{G}}_{\mathrm{r}}(\mathbf{r}_0,\mathbf{r}_0), \tag{7}$$

$$\bar{\mathbf{G}}_{\mathrm{r}}(\mathbf{r},\mathbf{r}') \stackrel{\text{def}}{=} \mathrm{i}\bar{\mathbf{G}}_0^I(\mathbf{r},\mathbf{r}') + \bar{\mathbf{G}}_{\mathrm{env}}(\mathbf{r},\mathbf{r}') = \bar{\mathbf{G}}(\mathbf{r},\mathbf{r}') - \Re[\bar{\mathbf{G}}_0(\mathbf{r},\mathbf{r}')] \Rightarrow \bar{\mathbf{G}}_{\mathrm{r}}^I(\mathbf{r},\mathbf{r}') = \bar{\mathbf{G}}^I(\mathbf{r},\mathbf{r}'). \tag{8}$$

Eq. (8) is a generalization of definition in [1] specifying only the value of $\bar{\mathbf{G}}_{\mathrm{r}}(\mathbf{r}_0,\mathbf{r}_0)$, and we used $[\bar{\mathbf{G}}_0(\mathbf{r},\mathbf{r}') + \bar{\mathbf{G}}_0^H(\mathbf{r}',\mathbf{r})]/2 = \Re[\bar{\mathbf{G}}_0(\mathbf{r},\mathbf{r}')]$. Moreover, $\bar{\boldsymbol{\chi}}$ is a natural defining quantity for absorption [1]:

$$W_{\mathrm{abs}} = -\frac{\omega}{2}\Im[\mathbf{p}^*\cdot\bar{\boldsymbol{\chi}}^{-1}\cdot\mathbf{p}] \Leftrightarrow \bar{\boldsymbol{\beta}} = -\bar{\boldsymbol{\alpha}}^H\cdot(\bar{\boldsymbol{\chi}}^{-1})^I\cdot\bar{\boldsymbol{\alpha}}, \tag{9}$$

which follows from Eq. (6). Interestingly, the last part of Eq. (9) can be rewritten as

$$\bar{\boldsymbol{\beta}} = [\bar{\mathbf{I}} - \omega^2\mu_0\bar{\mathbf{G}}_{\mathrm{r}}(\mathbf{r}_0,\mathbf{r}_0)\cdot\bar{\boldsymbol{\chi}}]^{-H}\cdot\bar{\boldsymbol{\chi}}^I\cdot[\bar{\mathbf{I}} - \omega^2\mu_0\bar{\mathbf{G}}_{\mathrm{r}}(\mathbf{r}_0,\mathbf{r}_0)\cdot\bar{\boldsymbol{\chi}}]^{-1} \cong \bar{\boldsymbol{\chi}}^I. \tag{10}$$

Here and further $\cong$ denotes that the equality is asymptotically exact when $x \to 0$ (hence, $k^3\|\bar{\boldsymbol{\chi}}\|/\varepsilon_0 \ll 1$). And this holds for any absorption in contrast to neglecting the second term in Eq. (6).

Next, we allow the point dipole to have the fluctuating dipole moment $\mathbf{p}_{\mathrm{fl}}$ in addition to the induced one. Requiring the thermal equilibrium of this dipole with the thermal bath (fluctuating electric field in the environment, for which the correlation is well-established [9]), we obtain

$$\langle \mathbf{p}_{\mathrm{fl}} \otimes \mathbf{p}_{\mathrm{fl}}^{*} \rangle = \frac{\Theta(\omega,T)}{\pi\omega}\bar{\boldsymbol{\beta}}, \quad \Theta(\omega,T) \stackrel{\mathrm{def}}{=} \frac{\hbar\omega}{1-\mathrm{e}^{-\hbar\omega/k_{\mathrm{B}}T}}, \tag{11}$$

where k_{B} and $\hbar$ is the Boltzmann and reduced Planck constant, respectively. This settles the controversy between Refs. [2–4], by showing that the error in [3] is due to erroneous use of $\bar{\boldsymbol{\alpha}}^{I}$ instead of $\bar{\boldsymbol{\beta}}$ in Eq. (11).

LIMIT OF A SMALL SPHERE

Consider a small sphere with radius a, volume V, and dielectric permittivity ε. In this section we limit ourselves to scalar ε and particles located in free space, in order to use the standard Mie theory. In contrast to the previous section, the most unambiguous quantity is electrostatic polarizability [1]:

$$\bar{\boldsymbol{\chi}} = 3V\varepsilon_0 \frac{\varepsilon-\varepsilon_0}{\varepsilon+2\varepsilon_0}\bar{\mathbf{I}}, \tag{12}$$

while definition of $\bar{\boldsymbol{\alpha}}$ is potentially ambiguous. It is common to express it through the first Mie coefficient a_1 [1]:

$$\bar{\boldsymbol{\alpha}} \cong \frac{6\pi\mathrm{i}\varepsilon_0}{k^3}a_1, \tag{13}$$

but there are two caveats. First, a_1 describes the dipole moment of a sphere in response to the projection of $\mathbf{E}_0(\mathbf{r})$ on the vector spherical harmonic $\mathbf{N}_{\mathrm{e}11}^{(1)}$ [10]; the latter is constant (and, hence, the projection equals $\mathbf{E}_0$) only in the limit $x \to 0$. Second, the same limit is required for the dipole approximation [e.g., Eq. (1)] to hold, which is equivalent to neglecting all other Mie coefficients. Therefore, while Eq. (13) can, in principle, be used for any a, the resulting $\bar{\boldsymbol{\alpha}}$ describes all optical properties of a sphere only when $x \ll 1$ (thus, $\cong$ in the equation).

Let us consider this limit in details [10]:

$$a_1 = -\mathrm{i}\frac{2x^3}{3}\frac{\varepsilon-\varepsilon_0}{\varepsilon+2\varepsilon_0} - \mathrm{i}\frac{2x^5}{5}\frac{(\varepsilon-\varepsilon_0)(\varepsilon-2\varepsilon_0)}{(\varepsilon+2\varepsilon_0)^2} + \left(\frac{2x^3}{3}\frac{\varepsilon-\varepsilon_0}{\varepsilon+2\varepsilon_0}\right)^2 + \mathcal{O}(x^7). \tag{14}$$

Taking a straightforward limit $x \to 0$ leaves only the first term in Eq. (14) and, hence, leads to $\bar{\boldsymbol{\alpha}} \approx \bar{\boldsymbol{\chi}}$ resulting in negative $\mathrm{W}_{\mathrm{abs}}$ for real ε, as discussed around Eqs. (6)–(10). To solve this problem, the limiting value should lead to both real and imaginary parts of a_1 (and $\bar{\boldsymbol{\alpha}}$) being asymptotically correct, even for real ε. This leaves the first and the third term in Eq. (14), leading to

$$\bar{\boldsymbol{\alpha}} \cong 3V\varepsilon_0 \frac{\varepsilon-\varepsilon_0}{\varepsilon+2\varepsilon_0}\left(1+\mathrm{i}\frac{2x^3}{3}\frac{\varepsilon-\varepsilon_0}{\varepsilon+2\varepsilon_0}\right)\bar{\mathbf{I}} = \bar{\boldsymbol{\chi}}\cdot[\bar{\mathbf{I}} + \mathrm{i}\omega^2\mu_0\bar{\mathbf{G}}_0^{I}(\mathbf{r}_0,\mathbf{r}_0)\cdot\bar{\boldsymbol{\chi}}] \cong \bar{\boldsymbol{\chi}}\cdot[\bar{\mathbf{I}} - \mathrm{i}\omega^2\mu_0\bar{\mathbf{G}}_0^{I}(\mathbf{r}_0,\mathbf{r}_0)\cdot\bar{\boldsymbol{\chi}}]^{-1}, \tag{15}$$

where the last transformation uses $k^3\|\bar{\boldsymbol{\chi}}\|/\varepsilon_0 \ll 1$ and makes the result equivalent to Eq. (9).

Next we consider the limiting values of scattering and absorption efficiencies:

$$Q_{\mathrm{abs}} = 4x\Im\left(\frac{\varepsilon-\varepsilon_0}{\varepsilon+2\varepsilon_0}\right)[1+\mathcal{O}(x^2)] \cong 4x\Im\left(\frac{\varepsilon-\varepsilon_0}{\varepsilon+2\varepsilon_0}\right), \tag{16}$$

$$Q_{sca} = \frac{8}{3}x^4\left|\frac{\varepsilon-\varepsilon_0}{\varepsilon+2\varepsilon_0}\right|^2 + \mathcal{O}(x^6) \cong \frac{8}{3}x^4\left|\frac{\varepsilon-\varepsilon_0}{\varepsilon+2\varepsilon_0}\right|^2, \tag{17}$$

which are related to the corresponding power rates as

$$W = \frac{c\varepsilon_0}{2}\pi a^2 Q. \tag{18}$$

Importantly, Eqs. (16) and (17) provide one-term asymptotic approximation (uniform for any ε), while the expression for $Q_{\mathrm{ext}} = Q_{\mathrm{abs}} + Q_{\mathrm{sca}}$ must contain both these terms. And all these expressions agree with Eqs. (4) and (6), if one uses Eqs. (10) and (15).

We are not analysing the FDT specifically for a sphere, since it would either use thermal-equilibrium arguments, as in previous section, or a cumbersome analysis of fluctuating currents inside the sphere, as in the next section.

SMALL PARTICLE OF ARBITRARY SHAPE AND COMPOSITION

Now we consider arbitrary particle with small volume V (characteristic size a) and again allow for arbitrary environment specified by $\bar{\mathbf{G}}(\mathbf{r},\mathbf{r}')$. Let the particle have dyadic electric permittivity $\bar{\boldsymbol{\varepsilon}}(\mathbf{r})$, denote total field everywhere in space $\mathbf{E}(\mathbf{r})$, polarization density $\mathbf{P}(\mathbf{r}) \stackrel{\mathrm{def}}{=} [\bar{\boldsymbol{\varepsilon}}(\mathbf{r}) - \varepsilon_0\bar{\mathbf{I}}]\cdot\mathbf{E}(\mathbf{r})$, and recall the defining property of the transition dyadic $\bar{\mathbf{T}}$ [11,12]

$$\omega^2\mu_0\mathbf{P}(\mathbf{r}) = \int_V \mathrm{d}^3\mathbf{r}'\bar{\mathbf{T}}(\mathbf{r},\mathbf{r}')\cdot\mathbf{E}_0(\mathbf{r}'). \tag{19}$$

The domain of $\bar{\mathbf{T}}$ (with respect to both arguments) is commonly extended to $\mathbb{R}^3$ by assuming that $\bar{\mathbf{T}}(\mathbf{r},\mathbf{r}') = \bar{\mathbf{0}}$ unless both $\mathbf{r}\in V$ and $\mathbf{r}'\in V$. Then the integration domain in Eq. (19) can be replaced by $\mathbb{R}^3$, and these domains are omitted in the following derivations. Further we assume $\bar{\mathbf{T}}(\mathbf{r},\mathbf{r}')$ a known quantity, since it is fully determined by $\bar{\boldsymbol{\varepsilon}}(\mathbf{r})$, e.g., through an integral equation [12].

Naturally, $\mathbf{p} = \int \mathrm{d}^3\mathbf{r}\,\mathbf{P}(\mathbf{r})$ implying together with Eq. (19) and assumption of constant $\mathbf{E}_0(\mathbf{r})$ inside V

$$\bar{\boldsymbol{\alpha}} \cong \frac{1}{\omega^2\mu_0}\iint \mathrm{d}^3\mathbf{r}\,\mathrm{d}^3\mathbf{r}'\bar{\mathbf{T}}(\mathbf{r},\mathbf{r}'), \tag{20}$$

which can, in principle, be used for arbitrary particles. But for larger particles the whole notion of polarizability is somewhat ambiguous, as discussed in the previous section. Eq. (20) only describes the response (total dipole moment) to the constant $\mathbf{E}_0(\mathbf{r})$, which is asymptotically equivalent to the response to $\mathbf{N}_{\mathrm{e}11}^{(1)}(\mathbf{r})$ as in Eq. (13). For extra insight, consider a general relation [12] for $\mathbf{r},\mathbf{r}'\notin V$:

$$\bar{\mathbf{G}}_\mathrm{s}(\mathbf{r},\mathbf{r}') = \bar{\mathbf{G}}(\mathbf{r},\mathbf{r}') + \iint \mathrm{d}^3\mathbf{r}''\mathrm{d}^3\mathbf{r}'''\bar{\mathbf{G}}(\mathbf{r},\mathbf{r}'')\cdot\bar{\mathbf{T}}(\mathbf{r}'',\mathbf{r}''')\cdot\bar{\mathbf{G}}(\mathbf{r}''',\mathbf{r}'), \tag{21}$$

which is equivalent to Eq. (1) if and only if both $\bar{\mathbf{G}}$ can be brought outside of the integral. The latter can be justified when $\bar{\mathbf{G}}(\mathbf{r},\mathbf{r}'') \cong \bar{\mathbf{G}}(\mathbf{r},\mathbf{r}_0)$ and $\bar{\mathbf{G}}(\mathbf{r}''',\mathbf{r}') \cong \bar{\mathbf{G}}(\mathbf{r}_0,\mathbf{r}')$, which in turn follows from $a \ll |\mathbf{r}-\mathbf{r}_0|, |\mathbf{r}'-\mathbf{r}_0|, \lambda$. To conclude, the simple Eq. (1), a quintessence of a point dipole, is valid only if the particle is very small. And the definition of $\bar{\boldsymbol{\alpha}}$ is only asymptotically unambiguous, i.e. all reasonable definitions converge to the same limit.

Let us further discuss the general definition for $\bar{\boldsymbol{\chi}}$. A natural generalization of Eq. (20) is

$$\bar{\boldsymbol{\chi}} = \frac{1}{\omega^2\mu_0}\iint \mathrm{d}^3\mathbf{r}\,\mathrm{d}^3\mathbf{r}'\bar{\mathbf{T}}_\mathrm{st}(\mathbf{r},\mathbf{r}') \tag{22}$$

(note the exact =), where $\bar{\mathbf{T}}_\mathrm{st}(\mathbf{r},\mathbf{r}')$ satisfies the same integral equation as $\bar{\mathbf{T}}(\mathbf{r},\mathbf{r}')$ [12], but with $\bar{\mathbf{G}}(\mathbf{r},\mathbf{r}')$ replaced by

$$\bar{\mathbf{G}}_\mathrm{st}(\mathbf{r},\mathbf{r}') = \frac{1}{k^2}\lim_{k\to 0} k^2\bar{\mathbf{G}}_0(\mathbf{r},\mathbf{r}')\,, \Rightarrow \bar{\mathbf{G}}_\mathrm{st}^I(\mathbf{r},\mathbf{r}') = \bar{\mathbf{0}}. \tag{23}$$

Importantly, $\bar{\mathbf{G}}_\mathrm{st}(\mathbf{r},\mathbf{r}')$, $\bar{\mathbf{T}}_\mathrm{st}(\mathbf{r},\mathbf{r}')$, and $\bar{\boldsymbol{\chi}}$ depend neither on the environment nor on k (nor ω). More specifically, $\bar{\mathbf{G}}_\mathrm{st}(\mathbf{r},\mathbf{r}')$ and $\bar{\mathbf{T}}_\mathrm{st}(\mathbf{r},\mathbf{r}')$ depend on k only through the common factor $k^{\pm 2}$ due to the used system of units. The abovementioned integral equations for $\bar{\mathbf{T}}(\mathbf{r},\mathbf{r}')$ and $\bar{\mathbf{T}}_\mathrm{st}(\mathbf{r},\mathbf{r}')$ imply

$$\bar{\mathbf{T}}_\mathrm{st}(\mathbf{r},\mathbf{r}') + \iint \mathrm{d}^3\mathbf{r}''\mathrm{d}^3\mathbf{r}'''\bar{\mathbf{T}}(\mathbf{r},\mathbf{r}'')\cdot[\bar{\mathbf{G}}(\mathbf{r}'',\mathbf{r}''') - \bar{\mathbf{G}}_\mathrm{st}(\mathbf{r}'',\mathbf{r}''')]\cdot\bar{\mathbf{T}}_\mathrm{st}(\mathbf{r}''',\mathbf{r}') = \bar{\mathbf{T}}(\mathbf{r},\mathbf{r}'). \tag{24}$$

Using Eq. (8) we obtain

$$\bar{\mathbf{G}}(\mathbf{r},\mathbf{r}') - \bar{\mathbf{G}}_\mathrm{st}(\mathbf{r},\mathbf{r}') = \bar{\mathbf{G}}_\mathrm{r}(\mathbf{r},\mathbf{r}') + \bar{\mathbf{G}}_1(\mathbf{r},\mathbf{r}'), \qquad \bar{\mathbf{G}}_1(\mathbf{r},\mathbf{r}') \stackrel{\mathrm{def}}{=} \Re[\bar{\mathbf{G}}_0(\mathbf{r},\mathbf{r}')] - \bar{\mathbf{G}}_\mathrm{st}(\mathbf{r},\mathbf{r}') = \mathcal{O}(|\mathbf{r}-\mathbf{r}'|^{-1}), \tag{25}$$

which together with Eq. (24), integrated twice over volume, Eqs. (20), (22), and constancy of $\bar{\mathbf{G}}_\mathrm{r}(\mathbf{r},\mathbf{r}')$ in V leads to

$$\bar{\boldsymbol{\alpha}} - \bar{\boldsymbol{\chi}} \cong \omega^2\mu_0\bar{\boldsymbol{\alpha}}\cdot\bar{\mathbf{G}}_\mathrm{r}(\mathbf{r}_0,\mathbf{r}_0)\cdot\bar{\boldsymbol{\chi}} + \frac{1}{\omega^2\mu_0}\iint\iint \mathrm{d}^3\mathbf{r}\,\mathrm{d}^3\mathbf{r}'\mathrm{d}^3\mathbf{r}''\mathrm{d}^3\mathbf{r}'''\bar{\mathbf{T}}(\mathbf{r},\mathbf{r}'')\cdot\bar{\mathbf{G}}_1(\mathbf{r},\mathbf{r}')\cdot\bar{\mathbf{T}}_{\mathbf{st}}(\mathbf{r}''',\mathbf{r}'). \tag{26}$$

Everything without integral corresponds exactly to Eq. (7), while the integral is tricky since $\bar{\mathbf{G}}_1(\mathbf{r},\mathbf{r}')$ is definitely non-constant inside V (and even weakly singular). Importantly, this integral describes the non-radiative correction [cf. $\mathcal{O}(x^5)$ term in Eq. (14)], since $\bar{\mathbf{G}}_1^I(\mathbf{r},\mathbf{r}') = \bar{\mathbf{0}}$, while the term with $\bar{\mathbf{G}}_\mathrm{r}(\mathbf{r}_0,\mathbf{r}_0)$ – the radiative one [$\bar{\mathbf{G}}_\mathrm{r}^I(\mathbf{r},\mathbf{r}') \neq \bar{\mathbf{0}}$]. The non-radiative correction is about $\mathcal{O}(1/x)$ times larger than the radiative one, but still $\mathcal{O}(x^2)$ of the main term. However, it can be shown that the non-radiative correction to $\bar{\boldsymbol{\alpha}}^I$ is $\mathcal{O}(x^2\|\bar{\boldsymbol{\chi}}\|^I)$, which is asymptotically negligible for any absorption, in contrast to radiative one, which is $\mathcal{O}(x^3\|\bar{\boldsymbol{\chi}}\|)$. Therefore, the integral can be completely neglected in Eq. (26).

One can further use the results of the phenomenological section, since they all follow from Eq. (1), but we provide a microscopic derivation below, based on [5]:

$$\begin{aligned} W_\mathrm{ext} &= -\frac{\omega}{2}\int \mathrm{d}^3\mathbf{r}\,\Im[\mathbf{E}_0(\mathbf{r})\mathbf{P}^*(\mathbf{r})] \cong \frac{\omega}{2}\Im(\mathbf{E}_0^*\cdot\mathbf{p}), \\ W_\mathrm{sca} &= \frac{\omega^3\mu_0}{2}\iint \mathrm{d}^3\mathbf{r}\,\mathrm{d}^3\mathbf{r}'\mathbf{P}^*(\mathbf{r})\cdot\bar{\mathbf{G}}^I(\mathbf{r},\mathbf{r}')\cdot\mathbf{P}(\mathbf{r}') \cong \frac{\omega^3\mu_0}{2}\mathbf{p}^*\cdot\bar{\mathbf{G}}^I(\mathbf{r}_0,\mathbf{r}_0)\cdot\mathbf{p}, \end{aligned} \tag{27}$$

where we used the constancy of $\mathbf{E}_0(\mathbf{r})$ and $\bar{\mathbf{G}}^I(\mathbf{r},\mathbf{r}')$ inside V. The final expression for W_sca has been already obtained in [5]. Expression for W_abs is then obtained exactly as in Eq. (6). To obtain the FDT in terms of $\mathbf{p}_\mathrm{fl}$, we recall that microscopically it is caused by internal fluctuating currents $\mathbf{J}_\mathrm{fl}(\mathbf{r})$, for which the FDT is unambiguously known [9,13]:

$$\langle \mathbf{J}_\mathrm{fl}(\mathbf{r})\otimes\mathbf{J}_\mathrm{fl}^*(\mathbf{r}')\rangle = \frac{\omega}{\pi}\Theta(\omega,T)\bar{\boldsymbol{\varepsilon}}^I(\mathbf{r})\delta(\mathbf{r}-\mathbf{r}'), \tag{28}$$

The fluctuation polarization density $\mathbf{P}(\mathbf{r})$ is produced both directly by $\mathbf{J}_{\mathrm{fl}}(\mathbf{r})$ and by induced currents due to the field

$$\mathbf{E}_0(\mathbf{r}) = \mathrm{i}\omega\mu_0 \int \overline{\mathbf{G}}(\mathbf{r},\mathbf{r}') \cdot \mathbf{J}_{\mathrm{fl}}(\mathbf{r}'), \tag{29}$$

where for brevity we omit the rigorous handling of the strong singularity of the Green's tensor. Using Eq. (19) we obtain

$$\mathbf{P}_{\mathrm{fl}}(\mathbf{r}) = \frac{\mathrm{i}}{\omega}\left[\mathbf{J}_{\mathrm{fl}}(\mathbf{r}) + \iint \mathrm{d}^3\mathbf{r}'\,\mathrm{d}^3\mathbf{r}''\,\overline{\mathbf{T}}(\mathbf{r},\mathbf{r}') \cdot \overline{\mathbf{G}}(\mathbf{r},\mathbf{r}'') \cdot \mathbf{J}_{\mathrm{fl}}(\mathbf{r}'')\right]. \tag{30}$$

After cumbersome derivations, using $\mathbf{p}_{\mathrm{fl}} = \int \mathrm{d}^3\mathbf{r}\,\mathbf{P}_{\mathrm{fl}}(\mathbf{r})$ and Eq. (28) we obtain

$$\langle \mathbf{p}_{\mathrm{fl}} \otimes \mathbf{p}_{\mathrm{fl}}^* \rangle = \frac{\Theta(\omega,T)}{\pi\omega^3\mu_0}\left[\iint \mathrm{d}^3\mathbf{r}\,\mathrm{d}^3\mathbf{r}'\,\overline{\mathbf{T}}^I(\mathbf{r},\mathbf{r}') - \iint\iint \mathrm{d}^3\mathbf{r}\,\mathrm{d}^3\mathbf{r}'\mathrm{d}^3\mathbf{r}''\mathrm{d}^3\mathbf{r}'''\,\overline{\mathbf{T}}^H(\mathbf{r}',\mathbf{r}) \cdot \overline{\mathbf{G}}^I(\mathbf{r}',\mathbf{r}'') \cdot \overline{\mathbf{T}}(\mathbf{r}'',\mathbf{r}''')\right], \tag{31}$$

which leads to Eq. (11) using Eq. (20) and constancy of $\overline{\mathbf{G}}^I(\mathbf{r}',\mathbf{r}'')$ inside V.

CONCLUSION

In this paper, we comprehensively considered the point-dipole concept from several points of view. First, as a singular point in space whose sole ability is to be polarized due to the external electric field. The polarizability $\overline{\boldsymbol{\alpha}}$ is then *empirically* defined as a constant in the expression for $\overline{\mathbf{G}}_{\mathrm{s}}(\mathbf{r},\mathbf{r}')$, from which we derived all optical properties of the dipole, including the FDT in terms of $\mathbf{p}_{\mathrm{fl}}$. The latter solves the long-standing controversy in the literature. Naturally, the same conclusions were obtained for a small sphere by taking the $x \to 0$ limit of the Mie theory. But it clearly illustrates that $\overline{\boldsymbol{\chi}}$ has unambiguous *microscopic* definition, while $\overline{\boldsymbol{\alpha}}$ – only asymptotically unambiguous one. It also shows that the limit of $x \to 0$ must be taken uniformly with respect to ε (the magnitude of absorption), which is required to get the asymptotically correct limits for both real and imaginary parts of $\overline{\boldsymbol{\alpha}}$ (and $\overline{\boldsymbol{\chi}}$). Next, we developed a general microscopic theory for particles of arbitrary shape and composition, based on volume-integral-formulation, which includes the definitions of $\overline{\boldsymbol{\alpha}}$ and $\overline{\boldsymbol{\chi}}$ as double integrals of the corresponding transition dyadics over V, and the relation between the two. This relation includes general expression for both radiative and non-radiative corrections. The latter is generally hard to compute, but can be asymptotically neglected for arbitrary absorption. Finally, we re-derived the FDT in terms of $\mathbf{p}_{\mathrm{fl}}$, starting with the $\mathbf{J}_{\mathrm{fl}}(\mathbf{r})$, for which the FDT is unambiguously known. Overall, this microscopic theory provides a solid foundation for the widely used phenomenological expressions related to a point dipole and sheds light on subtle points, which sometimes lead to confusion. The shape of the dipole does affect $\overline{\boldsymbol{\alpha}}$, but all its electromagnetic properties are further determined solely by this tensor.

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