The Fluctuation-Dissipation Theorem for a Point Dipole

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The ubiquitous concept of a point dipole is potentially ambiguous due to inherent singularity of electro-magnetic fields at its location. This leads to long-standing controversies in the literature, related to the definition of the dipole polarizability and formulation of the fluctuation-dissipation theorem (FDT) in terms of the dipole moment. 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. In addition to classical expressions for absorption and emission power, it allows us to rigorously derive the most general form of the FDT in terms of the fluctuating dipole moment. Next, we obtain the same results for a very small homogeneous sphere or even for a small particle of arbitrary shape, using the Lorenz–Mie theory and volume-integral formulation, respectively. This leads to unambiguous microscopic definition of the particle dipole moment and polarizability in terms of its size and refractive index.

INTRODUCTION

The "point dipole" is a convenient abstraction for small particles [1]. However, it is not a 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 a point dipole in free space, such as absorption and emission. However, some expressions are contradictory or excessive, and it is not obvious how to adapt these formulae to the case of complex environment. This problem becomes especially prominent when one tries to formulate the FDT for the fluctuating dipole moment $\mathbf{p}_{\text{fl}}$. For instance, Refs. [2,3] provided contradicting expressions for this FDT, and although more recent Ref. [4] pointed the inconsistency in [3], it did not attributed the issue to the formulation of the FDT, thus adding to the controversy rather than solving it.

To address this confusion, we 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 observables (e.g., scattering, absorption, and emission) [1], we limit ourselves to a single one – an expression for the source Green's dyadic $\bar{\mathbf{G}}_{\text{S}}(\mathbf{r},\mathbf{r}')$, 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 Lorenz–Mie theory, as is 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. Much more detailed description of these results can be found in [6,7].

PHENOMENOLOGICAL THEORY OF A POINT DIPOLE

Let us denote the wavelength λ, the wavenumber $k \stackrel{\mathrm{def}}{=} 2\pi/\lambda$, the internal (characteristic) size of a dipole a, and its size parameter $x \stackrel{\mathrm{def}}{=} ka$. 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 $\mathbf{J}_{\mathrm{s}}(\mathbf{r}')$:

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

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 and $\mathbf{p} = \bar{\boldsymbol{\alpha}}\cdot\mathbf{E}_0(\mathbf{r}_0)$ is the induced dipole moment. Moreover, Eq. (1) implicitly considers $\bar{\boldsymbol{\alpha}}$, as relating $\mathbf{p}$ with the external fields, thus corresponding to the dynamic (or *dressed*) polarizability [1].

We omit here the derivation of the extinction, absorption, and scattering powers by integrating the Poynting vector over the sphere with radius R_0 around the dipole ($a \ll R_0 \ll \lambda$). However, it brings forward the following convenient definitions. $\bar{\boldsymbol{\alpha}}^I \stackrel{\mathrm{def}}{=} (\bar{\boldsymbol{\alpha}} - \bar{\boldsymbol{\alpha}}^H)/(2\mathrm{i})$ (H denotes the Hermitian transpose),

$$\bar{\mathbf{G}}^I(\mathbf{r},\mathbf{r}') \stackrel{\mathrm{def}}{=} \frac{1}{2\mathrm{i}}[\bar{\mathbf{G}}(\mathbf{r},\mathbf{r}') - \bar{\mathbf{G}}^H(\mathbf{r}',\mathbf{r})] \tag{4}$$

is the kernel of self-adjoint emission operator [5] (regular for $\mathbf{r} \to \mathbf{r}'$), and

$$\bar{\boldsymbol{\beta}} \stackrel{\mathrm{def}}{=} \bar{\boldsymbol{\alpha}}^I - \omega^2\mu_0\bar{\boldsymbol{\alpha}}^H\cdot\bar{\mathbf{G}}^I(\mathbf{r}_0,\mathbf{r}_0)\cdot\bar{\boldsymbol{\alpha}}. \tag{5}$$

Note that $\bar{\boldsymbol{\beta}}$ is always Hermitian and asymptotically equivalent to $\bar{\boldsymbol{\chi}}^I$, where $\bar{\boldsymbol{\chi}}$ is the static (*bare*) polarizability. Importantly, the asymptotic equivalence is considered *uniformly for any level of absorption*, i.e. it implies the same leading order (in terms of x) both for absorbing and non-absorbing dipole material.

Next, we allow the point dipole to have the fluctuating dipole moment $\mathbf{p}_{\text{fl}}$ 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 [8]), we obtain

$$\langle \mathbf{p}_{\text{fl}} \otimes \mathbf{p}_{\text{fl}}^* \rangle = \frac{\Theta(\omega, T)}{\pi\omega} \bar{\boldsymbol{\beta}}, \quad \Theta(\omega, T) \stackrel{\text{def}}{=} \frac{\hbar\omega}{1 - \mathrm{e}^{-\hbar\omega/k_{\mathrm{B}}T}}, \tag{6}$$

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. (6).

A SMALL PARTICLE AS A MODEL OF A POINT DIPOLE

First, let us consider a small sphere with radius a, volume V, and scalar dielectric permittivity ε. Then, the roles of $\bar{\boldsymbol{\alpha}}$ and $\bar{\boldsymbol{\chi}}$ are reversed. The latter is well-known from electrostatics [1]:

$$\bar{\boldsymbol{\chi}} = 3V\varepsilon_0 \frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} \bar{\mathbf{I}}, \tag{7}$$

while definition of $\bar{\boldsymbol{\alpha}}$ is potentially ambiguous, although this fact is not universally acknowledged. The possible expressions are based on the first Mie coefficients a_1 or d_1 (in the expansion of the scattered or internal fields, respectively) or on the total dipole moment. The corresponding expressions have the same leading terms $O(x^3)$ and the, so-called, radiative-reaction term $O(x^6)$, which is non-negligible for real ε.

By contrast, they differ in terms $O(x^5)$, which are commonly called non-radiative corrections. There is a lot of papers advocating the use of one of this non-radiative corrections. However, our analysis clearly shows that they all are asymptotically negligible. In other words, they are significant only for large enough x, for which the scattered field is different from that for a point dipole. The differences between these $O(x^5)$ terms are related to the interchange of vector spherical wave functions (VSWFs) and their small-argument limits. The most accurate term depends on the specific application (measureable quantity of interest).

Overall, the conclusion stays the same for arbitrary small particle with anisotropic permittivity function $\bar{\boldsymbol{\varepsilon}}(\mathbf{r})$ in an arbitrary environment specified by $\bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}')$. While a closed-form solution is not available in this case, everything can be written in terms of the transition dyadic $\bar{\mathbf{T}}(\mathbf{r}, \mathbf{r}')$, which by definition relates the polarization density inside the particle to the incident field [9]. In particular, $\bar{\boldsymbol{\alpha}}$ and $\bar{\boldsymbol{\chi}}$ are expressed as double integrals of $\bar{\mathbf{T}}$ and its static limit $\bar{\mathbf{T}}_{\text{st}}$. Again, the electrostatic expression for $\bar{\boldsymbol{\chi}}$ is completely unambiguous, while several variations for $\bar{\boldsymbol{\alpha}}$ exist, based on adding VSWFs as multipliers inside the integral over $\bar{\mathbf{T}}$.

We also derived the FDT in terms of $\mathbf{p}_{\mathrm{fl}}$ [Eq. (6)] using this microscopic model. Now $\mathbf{p}_{\mathrm{fl}}$ is not postulated bur is rather expressed in terms of the fluctuating currents in the particle volume $\mathbf{J}_{\mathrm{fl}}(\mathbf{r})$, for which the FDT is unambiguously known [8,10]:

$$\langle \mathbf{J}_{\mathrm{fl}}(\mathbf{r}) \otimes \mathbf{J}_{\mathrm{fl}}^{*}(\mathbf{r}') \rangle = \frac{\omega}{\pi} \Theta(\omega, T) \bar{\boldsymbol{\varepsilon}}^{I}(\mathbf{r}) \delta(\mathbf{r} - \mathbf{r}'). \tag{8}$$

CONCLUSION

We showed that a point dipole is a singular point in space whose polarizability $\bar{\boldsymbol{\alpha}}$ is *empirically* defined as a constant in the expression for $\bar{\mathbf{G}}_{\mathrm{S}}(\mathbf{r}, \mathbf{r}')$. From the latter we derived all optical properties of the dipole, including the FDT in terms of $\mathbf{p}_{\mathrm{fl}}$, solving the long-standing controversy. Naturally, the same conclusions were obtained for a small sphere by taking the $x \to 0$ limit of the Lorenz–Mie theory. But in this case $\bar{\boldsymbol{\chi}}$ has unambiguous *microscopic* definition, while $\bar{\boldsymbol{\alpha}}$ – only asymptotically unambiguous one. General consideration of particles of arbitrary shape and composition, based on volume-integral-formulation, leads to the same conclusion. The shape and composition of the dipole does affect $\bar{\boldsymbol{\alpha}}$, but all its electromagnetic properties are further determined solely by this tensor. Finally, we provided a microscopic derivation of the FDT in terms of $\mathbf{p}_{\mathrm{fl}}$, starting from the $\mathbf{J}_{\mathrm{fl}}(\mathbf{r})$, for which the FDT is unambiguously known.

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