



A point electric dipole: From basic optical properties to the fluctuation–dissipation theorem

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ABSTRACT

We comprehensively review the deceptively simple concept of dipole scattering in order to uncover and resolve all ambiguities and controversies existing in the literature. First, we consider a point electric dipole in a non-magnetic environment as a singular point in space whose sole ability is to be polarized due to the external electric field. We show that the postulation of the Green's dyadic of the specific form provides the unified description of the contribution of the dipole into the electromagnetic properties of the whole space. This is the most complete, concise, and unambiguous definition of a point dipole and its polarizability. All optical properties, including the fluctuation–dissipation theorem for a fluctuating dipole, are derived from this definition. Second, we obtain the same results for a small homogeneous sphere by taking a small-size limit of the Lorenz–Mie theory. Third, and most interestingly, we generalize this microscopic description to small particles of arbitrary shape. Both bare (static) and dressed (dynamic) polarizabilities are defined as the double integrals of the corresponding dyadic transition operator over the particle's volume. While many derivations and some results are novel, all of them follow from or are connected with the existing literature, which we review throughout the paper.

1. Introduction

Light–matter interaction is the principal phenomenon for many aspects of life and technology. Generally, electromagnetic radiation may be absorbed by atoms and molecules with subsequent dissipation into another energy form or re-emission at the same or different frequency. Unfortunately, this quantum description becomes too complicated for objects consisting of many atoms, which is the case even for small particles. Absorption and scattering of light by particles much smaller than the wavelength has been widely discussed in theoretical physics since the pioneering works of Lord Rayleigh [1,2]. The Rayleigh approximation can be used for any small scatterer with the proper definition of the dipole moment. Although the dipole moment and polarizability seem intuitively clear, their calculation from the particle's size, shape, and internal properties may require cumbersome numerical computations for electrodynamic and/or quantum mechanical problems. It becomes especially complicated in nanophotonics, where atomic and electrodynamic theories meet [3]. Moreover, different definitions of polarizability exist.

The dipole moment is a well-defined concept when there is a charge distribution in space. The definition can be found, e.g., in Wikipedia [4] and classical textbooks [5]. However, the charge distribution is usually unknown in practice and should be found through a solution of a separate problem. For instance, an electrostatic problem should be solved to find the induced dipole moment

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of a particle in a uniform electric field. Many papers are devoted to the solution of this problem in order to obtain the polarizability of particles with complex shape, see, e.g., [6,7]. This polarizability relates the dipole moment to the incident electrostatic field. It is common to define a (quasi-)static, or *bare* polarizability, which is the same quantity as above but calculated with the use of a frequency-dependent permittivity. However, if the incident field is time-dependent, the induced dipole moment is given by the solution to the electrodynamic problem, and the corresponding *dynamic* polarizability is, in principle, different. Sometimes the static and dynamic polarizabilities are erroneously interchanged in various formulas, leading to confusion. Therefore, it is important to review their similarities and differences.

First, absorption and scattering by small dipolar particles are important from the experimental point of view. Since scattering is present only in the electrodynamic problems, the static polarizability is not generally applicable. For instance, the Rayleigh approximation for spheres was obtained (as a limiting case of the Lorenz–Mie theory) in [8]. It was shown that both the absorption and scattering efficiencies of a small sphere can be expressed in terms of its static polarizability only if scattering is negligible. In this case, the static polarizability is given by the Clausius–Mossotti formula, where the permittivity is considered at the frequency of the incident field. The correction to forward scattering amplitude was introduced by van de Hulst [9] to account for scattering. Since then the radiative corrections have been used for particles of various shapes in many applications. These corrections allow one to adapt the zero-frequency formulas to a system, which includes radiative (scattering) effects, i.e., to obtain the dynamic polarizability (also referred to as *dressed* to reflect the fact of correction), which better describes the experimental data at non-zero frequencies [10–12].

Second, the expression for the polarizability of a dipole affects the accuracy of computational methods, such as the coupled dipole approximation (CDA) and the discrete dipole approximation (DDA, [13,14]). In both methods, a system of point dipoles or voxels scatters and/or emits radiation. While the DDA employs radiative correction for each dipole, the equations for the CDA can be written using the static polarizability, but with an additional dipole *self-field* [15], whose absorption by the dipole compensates the radiated (scattered) energy [10]. In the end, both formulations are equivalent, but the self-field is an auxiliary (non-measurable) quantity, whose introduction may lead to confusion.

Third, the theory of radiative heat transfer relies on the fluctuation–dissipation theorem (FDT) [3]. In electromagnetics the FDT is well-established in terms of fluctuating currents or electric fields. However, in the presence of point dipoles, the most convenient formulation of the FDT is the one that relates the correlation of the fluctuating dipole moment with the dissipative (imaginary) part of its polarizability. Several possible definitions of the polarizability result in different versions of the FDT. This can lead to confusion when one form of the polarizability is used with the environmental correction intended for use with another form. For instance, the expressions for the detector readings in the same setup do not agree in [16] and [17], which was noted but not explained by the authors of the latter paper.

In this review we comprehensively describe the “point dipole” concept in order to carefully resolve all the ambiguities. We aim to give a unified and self-consistent description from all possible points of view in the framework of the frequency-domain electrodynamics. In Section 2, we consider a common abstraction of a singular point in space, whose sole ability is to be polarized due to the external electric field. We show that it can be defined by introducing the source Green’s dyadic $\bar{\mathbf{G}}_{\text{src}}(\mathbf{r}, \mathbf{r}')$ [18] of the special form. It provides a unified description of the contribution of such dipole to the whole space, and can even be measured experimentally. The polarizability is thus empirically defined as a constant parameter in the expression of $\bar{\mathbf{G}}_{\text{src}}(\mathbf{r}, \mathbf{r}')$. All optical properties of the point dipole, such as the absorption and emission powers, can be derived directly from this definition. It eliminates the need for additional, commonly used phenomenological expressions, which are sometimes contradictory, excessive, and even wrong in an inhomogeneous environment. In particular, we rigorously derive the fluctuation–dissipation theorem (FDT) in terms of the fluctuating dipole moment. This solves the long-standing controversy in the literature dealing with radiative heat transfer. Section 3 shows how the same conclusions can be obtained for a small sphere by taking the small-size limit of the Lorenz–Mie theory. In Section 4 we describe a general microscopic theory for particles of arbitrary shape and composition, based on the volume-integral-equation (VIE) formulation [19,20], which includes the definition of polarizability as the double integral of the corresponding transition dyadic (operator). We also review the relation of the polarizability of a small particle with its T-matrix, a widely used entity in light-scattering computations, which follows from the projection of the transition operator on the basis of the vector spherical wave functions (VSWFs). Throughout these sections we review the existing literature within a unified framework, and provide new derivations to fill the gaps. To facilitate the reading, the cumbersome details of these derivations are moved to Appendices A–F.

2. Phenomenological description of a point dipole

2.1. Basic definitions

The concept of a point dipole seems intuitively clear when speaking about absorption and scattering of light by atoms, molecules, and subwavelength particles. Although all these objects do have finite size, it is common to describe them mathematically as a delta-function in space having certain polarizability (and, hence, the dipole moment). This description contains a singularity by design, resulting in unexpected effects when one tries to relate the polarizability with the internal properties, such as the permittivity ϵ . Not surprisingly, the literature contains several definitions of the polarizability. Apart from the one for purely electrostatic problem with zero-frequency ϵ (irrelevant for scattering at frequency ω), these include bare or static, based on $\epsilon(\omega)$ [8], renormalized [15], and radiation-corrected (dressed or dynamic) ones [12,13,21]. But even those terms are not completely universal. For instance, a different definition of bare polarizability is used in [22], the terms “dynamic bare” and “dynamic dressed” are used in [23], and

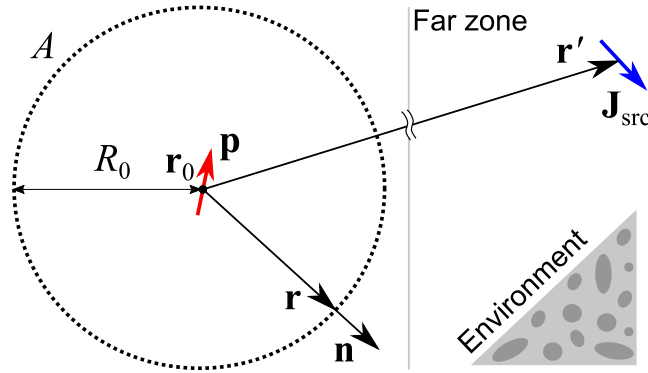


Fig. 1. Layout of the scattering problem for a point dipole.

the term “quasi-static” was used in [24] to indicate the frequency dependence through $\epsilon(\omega)$. The term *static* polarizability is indeed potentially confusing, but since ϵ is just a constant parameter (or a function of \mathbf{r}) in the electrostatic problem, one can take its value at frequency ω as well. Throughout the review we will use the latter convention. However, we defer the discussion of how the internal structure, including ϵ , and the polarizability of a small particle are related to Sections 3 and 4. Meanwhile we treat the polarizability as a prescribed constant.

We limit ourselves to the monochromatic electromagnetic waves with the time dependence $\exp(-i\omega t)$ and to non-magnetic materials, and use the SI units throughout the review. The description is based on the *dyadic Green's function* $\overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}')$, which by definition relates the field $\mathbf{E}(\mathbf{r})$ with the *point current source* \mathbf{J}_{src} placed at \mathbf{r}' [25]:

$$\mathbf{E}(\mathbf{r}) = i\omega\mu_0\overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}') \cdot \mathbf{J}_{\text{src}}. \quad (1)$$

Here and further we denote vectors and dyadics (3×3 matrices) by bold letters without and with overline, respectively; the centered dot denotes the bilinear *dot product* of vectors and/or dyadics (not to be confused with the sesquilinear *inner product* implying conjugation of the second argument — see Appendix E).

Naturally, the Green's dyadic depends on all details of the scattering problem. In vacuum it is a well-known *free-space Green's dyadic* $\overline{\mathbf{G}}_0$ [Eq. (A.1)]. Next we allow for an arbitrary *environment*, including other particles, a substrate, etc., and define $\overline{\mathbf{G}}_{\text{env}}$ as an additional contribution from it:

$$\overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}') = \overline{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}') + \overline{\mathbf{G}}_{\text{env}}(\mathbf{r}, \mathbf{r}'). \quad (2)$$

We further place a point dipole at \mathbf{r}_0 and assume that it is separated from all components of the environment (Fig. 1). Then $\overline{\mathbf{G}}_{\text{env}}(\mathbf{r}, \mathbf{r}')$ is necessarily smooth and finite when \mathbf{r} and/or \mathbf{r}' approach \mathbf{r}_0 , justifying its separation from $\overline{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}')$ in definition (2). Finally, the total Green's dyadic in presence of the dipole (or any scatterer) is called a *source Green's dyadic* $\overline{\mathbf{G}}_{\text{src}}(\mathbf{r}, \mathbf{r}')$ [18]. It is commonly defined with subscript “s” in the literature, as well as \mathbf{J}_{src} , but here we wish to avoid confusion with other subscripts. Note also that $\overline{\mathbf{G}}$ without subscripts denotes the *total Green's dyadic* without a particle, as defined by Eq. (2).

The point dipole with the *induced dipole moment* \mathbf{p} is equivalent to the current $-i\omega\mathbf{p}$ and, thus, produces the *scattered field* $\mathbf{E}_{\text{sca}}(\mathbf{r}) \triangleq \omega^2\mu_0\overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) \cdot \mathbf{p}$ around it [20], cf. Eq. (1). The dipole moment itself is induced by the *incident field* $\mathbf{E}_0(\mathbf{r}) = i\omega\mu_0\overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}') \cdot \mathbf{J}_{\text{src}}(\mathbf{r}')$ due to the source and the environment. A common postulate for the point dipole is

$$\mathbf{p} = \overline{\boldsymbol{\alpha}} \cdot \mathbf{E}_0, \quad (3)$$

where $\mathbf{E}_0 \triangleq \mathbf{E}_0(\mathbf{r}_0)$ and $\overline{\boldsymbol{\alpha}}$ is the (dynamic) *dipole polarizability tensor* (dyadic), discussed below. This postulate implies the following *total field* $\mathbf{E}(\mathbf{r})$:

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) + \mathbf{E}_{\text{sca}}(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) + \omega^2\mu_0\overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) \cdot \mathbf{p}, \quad \mathbf{r} \neq \mathbf{r}_0, \quad (4)$$

which together with Eq. (1) in the presence of the dipole leads to

$$\overline{\mathbf{G}}_{\text{src}}(\mathbf{r}, \mathbf{r}') = \overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}') + \omega^2\mu_0\overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) \cdot \overline{\boldsymbol{\alpha}} \cdot \overline{\mathbf{G}}(\mathbf{r}_0, \mathbf{r}'), \quad \mathbf{r}, \mathbf{r}' \neq \mathbf{r}_0. \quad (5)$$

This is the most complete mathematical description of the fact that the point dipole, placed in arbitrary environment with specified $\overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}')$, affects the electromagnetic properties of space outside its location. Note that $\mathbf{E}(\mathbf{r})$ and, hence, $\overline{\mathbf{G}}_{\text{src}}(\mathbf{r}, \mathbf{r}')$ are potentially measurable quantities, unlike the phenomenological \mathbf{p} and $\overline{\boldsymbol{\alpha}}$. Therefore, in this review we use Eq. (5) as a definition of the phenomenological constant $\overline{\boldsymbol{\alpha}}$. As we show further, this definition is self-sufficient, i.e., all other known properties of the point dipole follow from it. This obviously includes Eq. (3).

Let us now place this definition of the polarizability tensor $\overline{\boldsymbol{\alpha}}$ in existing context. It relates the dipole moment \mathbf{p} with the external fields oscillating with frequency ω and, therefore, implies a solution of a complete electrodynamic problem. Thus, we call this the

dynamic polarizability throughout the paper. It differs from the static polarizability $\bar{\chi}$, which is originally defined for constant electric fields ($\omega = 0$). The most rigorous way to relate the two quantities is

$$\bar{\chi} = \lim_{\omega \rightarrow 0} \bar{\alpha}, \quad (6)$$

where the limit implies $k \rightarrow 0$, but keeps $\mathbf{E}_0(\mathbf{r}_0)$ and all internal details of the dipole fixed. The latter is natural (and the only reasonable option) while the dipole is considered as a phenomenological entity. But if a microscopic model of the dipole is used, as in Sections 3 and 4, it also implies that ε is fixed at its value for the frequency of the original electrodynamic problem. This justifies the name “static polarizability” for $\bar{\chi}$, as discussed in the beginning of this section. Applying Eq. (6) to Eq. (4) and using Eq. (A.2), we obtain

$$\mathbf{E}_{\text{st}}(\mathbf{r}) = \mathbf{E}_{0,\text{st}}(\mathbf{r}) + \omega^2 \mu_0 \bar{\mathbf{G}}_{\text{st}}(\mathbf{r}, \mathbf{r}_0) \cdot \bar{\chi} \cdot \mathbf{E}_0, \quad (7)$$

where the subscript “st” refers to the static limit and $\bar{\mathbf{G}}_{\text{st}}(\mathbf{r}, \mathbf{r}_0)$ is defined by Eq. (A.2). The latter implicitly assumes that the static limit of $\bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0)$ is independent of the environment, which explains the omission of the additional subscript 0 in $\bar{\mathbf{G}}_{\text{st}}$. This independence follows from the smoothness of $\bar{\mathbf{G}}_{\text{env}}(\mathbf{r}, \mathbf{r}_0)$ in Eq. (2) — its static limit is negligible in comparison with Eq. (A.2). Similarly, Eq. (7) suggests that $\bar{\chi}$ is independent of the environment, which is further discussed in the following sections. Being general, Eq. (6) is instrumental if one has an expression (a model) for $\bar{\alpha}$, but not if $\bar{\alpha}$ is just a phenomenological constant. In particular, we are not aware how to use Eq. (7) to derive a relation between $\bar{\alpha}$ and $\bar{\chi}$, since it is not a priori clear what is the difference between $\mathbf{E}(\mathbf{r})$ and $\mathbf{E}_{\text{st}}(\mathbf{r})$.

The first approach to actually derive such a relation is to postulate that $\bar{\chi}$ acts on the *exciting field*, which in addition to the incident \mathbf{E}_0 includes the frequency-dependent *self-field*. The latter can be written as $\omega^2 \mu_0 \bar{\mathbf{G}}_{\text{r}}(\mathbf{r}_0, \mathbf{r}_0) \cdot \mathbf{p}$, where $\bar{\mathbf{G}}_{\text{r}}(\mathbf{r}, \mathbf{r}')$ is the *regularized Green’s dyadic* (i.e., finite when $\mathbf{r} \rightarrow \mathbf{r}'$) [26], leading to:

$$\mathbf{p} = \bar{\chi} \cdot \left[\mathbf{E}_0 + \omega^2 \mu_0 \bar{\mathbf{G}}_{\text{r}}(\mathbf{r}_0, \mathbf{r}_0) \cdot \mathbf{p} \right]. \quad (8)$$

Before discussing specific regularizations, let us define a decomposition of an arbitrary dyadic function:

$$\begin{aligned} \bar{\mathbf{A}}(\mathbf{r}, \mathbf{r}') &= \bar{\mathbf{A}}^R(\mathbf{r}, \mathbf{r}') + i\bar{\mathbf{A}}^I(\mathbf{r}, \mathbf{r}'), \\ \bar{\mathbf{A}}^R(\mathbf{r}, \mathbf{r}') &\stackrel{\text{def}}{=} \frac{1}{2} \left\{ \bar{\mathbf{A}}(\mathbf{r}, \mathbf{r}') + \left[\bar{\mathbf{A}}(\mathbf{r}', \mathbf{r}) \right]^H \right\}, \quad \bar{\mathbf{A}}^I(\mathbf{r}, \mathbf{r}') \stackrel{\text{def}}{=} \frac{1}{2i} \left\{ \bar{\mathbf{A}}(\mathbf{r}, \mathbf{r}') - \left[\bar{\mathbf{A}}(\mathbf{r}', \mathbf{r}) \right]^H \right\}, \end{aligned} \quad (9)$$

where H denotes the Hermitian transpose of a dyadic and its position in Eq. (9) emphasizes that it is applied to a dyadic at fixed coordinates in contrast to R and I which are applied to a dyadic function (operator) before considering specific coordinates. The same notation (R and I) applies to operators and coordinate-independent dyadics; all together this can be considered as a generalization of the real and imaginary parts of a complex number $z = \text{Re } z + i \text{Im } z$, respectively. See Appendix E for more details.

The most common approach for regularization of the Green’s dyadic is to isolate the skew-Hermitian part of $\bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}')$, or simply $i \text{Im } \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}')$, which is finite [15]. In terms of the above definitions, this can be expressed as:

$$\bar{\mathbf{G}}_{\text{r}}(\mathbf{r}, \mathbf{r}') \stackrel{\text{def}}{=} i\bar{\mathbf{G}}^I(\mathbf{r}, \mathbf{r}'), \quad (10)$$

We stick to this definition in this review, however, it should be considered as an assumption, since such regularization is inherently ambiguous [26–28], which is also discussed in several places below. Together with Eqs. (4), (8) this definition of $\bar{\mathbf{G}}_{\text{r}}(\mathbf{r}, \mathbf{r}')$ leads to:

$$\bar{\alpha}^{-1} = \bar{\chi}^{-1} - i\omega^2 \mu_0 \bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0), \quad (11)$$

evidently satisfying Eq. (6). Throughout this review we assume that both polarizabilities $\bar{\alpha}$ and $\bar{\chi}$ are non-singular (i.e., invertible) dyadics, since this allows one to obtain several simple expressions, such as Eq. (11), which are widely used in the literature. However, most formulas and conclusions stay valid even for singular polarizabilities. Finally, in vacuum

$$\bar{\mathbf{G}}_0^I(\mathbf{r}_0, \mathbf{r}_0) = \frac{k\mathbf{I}}{6\pi} \quad (12)$$

due to Eqs. (A.3), (E.8). While this regularization of the free-space Green’s dyadic is natural for many reasons (see Section 2.2), consideration of the environment immediately creates ambiguity. One may wonder if the Hermitian part of the environmental contribution, i.e., $\bar{\mathbf{G}}_{\text{env}}^R(\mathbf{r}_0, \mathbf{r}_0)$, should also be included in $\bar{\mathbf{G}}_{\text{r}}(\mathbf{r}_0, \mathbf{r}_0)$. However, as we show below, this leads to asymptotically negligible differences in any measurable quantities, i.e., only the I -part of $\bar{\mathbf{G}}_{\text{r}}(\mathbf{r}_0, \mathbf{r}_0)$ matters.

The second approach to relate $\bar{\alpha}$ and $\bar{\chi}$ is based on the fact that the main difference between a static and an oscillating dipole is the radiation, which transfers energy to infinity. Therefore, the relation can be derived from energy conservation. We pursue this line of thought in the next section and show that the final result is asymptotically equivalent to that based on the self-field.

2.2. Extinction, absorption, and scattering

Absorption and scattering by a point dipole are important from the experimental point of view. For instance, the amount of absorbed energy is vital for such applications as the molecular light conversion systems [29,30], solar cells [31], nanoparticles-based photothermal effect [32] and many others. In this respect, scattering does not only play a role as an energy sink, but is also a basis for far-field observations. The expressions for the absorption and scattering powers of the point dipole seem well-known and trivial, but there are three important caveats.

First, the expressions are often postulated (without derivation), which can potentially lead to misuse. Second, the use of expression $\text{Im}(\mathbf{E}_0 \cdot \mathbf{p}^*)$ for absorption is justified (in free space) only if the scattered power can be neglected (* denotes complex conjugation). The scattered power is proportional to the square of the particle volume (while absorption — to the first power), but this term cannot be neglected if the dipole is non-absorbing. Third, many applications contain a non-homogeneous environment, such as a plane substrate. There is no obvious way to transform formulas if the environment is not used explicitly in the derivation. However, absorption is an internal process and therefore should depend on the local field only. Indeed, as shown in [15], the corresponding power can be expressed through the static polarizability as follows:

$$W_{\text{abs}} = -\frac{\omega}{2} \text{Im}(\mathbf{p}^* \cdot \overline{\chi}^{-1} \cdot \mathbf{p}) = -\frac{\omega}{2} \mathbf{p}^* \cdot (\overline{\chi}^{-1})^I \cdot \mathbf{p}, \quad (13)$$

where we used Eq. (E.7).

By contrast, the expression involving $\overline{\alpha}$ would explicitly contain the environment. To show it, let us calculate the power absorbed by the dipole W_{abs} starting directly from Eq. (5) and integrating the time-averaged Poynting vector $\mathbf{S}(\mathbf{r})$ over a small spherical surface A (with the radius R_0) around the dipole [20]:

$$W_{\text{abs}} \stackrel{\text{def}}{=} - \lim_{R_0 \rightarrow 0} \oint_A d\mathbf{A} \cdot \mathbf{S}(\mathbf{r}), \quad (14)$$

where $d\mathbf{A} \stackrel{\text{def}}{=} \mathbf{n} d^2\mathbf{r}$, $\mathbf{n} \stackrel{\text{def}}{=} \mathbf{R}_0/R_0$ is the normal to the surface, $\mathbf{R}_0 \stackrel{\text{def}}{=} \mathbf{r} - \mathbf{r}_0$ (Fig. 1). In taking the limit $R_0 \rightarrow 0$ we assume that $R_0 \gg a$, where a is the internal size of the dipole. Using the standard decomposition of $\mathbf{S}(\mathbf{r})$ [Eq. (B.2)], we obtain the extinction and scattered powers:

$$W_{\text{abs}} = W_{\text{ext}} - W_{\text{sca}}, \quad W_{\text{ext}} \stackrel{\text{def}}{=} - \lim_{R_0 \rightarrow 0} \oint_A d\mathbf{A} \cdot \mathbf{S}_{\text{ext}}(\mathbf{r}), \quad W_{\text{sca}} \stackrel{\text{def}}{=} \lim_{R_0 \rightarrow 0} \oint_A d\mathbf{A} \cdot \mathbf{S}_{\text{sca}}(\mathbf{r}), \quad (15)$$

since the flow of $\mathbf{S}_0(\mathbf{r})$ over any closed surface not containing its sources is zero.

As shown in Appendix B, Eq. (15) can be rigorously evaluated to yield [Eq. (B.5)]:

$$W_{\text{ext}} = -\frac{\omega}{2} \text{Im}(\mathbf{E}_0 \cdot \mathbf{p}^*) = \frac{\omega}{2} \mathbf{E}_0^* \cdot \overline{\alpha}^I \cdot \mathbf{E}_0, \quad (16)$$

where we used Eq. (E.7). Note that $\overline{\alpha}^I = \text{Im} \overline{\alpha}$ for any symmetric polarizability [Eq. (E.9)], as is commonly the case. The scattered power is [Eq. (B.10)]:

$$W_{\text{sca}} = \frac{\omega^3 \mu_0}{2} \mathbf{p}^* \cdot \overline{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \mathbf{p} = \frac{\omega^3 \mu_0}{2} \mathbf{E}_0^* \cdot \overline{\alpha}^H \cdot \overline{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \overline{\alpha} \cdot \mathbf{E}_0. \quad (17)$$

Finally, the absorption power is found from Eqs. (15)–(17):

$$W_{\text{abs}} = \frac{\omega}{2} \mathbf{E}_0^* \cdot \overline{\beta} \cdot \mathbf{E}_0, \quad (18)$$

where we defined

$$\overline{\beta} \stackrel{\text{def}}{=} \overline{\alpha}^I - \omega^2 \mu_0 \overline{\alpha}^H \cdot \overline{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \overline{\alpha}, \quad (19)$$

which is always Hermitian (as it should be for real W_{abs}). Comparison of Eqs. (13), (18) leads to $\overline{\beta} = \overline{\alpha}^H \cdot (\overline{\chi}^{-1})^I \cdot \overline{\alpha}$, which after substitution into Eq. (19) leads to the relation between $\overline{\alpha}$ and $\overline{\chi}$:

$$(\overline{\alpha}^{-1})^I = (\overline{\chi}^{-1})^I - \omega^2 \mu_0 \overline{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0), \quad (20)$$

where we used Eq. (E.12).

We stress that Eq. (20) follows from but does not imply Eq. (11). In other words, using reasonable albeit still phenomenological Eq. (13) (the energy-conservation arguments) we obtained the relation between the I -parts of $\overline{\alpha}$ and $\overline{\chi}$, and thus justified the choice of the I -part of the regularized Green's dyadic, i.e., $\overline{\mathbf{G}}_r^I(\mathbf{r}_0, \mathbf{r}_0) = \overline{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0)$. There are, however, no rigorous arguments for the choice of the corresponding Hermitian part $\overline{\mathbf{G}}_r^R(\mathbf{r}_0, \mathbf{r}_0)$, except that it should be relatively small to satisfy Eq. (6). In particular, adding any Hermitian dyadic with a finite limit as $\omega \rightarrow 0$, e.g., $\overline{\mathbf{G}}_{\text{env}}^R(\mathbf{r}_0, \mathbf{r}_0)$, to $\overline{\mathbf{G}}_r^R(\mathbf{r}_0, \mathbf{r}_0)$ will keep the whole phenomenological theory self-consistent. This is not surprising, given that the self-field is not a measurable, but rather an auxiliary quantity. It is chosen such that the work rate done by the self-field on the dipole equals the radiated power [10]. Since this work rate is expressed through $\overline{\mathbf{G}}_r^I(\mathbf{r}_0, \mathbf{r}_0)$, only the latter can be reasonably constrained.

On the one hand, the only honest reason to postulate Eqs. (10), (11) (as we did in this review) is to choose zero as the simplest options of an infinite range or, equivalently, due to a symmetric truncation in some postulated basis, e.g., the Fourier basis in [26] or the VSWF basis, discussed after Eq. (D.15). While others have argued that there is a microscopic justification for that [11,33,34], we further show in Sections 3 and 4 that the microscopic analysis adds rigor to Eqs. (5) and (13), but only deepens the ambiguity with respect to $\overline{\mathbf{G}}_r^R(\mathbf{r}_0, \mathbf{r}_0)$. Note that *microscopic* denotes that some finite particle is used as a dipole model in contrast to the purely *phenomenological* approach of this section. However, in both cases we stay within the realms of the macroscopic electrodynamics, where the properties of materials are given by a permittivity. This is similar to the DDA, which solves the macroscopic VIE [14], but the size of the corresponding dipoles (discretization voxels) may well be smaller than atoms [35].

On the other hand, the dynamic (radiation-corrected) polarizability $\overline{\alpha}$ is successfully used in various applications, for instance, the surface-enhanced Raman scattering [36], plasmonics [37,38], and the DDA [14]. Moreover, in many applications the difference between $\overline{\alpha}$ and $\overline{\chi}$ is not significant at all, since the notion of a point dipole itself implies that a is negligibly small. Hence, its

polarizability is also small, specifically $k^3 \|\bar{\chi}\|/\varepsilon_0 \ll 1$, where $\|\dots\|$ denotes the norm of the dyadic (e.g., the L^2 one). Thus, the term with $\bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0)$ in Eq. (11) is also much smaller than other terms. However, that is not necessarily the case for Eq. (20), especially if $\|\bar{\chi}'\| \ll \|\bar{\chi}\|$. In the extreme case of $\bar{\chi}' = \bar{\mathbf{0}}$ (non-absorbing dipole), setting $\bar{\alpha} \approx \bar{\chi}$ will lead to negative W_{abs} in Eq. (18), while using Eq. (20) with all terms will correctly predict $W_{\text{abs}} = 0$. This brings us to explicit formulation of the point-dipole limit that we use throughout this review. We require the derived formulas for all quantities of interest to be asymptotically correct (i.e., to correctly describe the smallest order of smallness, e.g., in powers of a) *uniformly for any level of absorption*. In the last example above interchanging $\bar{\alpha}$ and $\bar{\chi}$ keeps W_{sca} , $\bar{\chi}^R$, and $\|\bar{\chi}\|$ asymptotically correct, but not W_{abs} , W_{ext} , and $\bar{\chi}'$.

We will further use \cong to denote the asymptotical equivalence under such uniform limit in contrast to the non-uniform approximation \approx . For instance, $\bar{\alpha} \approx \bar{\chi}$, but all values of $\bar{\alpha}$ obtained by introducing non-zero $\bar{\mathbf{G}}_r^R(\mathbf{r}_0, \mathbf{r}_0)$ are asymptotically equivalent, which alleviates the above discussed ambiguity. The use of \cong is much more specific than general arguments of energy conservation. While the latter is surely desirable for both theories and numerical methods, it is neither necessary nor sufficient for accurate simulations, as discussed with respect to the DDA in [20].

Finally, Eqs. (19), (20) lead to

$$\bar{\beta} = \left[\bar{\mathbf{I}} - \omega^2 \mu_0 \bar{\mathbf{G}}_r(\mathbf{r}_0, \mathbf{r}_0) \cdot \bar{\chi} \right]^{-H} \cdot \bar{\chi}' \cdot \left[\bar{\mathbf{I}} - \omega^2 \mu_0 \bar{\mathbf{G}}_r(\mathbf{r}_0, \mathbf{r}_0) \cdot \bar{\chi} \right]^{-1} \cong \bar{\chi}', \quad (21)$$

since the small corrections are applied multiplicatively to $\bar{\chi}'$ rather than additively. This means that instead of Eq. (13) one can use

$$W_{\text{abs}} \cong \frac{\omega}{2} \mathbf{E}_0^* \cdot \bar{\chi}' \cdot \mathbf{E}_0 \quad (22)$$

as a defining property of $\bar{\chi}$, and there are no compelling reasons to choose between the two. To reiterate, while we stay in the framework of the phenomenological theory, the most fundamental property is $\bar{\alpha}$, as a phenomenological constant in Eq. (5). It alone is sufficient to define any measurable quantity (including the FDT in Section 2.3). By contrast, $\bar{\chi}$ is an inherently microscopic quantity, for which many asymptotically equivalent definitions are possible.

2.3. Fluctuation–dissipation theorem

Thermal emission is of great importance for many experimental techniques and applications. Near-field radiative heat transfer is significant for thermal management of nanoscale devices, imaging, nanomanufacturing, thermal rectification, near-field thermal spectroscopy, and thermophotovoltaic power generation (see [39,40] and references therein). While the fluctuations (their correlation) are governed by quantum equations, thermal emission can be treated quasi-classically using the standard electromagnetics complemented by the FDT [40].

The FDT relates the rate of energy dissipation in a non-equilibrium system to the fluctuations that occur spontaneously at equilibrium [3]. In electromagnetics the FDT is well-established in terms of fluctuating currents or electric fields (see below). However, in the presence of point dipoles the most convenient formulation of the FDT is the one that relates the correlation of the fluctuating dipole moment with the imaginary part of its polarizability. But, as discussed in Section 2.2, the latter has several possible definitions, resulting in different versions of the FDT. This can lead to confusion when one polarizability is used with the environmental correction intended for another one. For instance, the expressions for the detector readings in the same setup do not agree in [16] and [17]. The difference stems from the fact that in [16] the authors used $\bar{\alpha}$ in the FDT formulated for $\bar{\chi}$. The same incorrect formulation of the FDT can be found in [41].

In this section, we derive the FDT for the fluctuating dipole moment straight from Eq. (5). For that we consider the equilibrium of the dipole with the environment (thermal bath) at temperature T without any other external sources. If the dipole is not present, the FDT for the fluctuating fields of the bath \mathbf{E}_b is well-known [42]:

$$\langle \mathbf{E}_b(\mathbf{r}) \otimes \mathbf{E}_b^*(\mathbf{r}') \rangle = \frac{\omega \mu_0}{\pi} \Theta(\omega, T) \bar{\mathbf{G}}^I(\mathbf{r}, \mathbf{r}'), \quad (23)$$

where $\langle \dots \rangle$ denotes time averaging over a period much larger than $1/\omega$, $\bar{\mathbf{G}}^I(\mathbf{r}, \mathbf{r}')$ is used instead of commonly used $\text{Im} \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}')$ according to [43],

$$\Theta(\omega, T) \stackrel{\text{def}}{=} \frac{\hbar \omega}{e^{\hbar \omega / k_B T} - 1} \quad (24)$$

is the mean energy of the quantum harmonic oscillator without the zero-point energy term, \hbar is the reduced Planck constant, and k_B is the Boltzmann constant. The only potential ambiguity is related to the specific expression for $\Theta(\omega, T)$, including the zero-point energy term. This issue is well discussed and referenced in Section 3.4 of [44], justifying the choice of Eq. (24).

When the dipole is placed in the thermal bath, there are both the induced dipole moment \mathbf{p} due to \mathbf{E}_b and the fluctuating one \mathbf{p}_{fl} . Before we derive a general result, let us use a simpler heuristic approach based on energy conservation. Substituting Eq. (23) into Eq. (18), we obtain the power absorbed by the dipole from the thermal bath:

$$W_{\text{abs}} = \frac{\omega}{2} \mathbf{E}_b^*(\mathbf{r}_0) \cdot \bar{\beta} \cdot \mathbf{E}_b(\mathbf{r}_0) = \frac{\omega}{2} \text{Tr} \left[\langle \mathbf{E}_b(\mathbf{r}_0) \otimes \mathbf{E}_b^*(\mathbf{r}_0) \rangle \cdot \bar{\beta} \right] = \frac{\omega^2 \mu_0}{2\pi} \Theta(\omega, T) \text{Tr} \left[\bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \bar{\beta} \right], \quad (25)$$

where Tr denotes the trace of a dyadic. The power emitted by the fluctuating dipole is [Eq. (17)]:

$$W_{\text{em}} = \frac{\omega^3 \mu_0}{2} \text{Tr} \left[\bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \langle \mathbf{p}_{\text{fl}} \otimes \mathbf{p}_{\text{fl}}^* \rangle \right]. \quad (26)$$

Next, we postulate that the spectral power density of absorption (of the fluctuating fields) is equal to those of emission (by the fluctuating dipole), since their temperatures are equal (corresponding to the thermal equilibrium); then:

$$\text{Tr}[\overline{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \langle \mathbf{p}_{f1} \otimes \mathbf{p}_{f1}^* \rangle] = \frac{\Theta(\omega, T)}{\pi\omega} \text{Tr}[\overline{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \overline{\boldsymbol{\beta}}]. \quad (27)$$

This (besides Tr and $\overline{\mathbf{G}}^I$) resembles the FDT for the point dipole. In particular, the need to use $\overline{\boldsymbol{\beta}}$ on the right-hand side of the FDT was pointed out earlier for a scalar polarizability [45,46].

For a rigorous derivation of the FDT we need to apply Eq. (23) to the total field \mathbf{E} , using the source Green's dyadic (5) on the right hand side (this approach was mentioned in Appendix A of [46]):

$$\langle \mathbf{E}(\mathbf{r}) \otimes \mathbf{E}^*(\mathbf{r}') \rangle = \frac{\omega\mu_0}{\pi} \Theta(\omega, T) \overline{\mathbf{G}}_{\text{src}}^I(\mathbf{r}, \mathbf{r}'). \quad (28)$$

The total field is made up of the field of the thermal bath \mathbf{E}_b , the scattered field \mathbf{E}_{sca} (due to \mathbf{E}_b) and the field emitted by the fluctuating dipole $\mathbf{E}_{\text{em}}(\mathbf{r})$. Combining it with Eq. (23) and using the fact that the fluctuations of the dipole and thermal bath (\mathbf{p}_{f1} and \mathbf{E}_b) are statistically independent, we obtain (see Appendix C for details):

$$\langle \mathbf{p}_{f1} \otimes \mathbf{p}_{f1}^* \rangle = \frac{\Theta(\omega, T)}{\pi\omega} \overline{\boldsymbol{\beta}}, \quad (29)$$

which is the usual formulation of the FDT for the point dipole. It is now clear that the difference between [16] and [17] was due to the use of $\overline{\boldsymbol{\alpha}}^I$ instead of $\overline{\boldsymbol{\beta}}$, which may lead to huge errors for small absorption, as discussed in the end of Section 2.2. By contrast, Eq. (21) is asymptotically correct for any absorption, thus

$$\langle \mathbf{p}_{f1} \otimes \mathbf{p}_{f1}^* \rangle \cong \frac{\Theta(\omega, T)}{\pi\omega} \overline{\boldsymbol{\chi}}^I. \quad (30)$$

The scalar equivalent of Eq. (30) was used, for instance, in [47] and in the thermal discrete-dipole approximation (T-DDA, [21]) assuming the Clausius–Mossotti polarizability for each dipole. The equivalence of these two formulations of the FDT was recently discussed in [22]. However, the authors stated that the factor $\boldsymbol{\beta}$ in Eq. (29) is not inherent to the FDT but arises because the definition of dipole moment does not account for the self-field. We disagree with this interpretation since, as discussed in Section 2.2, it is $\overline{\boldsymbol{\alpha}}$ (and, hence, $\overline{\boldsymbol{\beta}}$) which is the most fundamental quantity for this phenomenological scattering problem. Therefore, Eq. (29) is the primary result, while Eq. (30) is either a good approximation or even exactly equivalent to Eq. (29), depending on the specific definition of $\overline{\boldsymbol{\chi}}$.

3. Small-sphere limit

3.1. Polarizability of a small sphere

In Section 2 we noted multiple times that the lack of microscopic description leads to ambiguities, in particular, in the definition of the static polarizability $\overline{\boldsymbol{\chi}}$. Here we address this issue by considering a small sphere with the radius a , the volume V , and the permittivity ϵ and limit ourselves to scalar ϵ and particles located in free space, in order to use the standard Lorenz–Mie theory. Let us also denote the size parameter as $x \stackrel{\text{def}}{=} ka$ and the refractive index as $m_0 \stackrel{\text{def}}{=} \sqrt{\epsilon/\epsilon_0}$. Such model for a point dipole is, obviously, the simplest one and the one used in the vast majority of papers, which consider any microscopic model at all. It is also worth noting that all the formulas of this section can be extended to the case of a sphere in a homogeneous non-absorbing host medium with the permittivity ϵ_h through simple scaling [48]. In particular, ϵ_0 should be replaced by ϵ_h and k should be considered as the wavenumber in the host medium (multiplied by $\sqrt{\epsilon_h}$). Then the definitions of x and m_0 remain as given above.

In this microscopic theory the roles of $\overline{\boldsymbol{\chi}}$ and $\overline{\boldsymbol{\alpha}}$ are reversed. The static polarizability is unambiguously given by the Clausius–Mossotti formula [15]:

$$\overline{\boldsymbol{\chi}} = 3V\epsilon_0 \frac{\epsilon - \epsilon_0}{\epsilon + 2\epsilon_0} \overline{\mathbf{I}}, \quad (31)$$

where $\epsilon(\omega)$ is assumed, as discussed in Section 2.1 (see also p. 139 of [8]). By contrast, the definition of $\overline{\boldsymbol{\alpha}}$ is potentially ambiguous. It is common to express it through the first Lorenz–Mie coefficient a_1 , based on the similarity of the corresponding far-field partial scattering amplitudes to that of the point dipole [15]:

$$\overline{\boldsymbol{\alpha}} \cong \frac{6\pi i \epsilon_0}{k^3} a_1 \overline{\mathbf{I}}. \quad (32)$$

The asymptotic equivalence (\cong) has been discussed in Section 2.2, here it can be considered as a limit $x \rightarrow 0$. It is used because a_1 describes the dipolar response to the fields proportional to the first three VSWFs (see Appendix D), which are constant only in this limit.

Alternatively, one can compute the dipole moment of a small sphere by directly integrating the internal polarization $\mathbf{P}(\mathbf{r}) \stackrel{\text{def}}{=} (\epsilon - \epsilon_0)\mathbf{E}(\mathbf{r})$ [see Eq. (53) below] over the sphere volume, where $\mathbf{E}(\mathbf{r})$ – the field inside a sphere – is a superposition of the regular VSWFs with argument $m_0 k \mathbf{r}$ [8]. As shown in Appendix D [Eq. (D.6)], all these VSWFs except RgN_{1m} integrate to zero, while the expansion coefficients of $\mathbf{E}(\mathbf{r})$ before $\text{RgN}_{1m}(m_0 k \mathbf{r})$ are the same as for $\mathbf{E}_0(\mathbf{r})$ before $\text{RgN}_{1m}(k \mathbf{r})$, but multiplied by the Lorenz–Mie coefficient d_1 . Accounting for deviation of $\text{RgN}_{1m}(m_0 k \mathbf{r})$ from a constant [Eq. (D.11)], we obtain

$$\overline{\boldsymbol{\alpha}} \cong 3 \frac{j_1(m_0 x)}{m_0 x} V(\epsilon - \epsilon_0) d_1 \overline{\mathbf{I}}. \quad (33)$$

Although both Eqs. (32) and (33) make sense for arbitrary sizes, giving dipolar component of far-field response and the total dipole moment, respectively, the whole dipole approximation is valid only for a small sphere and if [49]

$$m_0 x \ll 1. \quad (34)$$

Let us consider this limit in details, starting with a_1 [8]:

$$a_1 = -i \frac{2x^3}{3} \frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} - i \frac{2x^5}{5} \frac{(\varepsilon - \varepsilon_0)(\varepsilon - 2\varepsilon_0)}{(\varepsilon + 2\varepsilon_0)^2} + \left(\frac{2x^3}{3} \frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} \right)^2 + \mathcal{O}(x^7). \quad (35)$$

Taking a straightforward limit $x \rightarrow 0$ leaves only the first term in Eq. (35) and, hence, leads to $\bar{\alpha} \approx \bar{\chi}$, which is not satisfactory. Instead, the limiting value should lead to both the real and imaginary parts of a_1 (and $\bar{\alpha}$) being asymptotically correct, even for real ε (see Section 2.2). This leaves the first and the third term in Eq. (35), leading to

$$\bar{\alpha} \cong 3V\varepsilon_0 \frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} \left(1 + i \frac{2x^3}{3} \frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} \right) \bar{\mathbf{I}} = \bar{\chi} \cdot \left[\bar{\mathbf{I}} + i\omega^2 \mu_0 \bar{\mathbf{G}}_0^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \bar{\chi} \right] \cong \bar{\chi} \cdot \left[\bar{\mathbf{I}} - i\omega^2 \mu_0 \bar{\mathbf{G}}_0^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \bar{\chi} \right]^{-1} \quad (36)$$

where the last transformation uses $k^3 \|\bar{\chi}\|/\varepsilon_0 \ll 1$ (follows from Eq. (34) and $x \ll 1$) and makes the result equivalent to Eq. (11).

It is often argued that accounting for the second term in Eq. (35) improves the accuracy of the dipole approximation (so-called, *non-radiative correction* [15,50,51]). Let us critically discuss it. On the one hand, the left-out Lorenz–Mie coefficients b_1 and a_2 are generally of the same order of magnitude as the non-radiative correction, since they are all proportional to x^5 . And the scattered fields corresponding to these coefficients (magnetic dipole and electric quadrupole, respectively) cannot, in principle, be described in the framework of the point-dipole approximation. On the other hand, the dependence of various Lorenz–Mie coefficients on ε is markedly different. In particular, in the case of dipole plasmon resonance (e.g., in metallic nanoparticles) ε is close to $-2\varepsilon_0$ amplifying all parts of a_1 relative to the other Lorenz–Mie coefficients. In this case the non-radiative correction to dipole polarizability becomes the dominant part of the $\mathcal{O}(x^5)$ correction. However, there is an ambiguity in this non-radiative term itself. Starting from the expression for d_1 [8,49], one can easily obtain

$$d_1 = \frac{3\varepsilon_0}{\varepsilon + 2\varepsilon_0} + \frac{3x^2}{10} \frac{(\varepsilon - \varepsilon_0)(\varepsilon + 10\varepsilon_0)}{(\varepsilon + 2\varepsilon_0)^2} + 2ix^3 \frac{\varepsilon_0(\varepsilon - \varepsilon_0)}{(\varepsilon + 2\varepsilon_0)^2} + \mathcal{O}(x^4), \quad (37)$$

where the first and the third terms exactly correspond to that in Eq. (35), if multiplied by $-2ix^3(\varepsilon - \varepsilon_0)/(9\varepsilon_0)$. This coefficient follows from comparing Eqs. (32) and (33). However, the non-radiative term in Eq. (37) is different from that in a_1 , even qualitatively. In particular, it is proportional to a larger power of $(\varepsilon - \varepsilon_0)$ after multiplication by the above coefficient.

Accounting for factor $3j_1(m_0x)/(m_0x)$ [Eq. (33)], modifies only the non-radiative term of d_1 into

$$\frac{3x^2}{10} \frac{\varepsilon_0(7\varepsilon - 10\varepsilon_0)}{(\varepsilon + 2\varepsilon_0)^2}, \quad (38)$$

which becomes qualitatively similar to that in a_1 , but still different. At this point one may wonder why the approaches based on a_1 and d_1 differ in the non-radiative term, even after applying correction to the latter. To answer this question, let us recall that the far-field quantity a_1 is related to $\bar{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}')$, which maps the regular VSWFs in the particle into the outgoing VSWFs. But since $\bar{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}')$ is unaware of a specific particle, both these VSWFs are in free space, see Eq. (D.13) and more detailed discussion in Section 4.4. Therefore, instead of projecting $\mathbf{P}(\mathbf{r})$ on a constant basis, as in Eq. (33), one may project it onto $\text{RgN}_{1m}(k\mathbf{r})$, the same basis as used for $\mathbf{E}_0(\mathbf{r})$. This can be done in the closed form [Eq. (D.8)], which together with the factor 6π due to normalization of RgN_{1m} [Eq. (D.10)] leads exactly to Eq. (32).

To conclude, the microscopic description asymptotically justifies Eq. (11), and also suggests three different next-order corrections. The one corresponding to d_1 [Eq. (37)] can be directly associated with the self-action of the dipole, while the other two provide corrections based on the non-constancy of the VSWFs, either when they are projected on a constant basis [Eq. (33)] or on the free-space VSWFs [Eq. (32)]. These options were also discussed in [52], both for spheres and spheroids. All of them correspond to the additional self-field, a regularized Green's dyadic, which is most meaningful for pure d_1 :

$$\bar{\mathbf{G}}_{r1}(\mathbf{r}_0, \mathbf{r}_0) \stackrel{\text{def}}{=} \frac{1}{40\pi a} \left(\frac{\varepsilon}{\varepsilon_0} + 10 \right) \bar{\mathbf{I}}. \quad (39)$$

For other two options, $\bar{\mathbf{G}}_{r1}$ would be inversely proportional to $(\varepsilon - \varepsilon_0)$, which seems artificial. However, even Eq. (39) has a couple of caveats. First, it is not finite when $a \rightarrow 0$, thus it is not really a regularized part but rather a weakly-singular one, i.e., $1/R$ term in Eq. (A.3), somehow averaged over the particle volume (further discussed in Section 4.2). Second, it does not provide a complete next-order correction for the scattering quantities. Therefore, the microscopic description does not resolve the ambiguity with respect to $\bar{\mathbf{G}}_r^R(\mathbf{r}_0, \mathbf{r}_0)$ or, more generally, to any asymptotically negligible correction to the self-field.

We finish this section by considering the limiting values of scattering and absorption efficiencies [8]:

$$Q_{\text{abs}} = 4x \text{Im} \left(\frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} \right) [1 + \mathcal{O}(x^2)] \cong 4x \text{Im} \left(\frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} \right), \quad (40)$$

$$Q_{\text{sca}} = \frac{8}{3} x^4 \left| \frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} \right|^2 + \mathcal{O}(x^6) \cong \frac{8}{3} x^4 \left| \frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} \right|^2, \quad (41)$$

which are related to the corresponding powers as

$$W = \frac{c\varepsilon_0}{2} \pi a^2 Q. \quad (42)$$

Importantly, Eqs. (40) and (41) provide a one-term asymptotic approximation (uniform for any ε), while the expression for $Q_{\text{ext}} = Q_{\text{abs}} + Q_{\text{sca}}$ must contain both these terms. And all these expressions are consistent with Eqs. (16), (17), and (18) if one uses Eqs. (21) and (36). One may try to use the specific expression for $\mathcal{O}(x^3)$ term in Eq. (40) [8] in order to justify the choice of either Eq. (13) or Eq. (18) over the other. But that is futile, since the latter two include only the influence of a_1 or d_1 , while the $\mathcal{O}(x^3)$ term in Q_{abs} also includes contributions from b_1 and a_2 . Finally, the radiative corrections can also be applied to multipolar polarizabilities of a sphere [11,53], but we discuss it in a more general setting in Section 4.4.

3.2. Fluctuation–dissipation theorem for a small sphere

In contrast to a point dipole, thermal emission of a sphere can be expressed through the fluctuations of its internal currents $\mathbf{J}_{\text{fl}}(\mathbf{r})$. In this case, we can start from the corresponding FDT for currents [3,40]:

$$[\mathbf{J}_{\text{fl}}(\mathbf{r}) \otimes \mathbf{J}_{\text{fl}}^*(\mathbf{r}')] = \frac{\omega}{\pi} \Theta(\omega, T) \text{Im}(\varepsilon) \delta(\mathbf{r} - \mathbf{r}') \bar{\mathbf{I}}. \quad (43)$$

Our goal here is to obtain the FDT in terms of the sphere's dipole moment. For simplicity, in this section we consider only a small sphere ($x \rightarrow 0$) – that is, we do not account for any corrections except the uniform limiting values (the largest terms for the real and imaginary parts). In particular, we ignore the second terms in the Lorenz–Mie coefficients a_1, d_1 [Eqs. (35), (37)] and the factor $3j_1(m_0x)/(m_0x)$. The dipole moment of the sphere is caused by fluctuating currents directly and by the induced electric fields:

$$\mathbf{p}_{\text{fl}} = \frac{i}{\omega} \int_V d^3\mathbf{r} \mathbf{J}_{\text{fl}}(\mathbf{r}) + (\varepsilon - \varepsilon_0) \int_V d^3\mathbf{r} \mathbf{E}(\mathbf{r}), \quad (44)$$

where $\mathbf{E}(\mathbf{r})$ is the internal field. In principle, this field can be expressed through \mathbf{J}_{fl} using the known expression of $\bar{\mathbf{G}}_{\text{src}}(\mathbf{r}, \mathbf{r}')$ for a sphere in terms of the VSWFs (see, e.g., Appendix B of [54]). But here we prefer to stay in the framework of the standard Lorenz–Mie theory, only slightly wandering into the general VIE framework, which is employed in its full power further in Section 4.3. First, we write down the field induced by \mathbf{J}_{fl} without the particle itself, an analogue of the incident field in Eq. (4):

$$\mathbf{E}_0(\mathbf{r}) = i\omega\mu_0 \lim_{V_0(\mathbf{r}) \rightarrow 0} \int_{V \setminus V_0(\mathbf{r})} d^3\mathbf{r}' \bar{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}') \cdot \mathbf{J}_{\text{fl}}(\mathbf{r}') - \frac{i\omega\mu_0}{3k^2} \mathbf{J}_{\text{fl}}(\mathbf{r}), \quad (45)$$

where the singularity is excluded as explained in Eq. (E.14).

Let us look at the integral $\int_V d^3\mathbf{r} \mathbf{E}_0(\mathbf{r})$ considering different components of $\bar{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}')$ [cf. Eq. (A.3)]. The contribution of $\bar{\mathbf{G}}_{\text{st}}(\mathbf{r}, \mathbf{r}')$ is exactly zero, since

$$\int_V d^3\mathbf{r} \lim_{V_0(\mathbf{r}) \rightarrow 0} \int_{V \setminus V_0(\mathbf{r})} d^3\mathbf{r}' \bar{\mathbf{G}}_{\text{st}}(\mathbf{r}, \mathbf{r}') \cdot \mathbf{J}_{\text{fl}}(\mathbf{r}') = \int_V d^3\mathbf{r}' \left[\lim_{V_0(\mathbf{r}') \rightarrow 0} \int_{V \setminus V_0(\mathbf{r}')} d^3\mathbf{r} \bar{\mathbf{G}}_{\text{st}}(\mathbf{r}, \mathbf{r}') \right] \cdot \mathbf{J}_{\text{fl}}(\mathbf{r}') = 0, \quad (46)$$

where the integral in square brackets is the difference of two surface integrals over spheres, so-called self-terms; each of them is equal to $1/(3k^2)$ for any \mathbf{r}' inside [19,55]. The contribution of $\bar{\mathbf{G}}^R(\mathbf{r}, \mathbf{r}') - \bar{\mathbf{G}}_{\text{st}}(\mathbf{r}, \mathbf{r}')$ is always asymptotically negligible in comparison with the remaining self-term in Eq. (45). We neglect it here, but it is discussed in a more general setting in Section 4.2, see Eq. (65). Thus, we are left with asymptotically constant $\bar{\mathbf{G}}_0^I(\mathbf{r}, \mathbf{r}')$. Analogous to the derivation of Eq. (33), we obtain

$$\int_V d^3\mathbf{r} \mathbf{E}(\mathbf{r}) \cong d_1 \int_V d^3\mathbf{r} \mathbf{E}_0(\mathbf{r}) \cong -d_1 \frac{i}{3\omega\varepsilon_0} \left(1 - i\frac{V k^3}{2\pi}\right) \int_V d^3\mathbf{r} \mathbf{J}_{\text{fl}}(\mathbf{r}). \quad (47)$$

Next, we substitute Eqs. (37), (45), (47) into Eq. (44):

$$\mathbf{p}_{\text{fl}} \cong \frac{i}{\omega} \left[1 - \frac{\varepsilon - \varepsilon_0}{3\varepsilon_0} d_1 \left(1 - \frac{2i}{3} x^3\right) \right] \int_V d^3\mathbf{r} \mathbf{J}_{\text{fl}}(\mathbf{r}) \cong \frac{i}{\omega} d_1 \int_V d^3\mathbf{r} \mathbf{J}_{\text{fl}}(\mathbf{r}), \quad (48)$$

where the last equality somewhat surprisingly follows from Eq. (37) (for leading terms in both the real and imaginary parts).

Finally, with the use of Eqs. (43) and (48) the FDT becomes:

$$\langle \mathbf{p}_{\text{fl}} \otimes \mathbf{p}_{\text{fl}}^* \rangle \cong \frac{\Theta(\omega, T)}{\pi\omega} |d_1|^2 V \text{Im} \varepsilon \cong \frac{\Theta(\omega, T)}{\pi\omega} V \left| \frac{3\varepsilon_0}{\varepsilon + 2\varepsilon_0} \right|^2 \text{Im} \varepsilon = \frac{\Theta(\omega, T)}{\pi\omega} \bar{\chi}^I, \quad (49)$$

where we used Eqs. (31) and (37). The final result is exactly the same as Eq. (30) for the point dipole, but since asymptotic equivalence has been used in its derivation, it equally well agrees with Eq. (29).

4. Arbitrary small particle

4.1. Microscopic definition of the polarizability

In this section we consider a regular particle with the dyadic inhomogeneous permittivity $\bar{\boldsymbol{\varepsilon}}(\mathbf{r})$ and a small arbitrary-shaped volume V (Fig. 2). Since there are no singular points in this problem, we may use the standard VIE formulation for the

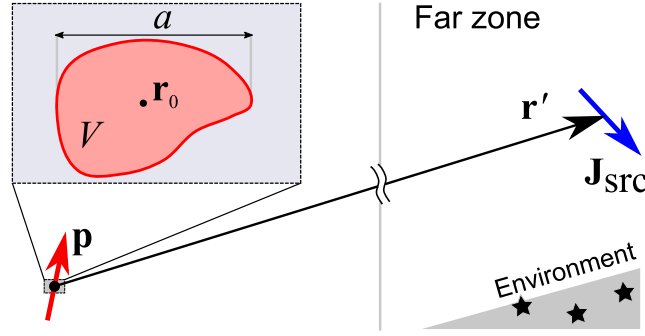


Fig. 2. An arbitrary-shaped small particle as a model of a point dipole.

electromagnetic scattering [18,19]. Let us denote the potential dyadic $\bar{\mathbf{U}}(\mathbf{r}) \triangleq \omega^2 \mu_0 [\bar{\boldsymbol{\epsilon}}(\mathbf{r}) - \varepsilon_0 \bar{\mathbf{I}}]$ and the polarization density $\mathbf{P}(\mathbf{r}) \triangleq \bar{\mathbf{U}}(\mathbf{r}) \cdot \mathbf{E}(\mathbf{r}) / (\omega^2 \mu_0)$ – the support of both is V . We limit ourselves to vacuum in the immediate vicinity of the particle (in addition to distant environment), but this can be replaced by any non-absorbing host medium as discussed in Section 3.1.

Employing the operator notation described in Appendix E, we write the VIE, also known as the Lippmann–Schwinger equation, in the form

$$\mathcal{E} = \mathcal{E}_0 + \mathcal{G}\mathcal{U}\mathcal{E}, \quad (50)$$

where $\mathcal{U}: \mathcal{H}_V \rightarrow \mathcal{H}_V$ is the multiplication operator [Eq. (E.13)] and $\mathcal{G}: \mathcal{H} \rightarrow \mathcal{H}$ is the singular Green’s operator [Eq. (E.14)]. \mathcal{H} and \mathcal{H}_V are spaces of square-integrable fields in \mathbb{R}^3 and V , respectively. One of the general ways to express the solution to this VIE is through the transition dyadic $\bar{\mathbf{T}}(\mathbf{r}, \mathbf{r}')$ or the operator $\mathcal{T}: \mathcal{H}_V \rightarrow \mathcal{H}_V$ [18,54], which by definition relates the incident field and the polarization:

$$\mathcal{U}\mathcal{E} = \omega^2 \mu_0 \mathcal{P} = \mathcal{T}\mathcal{E}_0 \Leftrightarrow \omega^2 \mu_0 \mathbf{P}(\mathbf{r}) = \int_V d^3 \mathbf{r}' \bar{\mathbf{T}}(\mathbf{r}, \mathbf{r}') \cdot \mathbf{E}_0(\mathbf{r}'), \quad (51)$$

where $\bar{\mathbf{T}}(\mathbf{r}, \mathbf{r}') = \bar{\mathbf{0}}$ unless both $\mathbf{r} \in V$ and $\mathbf{r}' \in V$, while $\mathcal{P} \in \mathcal{H}_V$. This operator is analogous to the transition operator in the quantum scattering theory, which was first applied to light scattering in [54]. Importantly, it is known to satisfy the following [18]:

$$\mathcal{T} = \mathcal{U} + \mathcal{U}\mathcal{G}\mathcal{T} = \mathcal{U} + \mathcal{T}\mathcal{G}\mathcal{U}, \quad (52)$$

which is also called the Lippmann–Schwinger equation due to its similarity to Eq. (50). Thus, \mathcal{T} or $\bar{\mathbf{T}}(\mathbf{r}, \mathbf{r}')$ is fully and uniquely determined by \mathcal{U} or $\bar{\boldsymbol{\epsilon}}(\mathbf{r})$, respectively, although evaluating $\bar{\mathbf{T}}(\mathbf{r}, \mathbf{r}')$ is generally a computationally-intensive problem. Moreover, the transition operator is closely related to the Waterman’s T-matrix widely used in computations [56–58], see Section 4.4. Using the integration map \mathcal{W}_0^H [Eq. (E.24)], we can express the total dipole moment as

$$\mathbf{p} \triangleq \int_V d^3 \mathbf{r} \mathbf{P}(\mathbf{r}) = \frac{1}{\omega^2 \mu_0} \mathcal{W}_0^H \mathcal{U}\mathcal{E}. \quad (53)$$

Note, however, that other (asymptotically equivalent) definitions of the dipole moment are based on the expansion of $\mathbf{P}(\mathbf{r})$ or the scattered field in free space into the VSWFs [59], which are discussed below.

Before deriving the expression for $\bar{\boldsymbol{\alpha}}$, let us consider the static case, keeping $\bar{\boldsymbol{\epsilon}}(\mathbf{r})$ (or \mathcal{U}/k^2) fixed to its value at ω , which corresponds to the definition of $\bar{\boldsymbol{\chi}}$ in Section 2.1. For that we apply the limit $\omega \rightarrow 0$ to Eq. (52) and obtain:

$$\mathcal{T}_{\text{st}} = \mathcal{U} + \mathcal{U}\mathcal{G}_{\text{st}}\mathcal{T}_{\text{st}} = \mathcal{U} + \mathcal{T}_{\text{st}}\mathcal{G}_{\text{st}}\mathcal{U}, \quad (54)$$

where $\mathcal{G}_{\text{st}}: \mathcal{H} \rightarrow \mathcal{H}$ is defined through its kernel, given by Eq. (A.2), and, similarly, $\mathcal{T}_{\text{st}}: \mathcal{H}_V \rightarrow \mathcal{H}_V$ have the kernel

$$\bar{\mathbf{T}}_{\text{st}}(\mathbf{r}, \mathbf{r}') \triangleq k^2 \lim_{k \rightarrow 0} \frac{1}{k^2} \bar{\mathbf{T}}(\mathbf{r}, \mathbf{r}'), \quad (55)$$

where the powers of k are due to the SI system of units. It is possible to renormalize the operators \mathcal{U} , \mathcal{G} , and \mathcal{T} in order to get rid of these coefficients, but we prefer to stick to the established definitions. Importantly, the environment contribution $\bar{\mathbf{G}}_{\text{env}}(\mathbf{r}, \mathbf{r}')$ disappears in this limit [cf. Eq. (7)], thus, \mathcal{T}_{st} , \mathcal{G}_{st} , and \mathcal{U} are independent of the environment and depend on k only through constant factors.

To our knowledge, \mathcal{T}_{st} has not been explicitly defined earlier, but its representation in terms of the VSWFs (the T-matrix) was obtained and used in [60–62], also with respect to the static polarizability [63]. Moreover, the electrostatic problem can be simplified by rewriting it in terms of surface-integral equation for the scalar potential, which is the usual method to find the static polarizability for various particles [6,64]. Thus, efficient numerical routines exist for evaluating $\bar{\mathbf{T}}_{\text{st}}(\mathbf{r}, \mathbf{r}')$, which is easier than that for $\bar{\mathbf{T}}(\mathbf{r}, \mathbf{r}')$, but still far from trivial for particles of arbitrary shape. We do not discuss the existence and uniqueness of a solution to Eqs. (52) and (54), but we assume that it is guaranteed for any sufficiently fine $\bar{\boldsymbol{\epsilon}}(\mathbf{r})$ – specific conditions are reviewed in [19].

Combining the static analogues of Eqs. (51), (53) we obtain:

$$\mathbf{p}_{\text{st}} = \frac{1}{\omega^2 \mu_0} \mathcal{W}_0^H \mathcal{T}_{\text{st}} \varepsilon_{0,\text{st}} \Rightarrow \bar{\boldsymbol{\chi}} = \frac{1}{\omega^2 \mu_0} \mathcal{W}_0^H \mathcal{T}_{\text{st}} \mathcal{W}_0 = \frac{1}{\omega^2 \mu_0} \iint_V d^3 \mathbf{r} d^3 \mathbf{r}' \bar{\mathbf{T}}_{\text{st}}(\mathbf{r}, \mathbf{r}'), \quad (56)$$

where we used the constancy of the incident field inside V , which can be expressed as $\mathcal{J}_V \varepsilon_{0,\text{st}} = \mathcal{W}_0 \cdot \mathbf{E}_0$. Here \mathcal{J}_V is the projector on \mathcal{T}_V , which zeroes the function values outside of V . Obviously, this expression for $\bar{\boldsymbol{\chi}}$ depends neither on the environment nor on k ; therefore, it is analogous to Eq. (31) for spheres.

The dynamic polarizability is obtained analogously to Eq. (56):

$$\bar{\boldsymbol{\alpha}} \cong \frac{1}{\omega^2 \mu_0} \mathcal{W}_0^H \mathcal{T} \mathcal{W}_0 = \frac{1}{\omega^2 \mu_0} \iint_V d^3 \mathbf{r} d^3 \mathbf{r}' \bar{\mathbf{T}}(\mathbf{r}, \mathbf{r}'), \quad (57)$$

but the equation is only asymptotic, since it is based on $\mathcal{J}_V \varepsilon_0 \cong \mathcal{W}_0 \cdot \mathbf{E}_0$ (the incident field is asymptotically constant inside V). Note that \mathbf{r}_0 (where \mathbf{E}_0 is evaluated) is an arbitrary fixed point inside V (Fig. 2). It may seem tempting to employ Eq. (57) for particles of non-negligible size, but then the whole notion of the polarizability is ambiguous, as discussed in Section 3.1. In particular, Eq. (57) describes the response (value of \mathbf{p}) to the constant \mathbf{E}_0 (its projection on three unit vectors), while all three approaches described in Section 3.1 – to its projection on three basic VSWFs. These two bases are asymptotically equivalent, but if the next-order corrections are desired, the most appropriate basis will depend on the specific incident field. Similarly, these approaches use different definitions of the dipole moment, mentioned above. This is further discussed in Section 4.2.

To gain more insight, we obtain the same result using the general relation for the source Green's operator [18], also known as the Dyson equation [65,66]:

$$\mathcal{G}_{\text{src}} = \mathcal{G} + \mathcal{G} \mathcal{T} \mathcal{G}, \quad (58)$$

and the fact that if $\mathbf{r} \in V$ and \mathbf{r}' is relatively far, then $\bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}') \cong \bar{\mathbf{G}}(\mathbf{r}_0, \mathbf{r}')$. This leads to:

$$\bar{\mathbf{G}}_{\text{src}}(\mathbf{r}, \mathbf{r}') \cong \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}') + \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) \cdot \mathcal{W}_0^H \mathcal{T} \mathcal{W}_0 \cdot \bar{\mathbf{G}}(\mathbf{r}_0, \mathbf{r}'), \quad (59)$$

which upon comparison with Eq. (5) gives the same definition of $\bar{\boldsymbol{\alpha}}$. This derivation is justified only when the characteristic particle size $a \ll |\mathbf{r} - \mathbf{r}_0|, |\mathbf{r}' - \mathbf{r}_0|, \lambda$. To conclude, the simple Eq. (5), a quintessence of the point dipole, is valid only if the particle is very small. And in this limit all reasonable definitions of $\bar{\boldsymbol{\alpha}}$ are asymptotically equivalent.

One can further use the results of Section 2, since they all follow from Eq. (5), but we provide a microscopic derivation below, based on [20]:

$$\begin{aligned} W_{\text{ext}} &= -\frac{\omega}{2} \text{Im} \langle \varepsilon_0, \mathcal{P} \rangle = \frac{1}{2\omega\mu_0} \langle \mathcal{T}^I \varepsilon_0, \varepsilon_0 \rangle \cong \frac{\omega}{2} \text{Im}(\mathbf{E}_0^* \cdot \mathbf{p}), \\ W_{\text{sca}} &= \frac{\omega^3 \mu_0}{2} \langle \mathcal{G}^I \mathcal{P}, \mathcal{P} \rangle \cong \frac{\omega^3 \mu_0}{2} \mathbf{p}^* \cdot \bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \mathbf{p}, \end{aligned} \quad (60)$$

where the inner product $\langle \cdot, \cdot \rangle$ is defined by Eq. (E.2) and we used Eq. (51) and the constancy of $\mathbf{E}_0(\mathbf{r})$ and $\bar{\mathbf{G}}^I(\mathbf{r}, \mathbf{r}')$ inside V ($\mathcal{J}_V \varepsilon_0 \cong \mathcal{W}_0 \cdot \mathbf{E}_0$, $\mathcal{J}_V \mathcal{G}^I \mathcal{J}_V \cong \mathcal{W}_0 \cdot \bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \mathcal{W}_0^H$). The asymptotic expression for W_{abs} is then obtained as in Eq. (18), but it is instructive to look at the exact operator expression:

$$\begin{aligned} W_{\text{abs}} &= \frac{1}{2\omega\mu_0} \langle (\mathcal{T}^I - \mathcal{T}^H \mathcal{G}^I \mathcal{T}) \varepsilon_0, \varepsilon_0 \rangle = \frac{1}{2\omega\mu_0} \langle [J - (\mathcal{G} - \mathcal{G}_{\text{st}}) \mathcal{T}_{\text{st}}]^{-H} \mathcal{T}_{\text{st}}^I [J - (\mathcal{G} - \mathcal{G}_{\text{st}}) \mathcal{T}_{\text{st}}]^{-1} \varepsilon_0, \varepsilon_0 \rangle \\ &= -\frac{\omega^3 \mu_0}{2} \langle (\mathcal{T}_{\text{st}}^{-1})^I \mathcal{P}, \mathcal{P} \rangle, \end{aligned} \quad (61)$$

where we used Eqs. (F.9), (F.11).

First, Eq. (61) shows that W_{abs} is proportional to $\mathcal{T}_{\text{st}}^I$, i.e., the latter determines the magnitude of absorption. Second, the last part of Eq. (61) is valid only when \mathcal{U}^{-1} exist [Eq. (F.10)] and is completely analogous to Eq. (13). This seems to justify the use of the latter as a starting assumption in the phenomenological theory. However, although $(\mathcal{T}_{\text{st}}^{-1})^I = (\mathcal{U}^{-1})^I$ [Eq. (F.10)], i.e., a trivial multiplication operator, there is generally no direct relation between it and $(\bar{\boldsymbol{\chi}}^{-1})^I$ such as Eq. (56). Moreover, multiplication operator is different from operators with almost constant kernel (e.g., \mathcal{G}^I), thus even for homogeneous scatterers W_{abs} is proportional to $\langle \mathcal{P}, \mathcal{P} \rangle$, which is generally not equal to $\langle \mathcal{W}_0 \mathcal{P}, \mathcal{W}_0 \mathcal{P} \rangle = \mathbf{p}^* \cdot \mathbf{p}$. The main problem is that $\mathbf{P}(\mathbf{r})$ is not constant even in the limit $a \rightarrow 0$ (in contrast to $\mathbf{E}_0(\mathbf{r})$), unless the particle is a homogeneous spheroid [9]. But even in this case it helps only to obtain the limiting values, but not the next-order corrections. For example, as discussed in Section 3.1 for spheres, the contribution of Lorenz–Mie coefficients (other than a_1) to \mathbf{p} is exactly zero, but contribution to $\langle \mathcal{P}, \mathcal{P} \rangle$ and, hence, to W_{abs} is small, but non-zero.

One may also argue that the last part of Eq. (61) justifies that absorption is a local quantity, since it doesn't depend on the environment if \mathcal{P} is fixed. However, the latter is a subtle assumption. One can indeed assume the fixed \mathbf{p} when varying the environment (since it can always be compensated by varying \mathbf{E}_0), but keeping the whole function \mathcal{P} fixed will require intricate modification of ε_0 according to Eqs. (51), (52).

4.2. Relation between the static and dynamic polarizabilities

To derive the analogue of Eqs. (11), (20) for small particles, let us use the following operator identity, which is derived in Appendix F [Eq. (F.2)]:

$$\mathcal{T} - \mathcal{T}_{\text{st}} = \mathcal{T} (\mathcal{G} - \mathcal{G}_{\text{st}}) \mathcal{T}_{\text{st}}. \quad (62)$$

Applying \mathcal{W}_0 on both sides of this identity and using Eq. (56) and (57), we have:

$$\bar{\alpha} - \bar{\chi} = \frac{1}{\omega^2 \mu_0} \mathcal{W}_0^H \mathcal{T} (\mathcal{G} - \mathcal{G}_{\text{st}}) \mathcal{T}_{\text{st}} \mathcal{W}_0. \quad (63)$$

To evaluate the integral (the inner product) on the right-hand side, we use the identity

$$\mathcal{G} - \mathcal{G}_{\text{st}} = \mathcal{G}_1 + i\mathcal{G}^I, \quad \mathcal{G}_1 \triangleq \mathcal{G}^R - \mathcal{G}_{\text{st}}. \quad (64)$$

where both \mathcal{G}_1 and \mathcal{G}^I are Hermitian and $\bar{\mathbf{G}}^I(\mathbf{r}, \mathbf{r}')$ is asymptotically constant inside V ($\mathcal{J}_V \mathcal{G}^I \mathcal{J}_V \cong \mathcal{W}_0 \cdot \bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \mathcal{W}_0^H$). Thus, we obtain:

$$\bar{\alpha} - \bar{\chi} \cong \frac{i}{\omega^2 \mu_0} \bar{\alpha} \cdot \bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \bar{\chi} + \frac{1}{\omega^2 \mu_0} \mathcal{W}_0^H \mathcal{T} \mathcal{G}_1 \mathcal{T}_{\text{st}} \mathcal{W}_0. \quad (65)$$

Everything without the last term exactly corresponds to Eq. (11), while this term is tricky since $\bar{\mathbf{G}}^R(\mathbf{r}, \mathbf{r}') - \bar{\mathbf{G}}_{\text{st}}(\mathbf{r}, \mathbf{r}')$ cannot be considered constant inside V and is even weakly singular — see the first term in the right-hand side of Eq. (A.3). It can only be evaluated if at least one of \mathcal{T} or \mathcal{T}_{st} is known either analytically or numerically, since one can be replaced by the other inside this term keeping the asymptotic accuracy. In terms of the small parameter $x = ka$, this term is $\mathcal{O}(1/x)$ times larger than the term with $\bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0)$, but still $\mathcal{O}(x^2)$ of the polarizability itself. However, it can be shown [see Eq. (F.9)] that the correction to $\bar{\alpha}^I$ due to \mathcal{G}_1 is $\mathcal{O}(x^2 \|\bar{\chi}\|^I)$, which is asymptotically negligible for any absorption, in contrast to the term with $\bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0)$, which is $\mathcal{O}(x^3 \|\bar{\chi}\|)$. This justifies the names non-radiative and radiative corrections for these two terms, respectively.

Trying to account for non-radiative correction (for a given particle shape) is not meaningless but the benefit will generally be of the same order of magnitude as accounting for the magnetic-dipole and electric-quadrupole contributions to the scattering quantities, which corresponds to non-constant corrections to $\mathbf{E}_0(\mathbf{r})$ (as was discussed for a sphere in Section 3.1). The notable exception is the dipole plasmon resonance of metallic nanoparticles, close to which staying within the electric-dipole approximation is well justified. But even then there are same-order differences based on specific basis to project \mathcal{T} : Eq. (57) is based on the constant basis, while three options discussed in Section 3.1 use the VSWFs on the right and different functions on the left. In particular, Eqs. (32) and (33) use $\text{RgN}_{1m}(kr)$ and the constant basis, respectively, for the projection from the left in Eq. (57), while Eqs. (37) and (39) use the expansion in terms of $\text{RgN}_{1m}(m_0kr)$ on the left instead of the projection. The latter has the closest resemblance to the last term in Eq. (65), in particular, the correction is proportional to $(\epsilon - \epsilon_0)^2$. However, it is not clear if this relation can be made explicit. Moreover, in the case of a non-symmetric particle, the operator \mathcal{T} is not diagonal in the VSWF basis. This implies that the dipole moment can be induced by other terms in the multipole expansion of $\mathbf{E}_0(\mathbf{r})$. This effect can be considered as a non-local contribution to the polarizability, which is comparable to the non-radiative correction even in the case of plasmon resonance, as was recently shown in [67].

Similar conclusions were reached in the framework of the DDA, where the non-radiative correction is given by the so-called M term [14,68]. Some integrals of the Green's dyadic over the volume of a dipole (a voxel in the DDA) can be computed, but they do not account for all corrections of $\mathcal{O}(x^2)$ with respect to the main term. Note, however, that the case of the DDA is somewhat easier, since the field inside a voxel is almost constant, when the voxel is surrounded on all sides by other voxels of the same material (that is the case for majority of voxels in real applications of the DDA). By contrast, the polarizability of the voxels close to the particle boundary can be corrected based on their surrounding [69], but this non-local (collective) correction is outside of the scope of this review.

The Hermitian part of the environmental contribution to the Green's dyadic, $\bar{\mathbf{G}}_{\text{env}}^R(\mathbf{r}_0, \mathbf{r}_0)$, is regular and can in principle be added to $\bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0)$ in Eq. (65), but it is also asymptotically negligible and even $\mathcal{O}(x)$ times smaller than the non-radiative corrections. This explains larger attention to the imaginary part of $\bar{\mathbf{G}}_{\text{env}}$ in numerical analysis [70]. To gain more insight into this issue and to add rigor to the above order-of-magnitude analysis, let us expand the polarizability in powers of a , which was discussed for a few specific examples in [15]. In doing so we assume that all other problem parameters are fixed, including the shape of the particle and distribution of $\bar{\mathbf{e}}(\mathbf{r})$ inside it (i.e., the argument \mathbf{r} scales with a). In free space, we may alternatively fix the particle including a and let $k \rightarrow 0$ (up to a factor a^3 due to the particle volume). In the presence of the environment, we need to additionally scale all its dimensions with wavelength so that $\bar{\mathbf{G}}_{\text{env}}(\mathbf{r}_0, \mathbf{r}_0)$ stays the same.

We start with $\bar{\chi} = \mathcal{O}(a^3)$, which follows from independence of $\bar{\chi}$ on k [cf. Eq. (31)]. This implies that

$$a \rightarrow 0: \bar{\mathbf{T}}_{\text{st}} \left(\frac{\mathbf{r}}{a}, \frac{\mathbf{r}'}{a} \right) = \mathcal{O}(a^{-3}), \quad \mathcal{T}_{\text{st}} = \mathcal{O}(1), \quad (66)$$

where the kernel estimate is rigorous, while the order of operators is a convenient notation to indicate its action on the smooth function (different from the kernel by a factor of volume). It can be made rigorous using the operator norms, but here we just use it to simplify equations. Analogously, we write down the unknown expansion for \mathcal{T} :

$$\mathcal{T} = \mathcal{T}_{\text{st}} + \mathcal{T}^{(1)} + \mathcal{T}^{(2)} + \mathcal{T}^{(3)} + \mathcal{O}(a^4), \quad (67)$$

where, by definition, $\mathcal{T}^{(n)} = \mathcal{O}(a^n)$, and $\mathcal{T}^{(0)} = \mathcal{T}_{\text{st}}$ due to the equivalence of the two limits discussed above. Next, Eqs. (64), (A.3) imply

$$\mathcal{J}_V (\mathcal{G} - \mathcal{G}_{\text{st}}) \mathcal{J}_V = \mathcal{J}_V \left(\mathcal{G}_1^{(2)} + \mathcal{G}^{(3)} \right) \mathcal{J}_V + \mathcal{O}(a^4), \quad (68)$$

where $\mathcal{G}_1^{(2)}$ is the leading-order term in the expansion of \mathcal{G}_1 with the kernel given by the first term in the right-hand side of Eq. (A.3), $\mathcal{G}^{(3)}$ is the operator with the constant kernel $i\bar{\mathbf{G}}_0^I(\mathbf{r}_0, \mathbf{r}_0) + \bar{\mathbf{G}}_{\text{env}}(\mathbf{r}_0, \mathbf{r}_0)$, and the orders in superscripts are correct only after projection on V .

Substituting Eqs. (66)–(68) into Eq. (62) we obtain:

$$\mathcal{T}^{(1)} = 0, \quad \mathcal{T}^{(2)} = \mathcal{T}_{\text{st}} \mathcal{G}_1^{(2)} \mathcal{T}_{\text{st}}, \quad \mathcal{T}^{(3)} = \mathcal{T}_{\text{st}} \mathcal{W}_0 \cdot \left[i\bar{\mathbf{G}}_0^I(\mathbf{r}_0, \mathbf{r}_0) + \bar{\mathbf{G}}_{\text{env}}(\mathbf{r}_0, \mathbf{r}_0) \right] \cdot \mathcal{W}_0^H \mathcal{T}_{\text{st}}, \quad (69)$$

which together with Eqs. (56) and (57) leads to [cf. Eq. (63)]:

$$\bar{\boldsymbol{\alpha}} = \bar{\boldsymbol{\chi}} + \frac{1}{\omega^2 \mu_0} \mathcal{W}_0^H \mathcal{T}_{\text{st}} \mathcal{G}_1^{(2)} \mathcal{T}_{\text{st}} \mathcal{W}_0 + \omega^2 \mu_0 \bar{\boldsymbol{\chi}} \cdot \left[i\bar{\mathbf{G}}_0^I(\mathbf{r}_0, \mathbf{r}_0) + \bar{\mathbf{G}}_{\text{env}}(\mathbf{r}_0, \mathbf{r}_0) \right] \cdot \bar{\boldsymbol{\chi}} + \mathcal{O}(a^7), \quad (70)$$

where the two correction terms are $\mathcal{O}(a^5)$ and $\mathcal{O}(a^6)$, respectively. Finally, the inverse polarizability is given by [cf. Eq. (11)]:

$$\bar{\boldsymbol{\alpha}}^{-1} = \bar{\boldsymbol{\chi}}^{-1} - \frac{1}{\omega^2 \mu_0} \bar{\boldsymbol{\chi}}^{-1} \cdot \mathcal{W}_0^H \mathcal{T}_{\text{st}} \mathcal{G}_1^{(2)} \mathcal{T}_{\text{st}} \mathcal{W}_0 \cdot \bar{\boldsymbol{\chi}}^{-1} - \omega^2 \mu_0 \left[i\bar{\mathbf{G}}_0^I(\mathbf{r}_0, \mathbf{r}_0) + \bar{\mathbf{G}}_{\text{env}}(\mathbf{r}_0, \mathbf{r}_0) \right] + \mathcal{O}(a), \quad (71)$$

where the terms on the right-hand side are $\mathcal{O}(a^{-3})$, $\mathcal{O}(a^{-1})$, and $\mathcal{O}(1)$, respectively. Thus, we have proved that the $\mathcal{O}(1)$ term in expansion of $\bar{\boldsymbol{\alpha}}^{-1}$ is constant and $\mathcal{O}(a^{-2})$ term is absent for any particle shape and $\bar{\boldsymbol{\epsilon}}(\mathbf{r})$. This has been conjectured and demonstrated for a few examples in free space in [15]. Moreover, this conclusion also remains valid for the polarizabilities defined in terms of the VSWFs [Eq. (79)], since the Taylor expansions of these VSWFs [Eq. (D.10)] include only even powers of kr , which affects only $\mathcal{O}(a^{-1})$, $\mathcal{O}(a)$ and further terms in Eq. (71).

Although this analysis assigns fundamental meaning to $i\bar{\mathbf{G}}_0^I(\mathbf{r}_0, \mathbf{r}_0) + \bar{\mathbf{G}}_{\text{env}}(\mathbf{r}_0, \mathbf{r}_0)$, including its Hermitian part, it does not provide a rigorous justification that this part should always be included in the correction. Thus, the ambiguity of this correction remains. Note also that one of the options may prove to be more accurate in real (experimental) applications. However, lack of a rigorous justification suggests that such empirical choice is not necessarily universal, but may depend on a specific configuration.

4.3. Fluctuation–dissipation theorem for a small particle

In this section, we derive the fluctuation–dissipation theorem for a particle with arbitrary shape, accounting for microscopic fluctuating currents. Let us first rewrite the FDT for currents [Eq. (43)] in a following operator form:

$$\langle \mathcal{J}_{\text{fl}} \otimes \mathcal{J}_{\text{fl}}^* \rangle = \frac{1}{\pi \omega \mu_0} \Theta(\omega, T) \mathcal{U}^I, \quad (72)$$

where \mathcal{J}_{fl} is the fluctuating current and $\mathcal{U}^I = (\mathcal{U} - \mathcal{U}^H)/2i$ is a generalization of $\text{Im } \epsilon$ to the case of tensorial and generally non-reciprocal permittivity (cf. Eq. 2.66 in [71]). Note that the time-averaging $\langle \dots \rangle$ should not be confused with the inner product. These fluctuating currents cause the fluctuating dipole moment \mathbf{p}_{fl} both directly as $(i/\omega) \mathcal{W}_0^H \mathcal{J}_{\text{fl}}$ and due to scattering of the corresponding field $\mathcal{E}_0 = i\omega \mu_0 \mathcal{G} \mathcal{J}_{\text{fl}}$, analogously to the discussion in Section 3.2 for a sphere. Using Eqs. (51), (53) we obtain:

$$\mathbf{p}_{\text{fl}} = \frac{i}{\omega} \mathcal{W}_0^H (\mathcal{J} + \mathcal{T} \mathcal{G}) \mathcal{J}_{\text{fl}} \quad (73)$$

(\mathcal{J} is the identity operator), which together with Eqs. (52), (72), (E.16), (E.22) leads to:

$$\begin{aligned} \langle \mathbf{p}_{\text{fl}} \otimes \mathbf{p}_{\text{fl}}^* \rangle &= \frac{1}{\omega^2} \mathcal{W}_0^H (\mathcal{J} + \mathcal{T} \mathcal{G}) \langle \mathcal{J}_{\text{fl}} \otimes \mathcal{J}_{\text{fl}}^* \rangle (\mathcal{J} + \mathcal{G}^H \mathcal{T}^H) \mathcal{W}_0 = \frac{\Theta(\omega, T)}{2i\pi\omega^3 \mu_0} \mathcal{W}_0^H \left[\mathcal{T} (\mathcal{J} + \mathcal{G}^H \mathcal{T}^H) - (\mathcal{J} + \mathcal{T} \mathcal{G}) \mathcal{T}^H \right] \mathcal{W}_0 \\ &= \frac{\Theta(\omega, T)}{\pi\omega^3 \mu_0} \mathcal{W}_0^H (\mathcal{T}^I - \mathcal{T} \mathcal{G}^I \mathcal{T}^H) \mathcal{W}_0 \cong \frac{\Theta(\omega, T)}{\pi\omega} \left[\bar{\boldsymbol{\alpha}}^I - \bar{\boldsymbol{\alpha}} \cdot \bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \bar{\boldsymbol{\alpha}}^H \right]. \end{aligned} \quad (74)$$

This result is the same as Eq. (29) for a point dipole and Eq. (49) for a small sphere. The last (asymptotic) equality in Eq. (74) is due to the asymptotic constancy of $\bar{\mathbf{G}}^I(\mathbf{r}, \mathbf{r}')$ inside V , as in Eq. (65), and due to Eq. (E.10). Moreover, using Eq. (F.9), we can rewrite Eq. (74) as:

$$\langle \mathbf{p}_{\text{fl}} \otimes \mathbf{p}_{\text{fl}}^* \rangle = \frac{\Theta(\omega, T)}{\pi\omega^3 \mu_0} \mathcal{W}_0^H \left[\mathcal{J} - (\mathcal{G} - \mathcal{G}_{\text{st}}) \mathcal{T}_{\text{st}} \right]^{-H} \mathcal{T}_{\text{st}}^I \left[\mathcal{J} - (\mathcal{G} - \mathcal{G}_{\text{st}}) \mathcal{T}_{\text{st}} \right]^{-1} \mathcal{W}_0 = -\frac{\Theta(\omega, T)}{\pi\omega^3 \mu_0} (\mathcal{T} \mathcal{W}_0)^H (\mathcal{T}_{\text{st}}^{-1})^I \mathcal{T} \mathcal{W}_0, \quad (75)$$

where the last part assumes the existence of $\mathcal{T}_{\text{st}}^{-1}$ [Eq. (F.10)]. Similar to Eq. (61) for W_{abs} , we see that $\langle \mathbf{p}_{\text{fl}} \otimes \mathbf{p}_{\text{fl}}^* \rangle$ is proportional to $\mathcal{T}_{\text{st}}^I$ and is asymptotically independent of the environment.

4.4. Relation to the T-matrix

The T-matrix method is widely used in light-scattering simulations [56]. In this method, incident and scattered fields are represented as superpositions of the VSWFs (Appendix D), and the matrix \mathbf{T} relates the decomposition coefficients. It was shown [58,72] that for a particle in free space \mathbf{T} is the projection of $ik\mathcal{T}$ onto the basis of the regular VSWFs. In particular, the elements of the lower right block of \mathbf{T} (denoted as \mathbf{T}^{22}) are expressed as

$$T_{nm'm'}^{22} = ik \iint_V d^3\mathbf{r} d^3\mathbf{r}' \text{Rg}\mathbf{N}_{nm}^*(k\mathbf{r}) \cdot \bar{\mathbf{T}}(\mathbf{r}, \mathbf{r}') \cdot \text{Rg}\mathbf{N}_{n'm'}(k\mathbf{r}'), \quad (76)$$

where $n \in \mathbb{N}$, $-n \leq m \leq n$, and the VSWFs $\text{Rg}\mathbf{N}_{nm}$ are defined by Eq. (D.3). Other three blocks of \mathbf{T} are expressed similarly employing the functions $\text{Rg}\mathbf{M}_{nm}$ (twice or in combination with the $\text{Rg}\mathbf{N}_{nm}$), but they correspond to the magnetic multipoles and vanish for small sizes due to Eq. (D.9). Note that the letter \mathbf{T} is used for the transition operator and the dyadic, as well as for the T-matrix as a whole (or its blocks) and its elements. However, this should not cause a confusion as each symbol is different by either its font or used indices.

Let us briefly recall the derivation of Eq. (76) to be able to generalize it to the arbitrary environment (at the end of this section). Overall, it is not surprising that \mathbf{T} is related to \mathcal{T} , since they both operate on the incident field \mathcal{E}_0 (either the function or its expansion). However, \mathcal{T} produces the internal polarization \mathcal{P} (up to a coefficient), while \mathbf{T} – the scattered field $\mathcal{E}_{\text{sca}} \stackrel{\text{def}}{=} \mathcal{E} - \mathcal{E}_0$. Let us define the scattering transition operator $\mathcal{T}_{\text{sca}} : \mathcal{H}_V \rightarrow \mathcal{H}$, given by

$$\mathcal{T}_{\text{sca}} \stackrel{\text{def}}{=} \mathcal{G}\mathcal{T} \Rightarrow \mathcal{E}_{\text{sca}} = \mathcal{T}_{\text{sca}}\mathcal{E}_0. \quad (77)$$

where we used Eqs. (50), (51). In principle, it produces \mathcal{E}_{sca} everywhere, but we are interested only in its values far from the particle, at least outside of the circumscribing sphere. Then a complete basis for the expansion of \mathcal{E}_{sca} is a set of the outgoing VSWFs \mathbf{N}_{nm} and \mathbf{M}_{nm} [Eq. (D.3)]. Therefore, in free space \mathbf{T} is the projection of \mathcal{T}_{sca} on the regular VSWFs on the right and the expansion in terms of the outgoing VSWFs on the left, respectively, see also [73].

In free space we use the diagonal expansion of \mathcal{G}_0 in terms of the VSWFs [Eq. (D.13)], which immediately implies Eq. (76). Let us define $\bar{\mathbf{T}}_{11}^{22}$ as a dyadic with components $(\bar{\mathbf{T}}_{11}^{22})_{\mu\nu} = T_{1,\mu-2,1,\nu-2}^{22}$, then we obtain

$$\bar{\mathbf{T}}_{11}^{22} = ik\mathcal{W}_s^H \mathcal{T}_0 \mathcal{W}_s \cong \frac{ik}{\delta\pi} \bar{\mathbf{Z}}^H \cdot \mathcal{W}_0^H \mathcal{T}_0 \mathcal{W}_0 \cdot \bar{\mathbf{Z}}, \quad (78)$$

which is the only part of Eq. (76) that survives the limit $x \rightarrow 0$. \mathcal{T}_0 is the transition operator in free space and \mathcal{W}_s is the projection operator [Eq. (E.17)] onto the basis $\bar{\mathbf{W}}_s(\mathbf{r})$, which is constructed from $\text{Rg}\mathbf{N}_{lm}(kr)$ and asymptotically proportional to the constant unitary dyadic $\bar{\mathbf{Z}}$ [Eq. (D.12)]. The difference between \mathcal{W}_s and $\mathcal{W}_0 \cdot \bar{\mathbf{Z}}/\sqrt{6\pi}$ explains the differences between various definitions of the polarizability and the dipole moment in Sections 3.1 and 4.1, and is related to [59]. Comparing Eq. (78) to Eq. (57), we obtain

$$\bar{\boldsymbol{\alpha}} \cong -i \frac{6\pi\epsilon_0}{k^3} \bar{\mathbf{Z}} \cdot \bar{\mathbf{T}}_{11}^{22} \cdot \bar{\mathbf{Z}}^H, \quad (79)$$

a general relation between $\bar{\boldsymbol{\alpha}}$ and the T-matrix elements for any particle in free space in any coordinate system. This relation (lacking the rotation of the basis, i.e., the dyadic $\bar{\mathbf{Z}}$) was previously mentioned in [11]. If different definition of the VSWFs is used, Eq. (79) will stay valid but $\bar{\mathbf{Z}}$ will change [64,74]. As an example, let us apply Eq. (79) to a particle symmetric with respect to the rotation over the z -axis. Then \mathbf{T} is diagonal with respect to m and $T_{n,-m,n,-m} = T_{nmnm}$ [56], leading to:

$$\bar{\boldsymbol{\alpha}} = -i \frac{6\pi\epsilon_0}{k^3} \begin{pmatrix} T_{1111}^{22} & 0 & 0 \\ 0 & T_{1111}^{22} & 0 \\ 0 & 0 & T_{1010}^{22} \end{pmatrix}. \quad (80)$$

which is consistent with Eq. (32) since $T_{nmnm}^{22} = -a_n$ (Eq. (5.44) of [25]).

Similar (generalized) relations with the elements of \mathbf{T} are known for the magnetic-dipole, electric-quadrupole, and higher-multipole polarizabilities. Moreover, a general form of the radiative correction was obtained [11], applicable to \mathbf{T} as a whole. This formulation is completely analogous to the operator description in Section 4.2. In particular, let us define the *symmetrized transition operator* \mathcal{T}_{sym} corresponding to the Hermitian (symmetric) part of \mathcal{G} , thus satisfying

$$\mathcal{T}_{\text{sym}} = \mathcal{U} + \mathcal{U}\mathcal{G}^R\mathcal{T}_{\text{sym}}. \quad (81)$$

Then, fully analogous to Eq. (62), we obtain:

$$\mathcal{T} - \mathcal{T}_{\text{sym}} = i\mathcal{T}\mathcal{G}^I\mathcal{T}_{\text{sym}}. \quad (82)$$

Next, the reactance matrix \mathbf{K} , defined in [11], is the projection–expansion of operator $-i\mathcal{G}\mathcal{T}_{\text{sym}}$ in the VSWF basis, same as for \mathcal{T}_{sca} above, which in free space equals the projection of $k\mathcal{T}_{\text{sym}}$ on the regular VSWF basis. Indeed, using the expansions of \mathcal{G}_0^I in terms of the VSWFs [Eq. (D.14)], Eq. (82) implies

$$\mathbf{T} - i\mathbf{K} = i\mathbf{TK}, \quad (83)$$

which is exactly Eq. (13) of [11]. Moreover, all symmetry properties of \mathbf{K} , discussed in [11], directly relate to the corresponding properties of \mathcal{T}_{sym} . For instance, for non-absorbing particles \mathcal{U} is Hermitian, then the same is true for \mathcal{T}_{sym} and, hence, for \mathbf{K} .

The standard radiative correction corresponds to neglecting the last term in Eq. (65), i.e., replacing $\mathcal{G} - \mathcal{G}_{\text{st}}$ by $i\mathcal{G}^I$ Eq. (62). Then, again in free space, we obtain exactly Eq. (21) of [11]:

$$\mathbf{T} - i\mathbf{K}^{(0)} = i\mathbf{TK}^{(0)}, \quad (84)$$

where $\mathbf{K}^{(0)}$ is the static limit of \mathbf{K} (or the projection of $k\mathcal{T}_{\text{st}}$ on the regular VSWF basis). In other words, a straightforward static limit of \mathbf{K} is asymptotically correct in contrast to \mathbf{T} , which fully agrees with conclusions of Section 4.2.

This property of \mathbf{K} was used in several applications. In particular, the reformulation of the Lorenz–Mie coefficients in terms of \mathbf{K} allows one to predict the morphology-dependent resonances for small spheres, which is impossible to do by simple Taylor series expansion [75]. It also simplifies consideration of the interaction of light with magnetic and electric resonant particles [76] and allows one to obtain the radiation-corrected approximate \mathbf{T} for small spheroids [77].

Now let us return to the general setting of this review, which includes arbitrary environment specified by the operator \mathcal{G}_{env} . In this case the whole notion of the T-matrix becomes ambiguous. First, it may relate to the particle in free space, e.g., [78], defined by Eq. (76) applied to \mathcal{T}_0 – we denote it as \mathbf{T}_0 . Second, one may expand the total scattered field \mathcal{E}_{sca} into the outgoing VSWFs (at

least in some region, depending on the environment) and relate it with the expansion coefficients of \mathcal{E}_0 . This corresponds to the expansion–projection of the scattering transition operator \mathcal{T}_{sca} , see Eq. (77), or to the T-matrix of the whole system (particle plus environment) with the only difference that \mathcal{E}_0 already accounts for the environment, i.e., it satisfies the Maxwell’s equations in the presence of the environment (without a particle) rather than in free space. More importantly, the environment generally causes \mathcal{E}_{sca} to have non-negligible multipole components even for a point dipole. More specifically, the Green’s operator \mathcal{G} in Eq. (77), including \mathcal{G}_{env} , propagates the polarization inside the particle into the field outside. This propagator may be cumbersome, but is independent of the particle, thus, we do not discuss it further. The third option is an intermediate one — we include the effect of \mathcal{G}_{env} on the internal polarization \mathcal{P} (self-action of the particle through the environment) but not its effect on the scattered field. Everything related to \mathcal{P} is encoded in the transition operator \mathcal{T} , and we follow the definition of the T-matrix given by Eq. (76), i.e., the projection of $ik\mathcal{T}$ on the basis of the regular VSWFs, and denote it as \mathbf{T} . The factor ik is taken from the definition of \mathbf{T}_0 , where it corresponds to the effect of the free-space propagator \mathcal{G}_0 (discussed above).

One of the most convenient properties of the chosen definition of \mathbf{T} is that for a small particle Eq. (79) remains valid in a general environment. To relate it with \mathbf{T}_0 we start with

$$\mathcal{T} - \mathcal{T}_0 = \mathcal{T}\mathcal{G}_{\text{env}}\mathcal{T}_0, \quad (85)$$

obtained analogously to Eqs. (62), (82). Next we expand \mathcal{G}_{env} in terms of the regular VSWFs inside the particle; the result is similar to Eq. (D.14) but with the matrix of coefficients $ik\mathbf{G}_{\text{env}}$ (indexed as the matrix \mathbf{T} and accounting for the cross terms between different orders and types of the VSWFs) instead of $k\mathbf{I}$. Alternatively, \mathbf{G}_{env} can be considered as a linear transformation of the outgoing VSWFs, generated by the particle and reflected by the environment, into the regular VSWFs acting back on the particle (see the matrix \mathbf{A} in [78]). Note that the regular behavior of such expansion requires that the sphere circumscribing the particle does not intersect any elements of the environment, e.g., substrate, otherwise the standard T-matrix approach is not valid, unless certain modifications are made [79]. Fortunately, this requirement is always satisfied in the limit of small particle size, since we consider this limit with all other distances kept fixed (Fig. 2). By contrast, this derivation does not apply to an oblate nanoparticle lying on a substrate. Substituting the expansion of \mathcal{G}_{env} into Eq. (85) and applying the VSWFs on both sides, we obtain the known expression [78]:

$$\mathbf{T} - \mathbf{T}_0 = \mathbf{T}\mathbf{G}_{\text{env}}\mathbf{T}_0. \quad (86)$$

Taking the limit of small size corresponds to considering only the blocks $\overline{\mathbf{T}}_{11}^{22}$, $\overline{\mathbf{T}}_{0,11}^{22}$, $\overline{\mathbf{G}}_{\text{env},11}^{22}$ [cf. Eq. (78)]. Due to $\overline{\mathbf{G}}_{\text{env}}(\mathbf{r}, \mathbf{r}')$ being almost constant inside the particle, we obtain

$$\overline{\mathbf{G}}_{\text{env}}(\mathbf{r}_0, \mathbf{r}_0) \cong ik \sum_{mm'} \left[G_{\text{env},1m1m'}^{22} \text{Rg}\mathbf{N}_{1m}(k\mathbf{r}) \otimes \text{Rg}\mathbf{N}_{1m'}^*(k\mathbf{r}') \right] \cong \frac{ik}{6\pi} \overline{\mathbf{Z}} \cdot \overline{\mathbf{G}}_{\text{env},11}^{22} \cdot \overline{\mathbf{Z}}^H, \quad (87)$$

which together with Eqs. (79) and (86) leads to the well-known relation [80,81]:

$$\overline{\boldsymbol{\alpha}} - \overline{\boldsymbol{\alpha}}_0 = \omega^2 \mu_0 \overline{\boldsymbol{\alpha}} \cdot \overline{\mathbf{G}}_{\text{env}}(\mathbf{r}_0, \mathbf{r}_0) \cdot \overline{\boldsymbol{\alpha}}_0, \quad (88)$$

where $\overline{\boldsymbol{\alpha}}_0$ is the polarizability in free space (given by Eq. (79) applied to $\overline{\mathbf{T}}_{0,11}^{22}$).

5. Conclusion

The seemingly simple concept of an electric dipole conceals pitfalls which may result in ambiguous expressions for the measurable quantities. We aimed to provide the description with minimal empirical assumptions and from as many points of view as possible. In the first part, we reviewed the results which follow from a point-dipole abstraction – a singular (infinitesimal) point in space whose sole ability is to be polarized due to the external electric field. It was shown that two existing approaches to obtain a relation between the static (bare) $\overline{\boldsymbol{\chi}}$ and the dynamic (dressed) $\overline{\boldsymbol{\alpha}}$ polarizabilities are asymptotically equivalent. This equivalence is defined as the one valid for any $\overline{\boldsymbol{\chi}}$, including purely real one (corresponding to a non-absorbing dipole), which explains the significance of the radiative correction as the main term of $\overline{\boldsymbol{\alpha}}^I$. We revisited classic results for absorption and emission by the integration of the Poynting vector over a surface enclosing the point dipole, thereby avoiding the singularity, in the most general case of inhomogeneous and not necessarily reciprocal environment. We maintained this generality throughout the review. As the main result of this part, we introduced the source Green’s dyadic $\overline{\mathbf{G}}_{\text{src}}(\mathbf{r}, \mathbf{r}')$ that describes the contribution of a point dipole into the electromagnetic properties of the whole space. In the same time, it serves as a single-equation unambiguous definition of the point dipole and all its optical properties. In particular, it allows one to rigorously derive the fluctuation–dissipation theorem (FDT) in terms of the fluctuating dipole moment \mathbf{p}_{fl} , resolving the existing confusion in the literature.

In the second part of this review, we discuss the most common microscopic model of a point dipole – a small sphere. In contrast to the phenomenological point dipole, this model implicitly includes the internal polarization $\mathbf{P}(\mathbf{r})$ and/or electric field $\mathbf{E}(\mathbf{r})$ inside the particle. As a result, the well-known Lorenz–Mie theory provides a completely rigorous theoretical description without any empirical assumptions. In particular, we showed that two definitions of the dipole moment, namely one with respect to the far-field scattering and another as the integral of $\mathbf{P}(\mathbf{r})$, are exactly the same. However, the usual expressions of the two for a small sphere are only asymptotically equivalent, while the second-order in a corrections are different. This result explains why there are several expressions for non-radiative correction in the literature. Next, we derived the FDT for \mathbf{p}_{fl} from the microscopic FDT for the fluctuating currents \mathbf{J}_{fl} inside the sphere and the definition of \mathbf{p}_{fl} through $\mathbf{E}(\mathbf{r})$. The result is equivalent to that for the point dipole, but avoids explicit consideration of the thermal equilibrium.

The third part of this review is devoted to the generalization of the microscopic description to small particles of arbitrary shape. It is done using the volume-integral-equation (VIE) formalism and the integral-operator calculus. Both $\bar{\chi}$ and $\bar{\alpha}$ are defined as the double integrals of the corresponding dyadic transition operators over the particle's volume. Interestingly, we obtained the general operator form of the radiative and non-radiative (finite-size) corrections to the static polarizability. This result agrees with recent papers where the radiative corrections were introduced in terms of the T-matrix. Moreover, we showed a general connection between the definitions of the polarizability through the VIE and the T-matrix approach. We also derived a rigorous expansion of $\bar{\alpha}^{-1}$ into powers of a . The leading $\mathcal{O}(a^{-3})$ term equals to $\bar{\chi}^{-1}$, $\mathcal{O}(a^{-2})$ term is absent, $\mathcal{O}(a^{-1})$ term corresponds to the non-radiative correction, and $\mathcal{O}(1)$ term, providing radiative correction, is completely independent of the particle. The latter has been previously conjectured in the literature. Finally, we re-derived the FDT in the most general (non-magnetic) case of a small particle with arbitrary shape, internal structure, and tensorial permittivity in an arbitrary environment.

Finally, all the derivations are made rigorous and detailed with the help of [Appendices A–F](#), some of which may be useful in other applications as well. For instance, we derived general algebraic properties concerning the decomposition of operators into Hermitian and skew-Hermitian parts (A^R and iA^I , respectively) and represented the I -part of the free-space Green's dyadic, $\bar{G}_0^I(\mathbf{r}, \mathbf{r}')$, in terms of the regular vector spherical wave functions. The latter should simplify the consideration of the scattering or emission problems for both point dipoles and finite particles. Overall, the review systemizes and pushes forward the understanding of a point electric dipole, and, thus, advances the theory of electromagnetic scattering by small particles.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Free-space dyadic Green's function and static limit

The free-space electric Green's dyadic is [19]:

$$\bar{G}_0(\mathbf{r}, \mathbf{r}') \stackrel{\text{def}}{=} \frac{\exp(ikR)}{4\pi R} \left[\left(\bar{\mathbf{I}} - \frac{\mathbf{R} \otimes \mathbf{R}}{R^2} \right) + \frac{ikR - 1}{k^2 R^2} \left(\bar{\mathbf{I}} - 3 \frac{\mathbf{R} \otimes \mathbf{R}}{R^2} \right) \right], \quad (\text{A.1})$$

where $\mathbf{R} = \mathbf{r} - \mathbf{r}'$, $R = |\mathbf{R}|$, $\bar{\mathbf{I}}$ is the identity dyadic, k is the wavenumber in the medium. The static limit of (A.1) is

$$\bar{G}_{\text{st}}(\mathbf{r}, \mathbf{r}') \stackrel{\text{def}}{=} \frac{1}{k^2} \lim_{kR \rightarrow 0} k^2 \bar{G}_0(\mathbf{r}, \mathbf{r}') = -\frac{1}{4\pi k^2 R^3} \left(\bar{\mathbf{I}} - 3 \frac{\mathbf{R} \otimes \mathbf{R}}{R^2} \right) = -\frac{1}{4\pi k^2} \nabla \otimes \frac{\mathbf{R}}{R^3}, \quad (\text{A.2})$$

where the remaining dependence of \bar{G}_{st} on k is an artefact of the used system of units. It is absent if the Gaussian system is used, e.g., [14]. It is straightforward to show that

$$\bar{G}_0(\mathbf{r}, \mathbf{r}') - \bar{G}_{\text{st}}(\mathbf{r}, \mathbf{r}') = \frac{1}{8\pi R} \left(\bar{\mathbf{I}} + \frac{\mathbf{R} \otimes \mathbf{R}}{R^2} \right) + \frac{k\bar{\mathbf{I}}}{6\pi} + \mathcal{O}(k^2 R). \quad (\text{A.3})$$

The free-space magnetic Green's dyadic [55], relating the magnetic field with the point source current (cf. Eq. (B.1) below), is:

$$\bar{G}_0^m(\mathbf{r}, \mathbf{r}') \stackrel{\text{def}}{=} \nabla \times \bar{G}_0(\mathbf{r}, \mathbf{r}') = (ikR - 1) \frac{\exp(ikR)}{4\pi R^3} \mathbf{R} \times \bar{\mathbf{I}}, \quad (\text{A.4})$$

which static limit is

$$\bar{G}_{\text{st}}^m(\mathbf{r}, \mathbf{r}') \stackrel{\text{def}}{=} \lim_{kR \rightarrow 0} \bar{G}_0^m(\mathbf{r}, \mathbf{r}') = -\frac{\mathbf{R} \times \bar{\mathbf{I}}}{4\pi R^3}. \quad (\text{A.5})$$

Note that this limit is not equal to $\nabla \times \bar{G}_{\text{st}}(\mathbf{r}, \mathbf{r}')$, since the latter equals $\bar{\mathbf{0}}$.

Appendix B. Integration of the poynting vector around a dipole

The magnetic field can be divided into two parts similarly to the electric field in Eq. (4):

$$\mathbf{H}(\mathbf{r}) = \frac{\nabla \times \mathbf{E}(\mathbf{r})}{i\omega\mu_0} = \left[\nabla \times \bar{G}_{\text{src}}(\mathbf{r}, \mathbf{r}') \right] \cdot \mathbf{J}_{\text{src}} = \mathbf{H}_0(\mathbf{r}) + \mathbf{H}_{\text{sca}}(\mathbf{r}). \quad (\text{B.1})$$

The standard decomposition of $\mathbf{S}(\mathbf{r})$ is then [20]:

$$\begin{aligned} \mathbf{S}(\mathbf{r}) &= \frac{1}{2} \text{Re} [\mathbf{E}(\mathbf{r}) \times \mathbf{H}^*(\mathbf{r})] = \mathbf{S}_0(\mathbf{r}) + \mathbf{S}_{\text{sca}}(\mathbf{r}) + \mathbf{S}_{\text{ext}}(\mathbf{r}), \\ \mathbf{S}_0(\mathbf{r}) &\stackrel{\text{def}}{=} \frac{1}{2} \text{Re} [\mathbf{E}_0(\mathbf{r}) \times \mathbf{H}_0^*(\mathbf{r})], \quad \mathbf{S}_{\text{sca}}(\mathbf{r}) \stackrel{\text{def}}{=} \frac{1}{2} \text{Re} [\mathbf{E}_{\text{sca}}(\mathbf{r}) \times \mathbf{H}_{\text{sca}}^*(\mathbf{r})], \\ \mathbf{S}_{\text{ext}}(\mathbf{r}) &\stackrel{\text{def}}{=} \frac{1}{2} \text{Re} [\mathbf{E}_0(\mathbf{r}) \times \mathbf{H}_{\text{sca}}^*(\mathbf{r}) + \mathbf{E}_{\text{sca}}(\mathbf{r}) \times \mathbf{H}_0^*(\mathbf{r})]. \end{aligned} \quad (\text{B.2})$$

First, we integrate two components of $\mathbf{S}_{\text{ext}}(\mathbf{r})$ analogous to Eqs. (25) and (28) in [19]:

$$\begin{aligned} \oint_A d\mathbf{A} \cdot [\mathbf{E}_0(\mathbf{r}) \times \mathbf{H}_{\text{sca}}^*(\mathbf{r})] &\xrightarrow{R_0 \rightarrow 0} i\omega \oint_A d\mathbf{A} \cdot \left\{ \mathbf{E}_0 \times \left[\nabla \times \overline{\mathbf{G}}_0^*(\mathbf{r}, \mathbf{r}_0) \cdot \mathbf{p}^* \right] \right\} \\ &\xrightarrow{R_0 \rightarrow 0} -\frac{i\omega}{4\pi R_0^2} \oint_A d\mathbf{A} \cdot [\mathbf{E}_0 \times (\mathbf{n} \times \mathbf{p}^*)] = -\frac{2}{3} i\omega \mathbf{E}_0 \cdot \mathbf{p}^*, \end{aligned} \quad (\text{B.3})$$

where we used Eqs. (B.1), (A.5) and the smoothness of $\overline{\mathbf{G}}_{\text{env}}(\mathbf{r}, \mathbf{r}_0)$, which implies that its contribution to the integral is negligible.

$$\begin{aligned} \oint_A d\mathbf{A} \cdot [\mathbf{E}_{\text{sca}}(\mathbf{r}) \times \mathbf{H}_0^*(\mathbf{r})] &= -\oint_A d^2\mathbf{r} [\mathbf{n} \times \mathbf{H}_0^*(\mathbf{r})] \cdot \mathbf{E}_{\text{sca}}(\mathbf{r}) \xrightarrow{R_0 \rightarrow 0} -\omega^2 \mu_0 \oint_A d^2\mathbf{r} [\mathbf{n} \times \mathbf{H}_0^*(\mathbf{r})] \cdot \overline{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}_0) \cdot \mathbf{p} \\ &\xrightarrow{R_0 \rightarrow 0} \frac{1}{\epsilon_0} \oint_A d^2\mathbf{r} [\mathbf{n} \times \mathbf{H}_0^*(\mathbf{r})] \cdot \left(\nabla \otimes \frac{\mathbf{R}_0}{4\pi R_0^3} \right) \cdot \mathbf{p} = \frac{1}{\epsilon_0} \oint_A d^2\mathbf{r} [\nabla \times \mathbf{H}_0^*(\mathbf{r})] \cdot \left(\frac{\mathbf{n} \otimes \mathbf{R}}{4\pi R_0^3} \right) \cdot \mathbf{p} = \frac{i\omega}{3} \mathbf{p} \cdot \mathbf{E}_0^*, \end{aligned} \quad (\text{B.4})$$

where we used Eqs. (4), (A.2) and interchanged ∇ with \mathbf{n} , due to Eq. (29) in [19]. Note that it is not possible to replace $\mathbf{H}_0(\mathbf{r})$ by $\mathbf{H}_0(\mathbf{r}_0)$ in Eq. (B.4), due to the strong singularity of $\mathbf{E}_{\text{sca}}(\mathbf{r})$. Combining Eqs. (15), (B.3), (B.4) yields:

$$W_{\text{ext}} = -\frac{\omega}{2} \text{Im}(\mathbf{E}_0 \cdot \mathbf{p}^*). \quad (\text{B.5})$$

Next, we integrate $\mathbf{S}_{\text{sca}}(\mathbf{r})$, for that we decompose

$$\mathbf{E}_{\text{sca}}(\mathbf{r}) = \omega^2 \mu_0 \left[\overline{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}_0) + \overline{\mathbf{G}}_{\text{env}}(\mathbf{r}, \mathbf{r}_0) \right] \cdot \mathbf{p} \quad (\text{B.6})$$

and integrate four combinations of $\overline{\mathbf{G}}_0$ and $\overline{\mathbf{G}}_{\text{env}}$ separately. Contribution from the free-space dyadics can be integrated explicitly for any R_0 (see Eq. (26) of [20]):

$$\begin{aligned} &\oint_A d^2\mathbf{A} \cdot \left\{ \left[\overline{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}_0) \cdot \mathbf{p} \right] \times \left[\nabla \times \overline{\mathbf{G}}_0^*(\mathbf{r}, \mathbf{r}_0) \cdot \mathbf{p}^* \right] \right\} \\ &= \frac{1}{(4\pi)^2 R^3} \left(1 + \frac{ikR-1}{k^2 R^2} \right) (ikR+1) \oint_A d^2\mathbf{r} [\mathbf{n} \times \mathbf{p}] \cdot [\mathbf{n} \times \mathbf{p}^*] = \frac{1}{6\pi} \left(\frac{1}{k^2 R^3} + ik \right) |\mathbf{p}|^2. \end{aligned} \quad (\text{B.7})$$

The cross-terms are evaluated analogously to Eqs. (B.3), (B.4):

$$\oint_A d^2\mathbf{A} \cdot \left\{ \left[\overline{\mathbf{G}}_{\text{env}}(\mathbf{r}, \mathbf{r}_0) \cdot \mathbf{p} \right] \times \left[\nabla \times \overline{\mathbf{G}}_0^*(\mathbf{r}, \mathbf{r}_0) \cdot \mathbf{p}^* \right] \right\} \xrightarrow{R_0 \rightarrow 0} -\frac{2}{3} \mathbf{p}^* \cdot \overline{\mathbf{G}}_{\text{env}}(\mathbf{r}_0, \mathbf{r}_0) \cdot \mathbf{p}, \quad (\text{B.8})$$

$$\oint_A d^2\mathbf{A} \cdot \left\{ \left[\overline{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}_0) \cdot \mathbf{p} \right] \times \left[\nabla \times \overline{\mathbf{G}}_{\text{env}}^*(\mathbf{r}, \mathbf{r}_0) \cdot \mathbf{p}^* \right] \right\} \xrightarrow{R_0 \rightarrow 0} \frac{1}{3} \mathbf{p} \cdot \overline{\mathbf{G}}_{\text{env}}^*(\mathbf{r}_0, \mathbf{r}_0) \cdot \mathbf{p}^*, \quad (\text{B.9})$$

The term containing only $\overline{\mathbf{G}}_{\text{env}}$ vanish, since it is an energy flow over the closed surface for the field, which does not have sources inside this surface (analogously to \mathbf{S}_0). Combining Eqs. (15), (B.6)–(B.9) we obtain:

$$W_{\text{sca}} = \frac{\omega^3 \mu_0}{2} \left[\frac{k}{6\pi} |\mathbf{p}|^2 + \text{Im} \left(\mathbf{p}^* \cdot \overline{\mathbf{G}}_{\text{env}}(\mathbf{r}_0, \mathbf{r}_0) \cdot \mathbf{p} \right) \right] = \frac{\omega^2 \mu_0}{2} \mathbf{p}^* \cdot \overline{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \mathbf{p}, \quad (\text{B.10})$$

using Eqs. (12), (E.7).

Finally, we note that the derivations in this section can be made shorter by decomposing the total field into the regular and singular parts from the beginning:

$$\mathbf{E}(\mathbf{r}) = \left[\mathbf{E}_0(\mathbf{r}) + \omega^2 \mu_0 \overline{\mathbf{G}}_{\text{env}}(\mathbf{r}, \mathbf{r}_0) \cdot \mathbf{p} \right] + \omega^2 \mu_0 \overline{\mathbf{G}}_{\text{env}}(\mathbf{r}, \mathbf{r}_0) \cdot \mathbf{p} \quad (\text{B.11})$$

at the expense of less trivial combination of the corresponding integrals into W_{ext} and W_{sca} .

Appendix C. Derivation of the FDT for a point dipole

As explained after Eq. (28), we expand the total field as

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_b(\mathbf{r}) + \mathbf{E}_{\text{sca}}(\mathbf{r}) + \mathbf{E}_{\text{em}}(\mathbf{r}), \quad (\text{C.1})$$

$$\mathbf{E}_{\text{sca}}(\mathbf{r}) = \omega^2 \mu_0 \overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) \cdot \overline{\boldsymbol{\alpha}} \cdot \mathbf{E}_b(\mathbf{r}_0), \quad \mathbf{E}_{\text{em}}(\mathbf{r}) = \omega^2 \mu_0 \overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) \cdot \mathbf{p}_{\text{fl}}, \quad (\text{C.2})$$

similar to Eq. (4). Then we correlate it with itself:

$$\begin{aligned} &\langle \mathbf{E}(\mathbf{r}) \otimes \mathbf{E}^*(\mathbf{r}') \rangle \\ &= \langle \mathbf{E}_b(\mathbf{r}) \otimes \mathbf{E}_b^*(\mathbf{r}') \rangle + \langle \mathbf{E}_b(\mathbf{r}) \otimes \mathbf{E}_{\text{sca}}^*(\mathbf{r}') \rangle + \langle \mathbf{E}_{\text{sca}}(\mathbf{r}) \otimes \mathbf{E}_b^*(\mathbf{r}') \rangle + \langle \mathbf{E}_{\text{sca}}(\mathbf{r}) \otimes \mathbf{E}_{\text{sca}}^*(\mathbf{r}') \rangle + \langle \mathbf{E}_{\text{em}}(\mathbf{r}) \otimes \mathbf{E}_{\text{em}}^*(\mathbf{r}') \rangle, \end{aligned} \quad (\text{C.3})$$

where the other four terms vanish due to the statistical independence of \mathbf{E}_b and \mathbf{E}_{em} . The first two correlations in Eq. (2) are given by Eqs. (28) and (23), respectively, and are related through the identity

$$\overline{\mathbf{G}}_{\text{src}}^I(\mathbf{r}, \mathbf{r}') = \overline{\mathbf{G}}^I(\mathbf{r}, \mathbf{r}') + \frac{1}{2i} \left[\overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) \cdot \overline{\boldsymbol{\alpha}} \cdot \overline{\mathbf{G}}(\mathbf{r}_0, \mathbf{r}') - \overline{\mathbf{G}}^H(\mathbf{r}, \mathbf{r}_0) \cdot \overline{\boldsymbol{\alpha}}^H \cdot \overline{\mathbf{G}}^H(\mathbf{r}_0, \mathbf{r}') \right], \quad (\text{C.4})$$

where H immediately after $\bar{\mathbf{G}}$ denotes the adjoint of the operator before considering the specific arguments, i.e., $\bar{\mathbf{G}}^H(\mathbf{r}, \mathbf{r}') = [\bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}')]^H$ [Eq. (E.3)]. Let us express the remaining four terms using the dyadic identities

$$(\bar{\mathbf{A}} \cdot \mathbf{a}) \otimes \mathbf{b} = \bar{\mathbf{A}} \cdot (\mathbf{a} \otimes \mathbf{b}), \quad \mathbf{a} \otimes (\bar{\mathbf{A}} \cdot \mathbf{b})^* = (\mathbf{a} \otimes \mathbf{b}) \cdot \bar{\mathbf{A}}. \quad (\text{C.5})$$

We obtain:

$$\langle \mathbf{E}_b(\mathbf{r}) \otimes \mathbf{E}_{\text{sca}}^*(\mathbf{r}') \rangle = \omega^2 \mu_0 \langle \mathbf{E}_b(\mathbf{r}) \otimes \mathbf{E}_b^*(\mathbf{r}_0) \rangle \cdot \bar{\boldsymbol{\alpha}}^H \cdot \bar{\mathbf{G}}^H(\mathbf{r}_0, \mathbf{r}'), \quad (\text{C.6})$$

$$\langle \mathbf{E}_{\text{sca}}(\mathbf{r}) \otimes \mathbf{E}_b^*(\mathbf{r}') \rangle = \omega^2 \mu_0 \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) \cdot \bar{\boldsymbol{\alpha}} \cdot \langle \mathbf{E}_b(\mathbf{r}_0) \otimes \mathbf{E}_b^*(\mathbf{r}') \rangle, \quad (\text{C.7})$$

$$\langle \mathbf{E}_{\text{sca}}(\mathbf{r}) \otimes \mathbf{E}_{\text{sca}}^*(\mathbf{r}') \rangle = \omega^4 \mu_0^2 \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) \cdot \bar{\boldsymbol{\alpha}} \cdot \langle \mathbf{E}_b(\mathbf{r}_0) \otimes \mathbf{E}_b^*(\mathbf{r}_0) \rangle \cdot \bar{\boldsymbol{\alpha}}^H \cdot \bar{\mathbf{G}}^H(\mathbf{r}_0, \mathbf{r}'), \quad (\text{C.8})$$

$$\langle \mathbf{E}_{\text{em}}(\mathbf{r}) \otimes \mathbf{E}_{\text{em}}^*(\mathbf{r}') \rangle = \omega^4 \mu_0^2 \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) \cdot \langle \mathbf{p}_{\text{fl}} \otimes \mathbf{p}_{\text{fl}}^* \rangle \cdot \bar{\mathbf{G}}^H(\mathbf{r}_0, \mathbf{r}'), \quad (\text{C.9})$$

Combining it all into Eq. (C.3), using again Eq. (23) and reducing the common factor $\omega^3 \mu_0^2 \Theta(\omega, T)/\pi$, we obtain

$$\begin{aligned} & \frac{1}{2i} \left\{ \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) \cdot \bar{\boldsymbol{\alpha}} \cdot \bar{\mathbf{G}}(\mathbf{r}_0, \mathbf{r}') - \bar{\mathbf{G}}^H(\mathbf{r}, \mathbf{r}_0) \cdot \bar{\boldsymbol{\alpha}}^H \cdot \bar{\mathbf{G}}^H(\mathbf{r}_0, \mathbf{r}') - \left[\bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) - \bar{\mathbf{G}}^H(\mathbf{r}, \mathbf{r}_0) \right] \cdot \bar{\boldsymbol{\alpha}}^H \cdot \bar{\mathbf{G}}^H(\mathbf{r}_0, \mathbf{r}') \right. \\ & \quad \left. - \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) \cdot \bar{\boldsymbol{\alpha}} \cdot \left[\bar{\mathbf{G}}(\mathbf{r}_0, \mathbf{r}') - \bar{\mathbf{G}}^H(\mathbf{r}_0, \mathbf{r}') \right] \right\} \\ & = \omega^2 \mu_0 \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) \cdot \bar{\boldsymbol{\alpha}} \cdot \bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \bar{\boldsymbol{\alpha}}^H \cdot \bar{\mathbf{G}}^H(\mathbf{r}_0, \mathbf{r}') + \frac{\pi\omega}{\Theta(\omega, T)} \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) \cdot \langle \mathbf{p}_{\text{fl}} \otimes \mathbf{p}_{\text{fl}}^* \rangle \cdot \bar{\mathbf{G}}^H(\mathbf{r}_0, \mathbf{r}'), \end{aligned} \quad (\text{C.10})$$

which is further transformed into:

$$\bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0) \cdot \left[\frac{\pi\omega}{\Theta(\omega, T)} \langle \mathbf{p}_{\text{fl}} \otimes \mathbf{p}_{\text{fl}}^* \rangle - \frac{1}{2i} (\bar{\boldsymbol{\alpha}} - \bar{\boldsymbol{\alpha}}^H) + \omega^2 \mu_0 \bar{\boldsymbol{\alpha}} \cdot \bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \bar{\boldsymbol{\alpha}}^H \right] \cdot \bar{\mathbf{G}}^H(\mathbf{r}_0, \mathbf{r}') = \bar{\mathbf{0}}. \quad (\text{C.11})$$

Since Eq. (C.11) is valid for any \mathbf{r}, \mathbf{r}' , the expression in square brackets must be identically zero, which leads to Eq. (29). Note that $\bar{\boldsymbol{\alpha}} \cdot \bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \bar{\boldsymbol{\alpha}}^H = \bar{\boldsymbol{\alpha}}^H \cdot \bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0) \cdot \bar{\boldsymbol{\alpha}}$ due to the Hermitian property of the dyadic $\bar{\mathbf{G}}^I(\mathbf{r}_0, \mathbf{r}_0)$.

Appendix D. Vector spherical wave functions

In this appendix we recall some properties of the spherical functions, largely following Appendices A and C of [25]. The *scalar spherical harmonics* (orthonormal on a unit sphere) are given as:

$$Y_{nm}(\hat{\mathbf{r}}) \stackrel{\text{def}}{=} \sqrt{\frac{(2n+1)(n-m)!}{4\pi(n+m)!}} P_n^m(\cos\theta) \exp(im\varphi), \quad (\text{D.1})$$

where $n \geq 0$, $-n \leq m \leq n$, the hat denotes unit vectors, i.e., $\hat{\mathbf{r}} \stackrel{\text{def}}{=} \mathbf{r}/r$, θ , φ are the spherical angles and P_n^m is the associated Legendre polynomial. When multiplied with the spherical Hankel and Bessel functions, $h_n^{(1)}(kr)$ and $j_n(kr)$, $Y_{nm}(\hat{\mathbf{r}})$ leads (up to a coefficient) to the *scalar spherical wave functions* ψ_{nm} and $\text{Rg}\psi_{nm}$, respectively. Both are the solutions to the scalar Helmholtz equation, but *outgoing* ψ_{nm} satisfies the Sommerfeld radiation condition at infinity, while *regular* $\text{Rg}\psi_{nm}$ is finite when $kr \rightarrow 0$. In particular, we use the not-normalized functions

$$\begin{pmatrix} \psi_{nm}(k\mathbf{r}) \\ \text{Rg}\psi_{nm}(k\mathbf{r}) \end{pmatrix} \stackrel{\text{def}}{=} \begin{pmatrix} h_n^{(1)}(kr) \\ j_n(kr) \end{pmatrix} P_n^m(\cos\theta) \exp(im\varphi). \quad (\text{D.2})$$

While we follow the notation of [25], we interchange the order of indices n and m to ψ and other functions below as in [58], since n is the primary index.

The VSWFs are the solutions to the vector Helmholtz equation. The only ones which are divergence-free (suitable for the expansion of electric fields) are

$$\begin{pmatrix} \mathbf{M}_{nm}(k\mathbf{r}) \\ \text{Rg}\mathbf{M}_{nm}(k\mathbf{r}) \end{pmatrix} \stackrel{\text{def}}{=} \gamma_{nm} \nabla \times \left[\mathbf{r} \begin{pmatrix} \psi_{nm}(k\mathbf{r}) \\ \text{Rg}\psi_{nm}(k\mathbf{r}) \end{pmatrix} \right], \quad \begin{pmatrix} \mathbf{N}_{nm}(k\mathbf{r}) \\ \text{Rg}\mathbf{N}_{nm}(k\mathbf{r}) \end{pmatrix} \stackrel{\text{def}}{=} \frac{1}{k} \nabla \times \left[\begin{pmatrix} \mathbf{M}_{nm}(k\mathbf{r}) \\ \text{Rg}\mathbf{M}_{nm}(k\mathbf{r}) \end{pmatrix} \right], \quad (\text{D.3})$$

where $n \geq 1$ and the normalization coefficient is similar to that in Eq. (D.1):

$$\gamma_{nm} \stackrel{\text{def}}{=} \sqrt{\frac{(2n+1)(n-m)!}{4\pi n(n+1)(n+m)!}}. \quad (\text{D.4})$$

The outgoing VSWFs satisfies the Sommerfeld radiation condition and are, thus, a complete basis for the expansion of the scattered electric fields, while the regular ones — for the electric fields inside a particle.

The alternative expressions for the VSWFs are:

$$\begin{aligned} \mathbf{M}_{nm}(k\mathbf{r}) &= \gamma_{nm} \frac{h_n^{(1)}(kr)}{kr} \mathbf{C}_{nm}(\hat{\mathbf{r}}), \\ \mathbf{N}_{nm}(k\mathbf{r}) &= \gamma_{nm} \left[n(n+1) \frac{h_n^{(1)}(kr)}{kr} \mathbf{P}_{nm}(\hat{\mathbf{r}}) + \frac{1}{kr} \frac{d}{d\epsilon} \left[r h_n^{(1)}(kr) \right] \mathbf{B}_{nm}(\hat{\mathbf{r}}) \right], \end{aligned} \quad (\text{D.5})$$

and the regular ones are obtained by replacing $h_n^{(1)}(kr)$ by $j_n(kr)$. Here $\mathbf{C}_{nm}(\hat{\mathbf{r}})$, $\mathbf{B}_{nm}(\hat{\mathbf{r}})$, $\mathbf{P}_{nm}(\hat{\mathbf{r}})$ are the not-normalized *vector spherical harmonics* [25] (one of the existing conventions). We do not provide the specific expressions here, but they are orthogonal between

different types and different indices. Moreover, it can be shown (pp. 1898–1899 of [82]) that $\mathbf{P}_{nm}(\hat{\mathbf{r}})$ and $\mathbf{B}_{nm}(\hat{\mathbf{r}})$ are linear combinations of $Y_{n'm'}(\hat{\mathbf{r}})$ with $n' = n \pm 1$ and $|m' - m| \leq 1$, while $\mathbf{C}_{nm}(\hat{\mathbf{r}})$ is a linear combination of $Y_{nm'}(\hat{\mathbf{r}})$. This together with orthogonality of $Y_{nm'}(\hat{\mathbf{r}})$ implies

$$\begin{aligned} \forall n \geq 1: \oint d^2\mathbf{r} \mathbf{M}_{nm}(\mathbf{r}) &= \oint d^2\mathbf{r} \text{Rg}\mathbf{M}_{nm}(\mathbf{r}) = 0, \\ \forall n \geq 2: \oint d^2\mathbf{r} \mathbf{N}_{nm}(\mathbf{r}) &= \oint d^2\mathbf{r} \text{Rg}\mathbf{N}_{nm}(\mathbf{r}) = 0, \end{aligned} \quad (\text{D.6})$$

where we used that the integration without a multiplier is equivalent to the integration with $Y_{00}(\hat{\mathbf{r}})$. In other words, only $\mathbf{N}_{1m}(k\mathbf{r})$ and $\text{Rg}\mathbf{N}_{1m}(k\mathbf{r})$ have non-zero integral over any spherical surface.

The basis consisting of $\text{Rg}\mathbf{N}_{nm}$ and $\text{Rg}\mathbf{M}_{nm}$ is orthogonal, i.e., all cross terms (inner products — see Eq. (E.2)) over any spherical surface and, hence, over the volume of any sphere are exactly zero. This holds even for different wave vectors (see below), since this changes only the radial functions, while spherical harmonics stay orthogonal. Using the normalization of the spherical harmonics [25]

$$n(n+1) \oint d^2\hat{\mathbf{r}} \mathbf{P}_{nm}^*(\hat{\mathbf{r}}) \cdot \mathbf{P}_{nm}(\hat{\mathbf{r}}) = \oint d^2\hat{\mathbf{r}} \mathbf{B}_{nm}^*(\hat{\mathbf{r}}) \cdot \mathbf{B}_{nm}(\hat{\mathbf{r}}) = \frac{1}{\gamma_{nm}^2}, \quad (\text{D.7})$$

we obtain the inner product of functions $\text{Rg}\mathbf{N}_{nm}$ with the same spherical indices but different wave vectors:

$$\begin{aligned} \int_{r \leq a} d^3\mathbf{r} \text{Rg}\mathbf{N}_{nm}^*(k\mathbf{r}) \cdot \text{Rg}\mathbf{N}_{nm}(m_0k\mathbf{r}) &= \frac{1}{m_0k^3} \int_0^x dz \left\{ n(n+1) j_n(z) j_n(m_0z) + [z j_n(z)]' [z j_n(m_0z)]' \right\} \\ &= \frac{m_0 j_n(m_0x) [x j_n(x)]' - j_n(x) [x j_n(m_0x)]'}{m_0(m_0^2 - 1)k^3} = \frac{i}{(m_0^2 - 1)k^3} \frac{a_n}{d_n}, \end{aligned} \quad (\text{D.8})$$

where $z \stackrel{\text{def}}{=} kr$, $x \stackrel{\text{def}}{=} ka$, k is assumed to be real, and m_0 can be complex. The primes denote differentiation with respect to either z or x , and the last part is expressed through the Lorenz–Mie coefficients for a sphere with the size parameter x and the refractive index m_0 [49]. The simplest way to verify the specified antiderivative in Eq. (D.8) is through the differential equation for the Riccati–Bessel function $x j_n(x)$ [83]. For $\text{Rg}\mathbf{M}_{nm}$ similar simple relations can only be derived with the additional weighting factor r^2 in the integral, then it is proportional to b_n/c_n .

Eq. (D.4) also implies that

$$\text{Rg}\mathbf{N}_{nm}(k\mathbf{r}) = \mathcal{O}(r^{n-1}), \quad \text{Rg}\mathbf{M}_{nm}(k\mathbf{r}) = \mathcal{O}(r^n), \quad (\text{D.9})$$

i.e., of all regular divergence-free VSWFs only a set of $\text{Rg}\mathbf{N}_{1m}(k\mathbf{r})$ does not vanish when $kr \rightarrow 0$ and is given by:

$$\begin{aligned} \text{Rg}\mathbf{N}_{10}(k\mathbf{r}) &= \sqrt{\frac{3}{8\pi}} \left\{ 2 \frac{j_1(kr)}{kr} \cos\theta \hat{\mathbf{e}}_r - \left[j_0(kr) - \frac{j_1(kr)}{kr} \right] \sin\theta \hat{\mathbf{e}}_\theta \right\} = \frac{\hat{\mathbf{e}}_z}{\sqrt{6\pi}} + \mathcal{O}[(kr)^2], \\ \text{Rg}\mathbf{N}_{1,\pm 1}(k\mathbf{r}) &= \mp \sqrt{\frac{3}{16\pi}} \left\{ 2 \frac{j_1(kr)}{kr} \sin\theta \hat{\mathbf{e}}_r + \left[j_0(kr) - \frac{j_1(kr)}{kr} \right] (\cos\theta \hat{\mathbf{e}}_\theta \pm \hat{\mathbf{e}}_\varphi) \right\} \exp(\pm i\varphi) = \frac{\mp \hat{\mathbf{e}}_x - i \hat{\mathbf{e}}_y}{2\sqrt{3\pi}} + \mathcal{O}[(kr)^2], \end{aligned} \quad (\text{D.10})$$

where $\hat{\mathbf{e}}$ are the unit basic vectors for the spherical and Cartesian coordinate systems. When integrated over the unit sphere, the terms with $j_1(kr)$ vanish, allowing easy averaging of the VSWFs over a sphere of arbitrary radius:

$$\begin{aligned} \int_{r \leq a} d^3\mathbf{r} \text{Rg}\mathbf{N}_{10}(k\mathbf{r}) &= 3 \frac{j_1(ka)}{ka} \frac{\hat{\mathbf{e}}_z}{\sqrt{6\pi}}, \\ \int_{r \leq a} d^3\mathbf{r} \text{Rg}\mathbf{N}_{1,\pm 1}(k\mathbf{r}) &= 3 \frac{j_1(ka)}{ka} \frac{\mp \hat{\mathbf{e}}_x - i \hat{\mathbf{e}}_y}{2\sqrt{3\pi}}. \end{aligned} \quad (\text{D.11})$$

Importantly, we can use the set of $\text{Rg}\mathbf{N}_{1m}(k\mathbf{r})$ as a (incomplete) basis, which is asymptotically constant and orthogonal:

$$\begin{aligned} \overline{\mathbf{W}}_s(\mathbf{r}) &\stackrel{\text{def}}{=} \{ \text{Rg}\mathbf{N}_{1,-1}(k\mathbf{r}), \text{Rg}\mathbf{N}_{10}(k\mathbf{r}), \text{Rg}\mathbf{N}_{11}(k\mathbf{r}) \} \cong \frac{1}{\sqrt{6\pi}} \overline{\mathbf{Z}}, \\ \overline{\mathbf{Z}} &\stackrel{\text{def}}{=} \begin{pmatrix} 1/\sqrt{2} & 0 & -1/\sqrt{2} \\ -i/\sqrt{2} & 0 & -i/\sqrt{2} \\ 0 & 1 & 0 \end{pmatrix}, \end{aligned} \quad (\text{D.12})$$

where $\overline{\mathbf{W}}_s(\mathbf{r})$ (“s” denotes spherical) combines basis functions as columns and $\overline{\mathbf{Z}}$ is a unitary (rotation) matrix ($\overline{\mathbf{Z}}^{-1} = \overline{\mathbf{Z}}^H$). The latter would not be needed if we used the notation of the real VSWFs as is common in the Lorenz–Mie theory [8]. Then $\mathbf{N}_{e11}^{(1)}$, $\mathbf{N}_{o11}^{(1)}$, and $\mathbf{N}_{e01}^{(1)}$ are directly proportional to the Cartesian unit vectors (see also Eq. (C47) of [25]).

One of the illustrating features of the VSWFs is the expansion of the free-space Green’s dyadic [25,72]:

$$\overline{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}') = ik \sum_{nm} \left[\mathbf{M}_{nm}(k\mathbf{r}) \otimes \text{Rg}\mathbf{M}_{nm}^*(k\mathbf{r}') + \mathbf{N}_{nm}(k\mathbf{r}) \otimes \text{Rg}\mathbf{N}_{nm}^*(k\mathbf{r}') \right], \quad r > r', \quad (\text{D.13})$$

and the case of $r' > r$ is obtained from $\overline{\mathbf{G}}_0(\mathbf{r}', \mathbf{r}) = \left[\overline{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}') \right]^T$. Taking the imaginary part of Eq. (D.13), we obtain

$$\overline{\mathbf{G}}_0^I(\mathbf{r}, \mathbf{r}') = k \sum_{nm} \left[\text{Rg}\mathbf{M}_{nm}(k\mathbf{r}) \otimes \text{Rg}\mathbf{M}_{nm}^*(k\mathbf{r}') + \text{Rg}\mathbf{N}_{nm}(k\mathbf{r}) \otimes \text{Rg}\mathbf{N}_{nm}^*(k\mathbf{r}') \right], \quad (\text{D.14})$$

using

$$\begin{aligned} & \operatorname{Re} \left[\mathbf{M}_{nm}(k\mathbf{r}) \otimes \operatorname{Rg}\mathbf{M}_{nm}^*(k\mathbf{r}') + \mathbf{M}_{n,-m}(k\mathbf{r}) \otimes \operatorname{Rg}\mathbf{M}_{n,-m}^*(k\mathbf{r}') \right] \\ &= \frac{\mathbf{M}_{nm}(k\mathbf{r}) + (-1)^m \mathbf{M}_{n,-m}^*(k\mathbf{r})}{2} \otimes \operatorname{Rg}\mathbf{M}_{nm}^*(k\mathbf{r}') + \frac{\mathbf{M}_{n,-m}(k\mathbf{r}) + (-1)^m \mathbf{M}_{nm}^*(k\mathbf{r})}{2} \otimes \operatorname{Rg}\mathbf{M}_{n,-m}^*(k\mathbf{r}') \\ &= \operatorname{Rg}\mathbf{M}_{nm}(k\mathbf{r}) \otimes \operatorname{Rg}\mathbf{M}_{nm}^*(k\mathbf{r}') + \operatorname{Rg}\mathbf{M}_{n,-m}(k\mathbf{r}) \otimes \operatorname{Rg}\mathbf{M}_{n,-m}^*(k\mathbf{r}') \end{aligned} \quad (\text{D.15})$$

and the analogous relation for \mathbf{N}_{nm} , which follows from $\operatorname{Re} h_n^{(1)}(x) = j_n(x)$ and the symmetry relations [25]:

$$\gamma_{n,-m} \mathbf{B}_{n,-m}(\hat{\mathbf{r}}) = (-1)^m \gamma_{nm} \mathbf{B}_{nm}^*(\hat{\mathbf{r}}), \quad \operatorname{Rg}\mathbf{M}_{n,-m}(k\mathbf{r}) = (-1)^m \operatorname{Rg}\mathbf{M}_{nm}^*(k\mathbf{r}) \quad (\text{D.16})$$

and the same for \mathbf{C}_{nm} , \mathbf{P}_{nm} and $\operatorname{Rg}\mathbf{N}_{nm}$, respectively. For $r = r'$ Eq. (D.14) follows from continuity on both sides ($r < r'$ and $r > r'$) and from the regularity of $\overline{\mathbf{G}}_0^I$ [Eq. (12)].

Alternatively, Eq. (D.14) can be derived, albeit less rigorously, from this regularity and the requirement of the corresponding operator \mathcal{G}^I to be Hermitian [Eq. (A.5)]. Then one has to remove all irregularity from the outgoing VSWFs, which naturally leaves only the regular ones. Therefore, Eqs. (D.13), (D.14) can be viewed as a justification that $i\overline{\mathbf{G}}_0^I(\mathbf{r}, \mathbf{r}')$ is the regular part of $\overline{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}')$, i.e., Eq. (10). We are not aware of previous occurrence of Eq. (D.14) in the literature.

Appendix E. Integral operators, projectors, and inner product

The following definitions are partly based on [18,84], but with a slightly different notation involving the script font. The general definition of a *dyadic integral operator* \mathcal{A} with the kernel $\overline{\mathbf{A}}(\mathbf{r}, \mathbf{r}')$ is

$$\mathcal{g} = \mathcal{A}f \Leftrightarrow \mathbf{g}(\mathbf{r}) = \int_{\mathbb{R}^3} d^3\mathbf{r}' \overline{\mathbf{A}}(\mathbf{r}, \mathbf{r}') \cdot \mathbf{f}(\mathbf{r}'), \quad (\text{E.1})$$

where f and g are *vector functions* (fields) with coordinate representations $\mathbf{f}(\mathbf{r})$ and $\mathbf{g}(\mathbf{r})$, respectively. Script letters denote operators and elements of a *function space* (for which we reserve the letter \mathcal{H}). The physically meaningful space is the Hilbert one $\mathcal{H} \triangleq L^2(\mathbb{R}^3)^3$, i.e., each field component is square-integrable, which corresponds to the finite energy of the electromagnetic field [85]. Some functions have finite support V , i.e., they are non-zero only inside this volume — they belong to $\mathcal{H}_V \triangleq L^2(V)^3 \subset \mathcal{H}$. The enclosure is due to the equivalence of functions defined on V and functions equal to zero outside of V . Similarly, some operators act inside \mathcal{H}_V (although formally from \mathcal{H} to \mathcal{H}), i.e., their kernels are zero if either argument is outside V (and the integration domain in Eq. (E.1) can be changed to V). The particular operator domain (V or \mathbb{R}^3) is in most cases clear from the context, the distinction becomes important only when the inverse operators are discussed. To simplify further discussion, we define the projector $\mathcal{J}_V: \mathcal{H} \rightarrow \mathcal{H}_V$, which zeroes the function value outside of V . Note also that an infinite plane wave, which is widely used in scattering problems, belongs only to $L_{\text{loc}}^2(\mathbb{R}^3)^3$, i.e., each field component is square-integrable in any finite domain. This is because such wave cannot be generated by finite sources [85]. However, in this review such fields are used only after the projection into \mathcal{H}_V .

Both \mathcal{H} and \mathcal{H}_V have the natural *inner product*:

$$\langle f, g \rangle \triangleq \int_{\mathbb{R}^3} d^3\mathbf{r} \mathbf{f}(\mathbf{r}) \cdot \mathbf{g}^*(\mathbf{r}) \quad (\text{E.2})$$

(not to be confused with the time averaging in the formulation of the FDT), where for \mathcal{H}_V the integration domain can be limited to V . This allows one to define the (*Hermitian*) *adjoint* of operator \mathcal{A}^H :

$$\forall f, g: \langle f, \mathcal{A}^H g \rangle \triangleq \langle \mathcal{A} f, g \rangle, \quad (\text{E.3})$$

with the kernel $[\overline{\mathbf{A}}(\mathbf{r}', \mathbf{r})]^H$ (note the change of arguments). The *Hermitian (self-adjoint) operator* satisfies:

$$\mathcal{A}^H = \mathcal{A} \Rightarrow \forall f: \langle \mathcal{A} f, f \rangle \in \mathbb{R}. \quad (\text{E.4})$$

For instance, \mathcal{J}_V is Hermitian.

Next, recalling that Hermitian operators are analogous to real numbers, we define the *R*- and *I*-parts of an arbitrary operator:

$$\mathcal{A}^R \triangleq \frac{\mathcal{A} + \mathcal{A}^H}{2}, \quad \mathcal{A}^I \triangleq \frac{\mathcal{A} - \mathcal{A}^H}{2i} \Rightarrow \mathcal{A} = \mathcal{A}^R + i\mathcal{A}^I, \quad (\text{E.5})$$

which is a straightforward generalization of Toeplitz decomposition of a matrix (into Hermitian and skew-Hermitian parts). The kernels of these operators are given by Eq. (9). Note that both \mathcal{A}^R and \mathcal{A}^I are Hermitian by definition, which implies

$$\operatorname{Re} \langle \mathcal{A} f, f \rangle = \langle \mathcal{A}^R f, f \rangle, \quad \operatorname{Im} \langle \mathcal{A} f, f \rangle = \langle \mathcal{A}^I f, f \rangle. \quad (\text{E.6})$$

Naturally, this decomposition applies to dyadics as well, then Eq. (E.6) becomes:

$$\operatorname{Re} (\mathbf{b}^* \cdot \overline{\mathbf{A}} \cdot \mathbf{b}) = \mathbf{b}^* \cdot \overline{\mathbf{A}}^R \cdot \mathbf{b}, \quad \operatorname{Im} (\mathbf{b}^* \cdot \overline{\mathbf{A}} \cdot \mathbf{b}) = \mathbf{b}^* \cdot \overline{\mathbf{A}}^I \cdot \mathbf{b}. \quad (\text{E.7})$$

In contrast to the inner product of functions, the dot product of vectors is defined without conjugation, as is the standard way when using dyadics (then application of dyadics is analogous to that of an operator, i.e., the argument is not conjugated).

We have previously shown that the I -part of the Green's dyadic is very convenient for emission problems [20], and derived the above relations involving A^I or \bar{A}^I . The I -part of matrices has also been used in [11]. With respect to naming, A^R and iA^I can be called the Hermitian and skew-Hermitian parts of A , respectively, but we are not aware of any suitable term for A^I . While it can be called an imaginary part of A , in application to matrices it may be confused with taking the imaginary part of each element, i.e., $\text{Im}\bar{A}$ (although see Eqs. (E.8), (E.9) below). Therefore, we use the term I -part in this review. Moreover, by the Hermitian, skew-Hermitian, and I -parts of a dyadic function (an operator kernel) we denote kernels of the corresponding parts of the operator.

In a common case of a reciprocal environment, many electromagnetic operators are *pseudo-self-adjoint* [18,85], i.e., $A = A^T = A^{H*}$, where T denotes the pseudo-adjoint (a generalization of matrix transpose) and $*$ denotes the complex conjugation applied to the operator kernel. For such operators the kernels of the R - and I -parts are obtained by the standard complex analogues:

$$A^H = A^* \Leftrightarrow \bar{A}^R(\mathbf{r}, \mathbf{r}') = \text{Re}\bar{A}(\mathbf{r}, \mathbf{r}'), \quad \bar{A}^I(\mathbf{r}, \mathbf{r}') = \text{Im}\bar{A}(\mathbf{r}, \mathbf{r}'), \quad (\text{E.8})$$

Similarly, the polarizability in a reciprocal environment is complex-symmetric. For such dyadics

$$\bar{A}^T = \bar{A} \Leftrightarrow \bar{A}^H = \bar{A}^* \Leftrightarrow \bar{A}^R = \text{Re}\bar{A}, \quad \bar{A}^I = \text{Im}\bar{A}. \quad (\text{E.9})$$

However, we keep the general notation in this review to allow for non-reciprocal configurations.

Continuing the analogy between operators and complex numbers, one can show that

$$\forall A, B: (A^H)^R = A^R, \quad (A^H)^I = -A^I, \quad (A^H B A)^R = A^H B^R A, \quad (A^H B A)^I = A^H B^I A, \quad (\text{E.10})$$

$$\forall A: A = A^H \Rightarrow A^R = A, \quad A^I = 0, \quad (\text{E.11})$$

and for any invertible operator (or dyadic):

$$(A^{-1})^R = A^{-H} A^R A^{-1} = A^{-1} A^R A^{-H}, \quad (A^{-1})^I = -A^{-H} A^I A^{-1} = -A^{-1} A^I A^{-H}, \quad (\text{E.12})$$

where $A^{-H} \triangleq (A^{-1})^H = (A^H)^{-1}$. More advanced properties are derived in [Appendix F](#).

The definition (E.1) may easily incorporate the three-dimensional delta function $\delta(\mathbf{r} - \mathbf{r}')$, corresponding to a pointwise-multiplication operator. In particular, the potential operator \mathcal{U} with the kernel $\bar{\mathbf{U}}(\mathbf{r})\delta(\mathbf{r} - \mathbf{r}')$ acts as

$$\mathcal{g} = \mathcal{U}\mathcal{f} \Leftrightarrow \mathbf{g}(\mathbf{r}) = \bar{\mathbf{U}}(\mathbf{r}) \cdot \mathbf{f}(\mathbf{r}). \quad (\text{E.13})$$

Many of the dyadic Green's functions discussed in this review are strongly singular (with the notable exception of their I -parts). Therefore, we define the corresponding operators with the explicit treatment of the singularity [19]:

$$\mathcal{g} = \mathcal{G}\mathcal{f} \Leftrightarrow \mathbf{g}(\mathbf{r}) = \lim_{V_0 \rightarrow 0} \int_{\mathbb{R}^3 \setminus V_0} d^3\mathbf{r}' \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}') \cdot \mathbf{f}(\mathbf{r}') - \frac{1}{3k^2} \mathbf{f}(\mathbf{r}), \quad (\text{E.14})$$

where V_0 is the spherical exclusion volume centered at \mathbf{r} , and its shape is tightly connected with the specific coefficient in the last term [55]. The latter coefficient naturally depends on the singular part ($\sim R^{-3}$) of $\bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}')$, also known as the operator symbol [86]. Fortunately, this part is the same for $\bar{\mathbf{G}}$, $\bar{\mathbf{G}}_0$, and $\bar{\mathbf{G}}_{\text{st}}$, hence Eq. (E.14) applies to all these operators.

The outer product of two functions is

$$A = \mathcal{f} \otimes \mathcal{g} \Leftrightarrow \bar{A}(\mathbf{r}, \mathbf{r}') = \mathbf{f}(\mathbf{r}) \otimes \mathbf{g}(\mathbf{r}') \Leftrightarrow \forall \mathcal{h}: A\mathcal{h} = \mathcal{f} \langle \mathcal{g}, \mathcal{h}^* \rangle, \quad (\text{E.15})$$

which is a generalization of the dyadic product (and neither of them includes conjugation). It is easy to show that:

$$\forall \mathcal{f}, \mathcal{g}, A: \mathcal{g} \otimes \mathcal{f} = (\mathcal{f} \otimes \mathcal{g})^T, \quad (A\mathcal{f}) \otimes \mathcal{g} = A(\mathcal{f} \otimes \mathcal{g}), \quad \mathcal{f} \otimes (A\mathcal{g}) = (\mathcal{f} \otimes \mathcal{g})A^T. \quad (\text{E.16})$$

Similarly, we may construct outer products of vectors and functions resulting in linear maps (operating between \mathbb{C}^3 and the function space). In particular, let us define:

$$\mathcal{W} = \sum_{\mu} w_{\mu} \otimes \hat{\mathbf{e}}_{\mu}, \quad (\text{E.17})$$

where μ indexes the Cartesian coordinates, $\hat{\mathbf{e}}_{\mu}$ are the unit vectors in \mathbb{C}^3 , and w_{μ} are three basic functions (to be specified below). Then

$$\mathcal{W}\mathbf{a} = \sum_{\mu} a_{\mu} w_{\mu}, \quad \mathcal{W}^H \mathcal{f} = \sum_{\mu} \langle \mathcal{f}, w_{\mu} \rangle \hat{\mathbf{e}}_{\mu}, \quad (\text{E.18})$$

i.e., \mathcal{W} produces a linear combination of w_{μ} and \mathcal{W}^H projects any function on this (incomplete) basis. For operators such projection leads to a dyadic:

$$(\mathcal{W}^H A \mathcal{W})_{\mu, \nu} = \langle A w_{\nu}, w_{\mu} \rangle = \iint_{\mathbb{R}^3} d^3\mathbf{r} d^3\mathbf{r}' w_{\mu}^*(\mathbf{r}) \cdot \bar{A}(\mathbf{r}, \mathbf{r}') \cdot w_{\nu}(\mathbf{r}'), \quad (\text{E.19})$$

Moreover, we can combine $w_{\mu}(\mathbf{r})$ as columns of the dyadic function $\bar{\mathbf{W}}(\mathbf{r})$ [cf. Eq. (E.17)]:

$$\bar{\mathbf{W}}(\mathbf{r}) \triangleq \{w_1(\mathbf{r}), w_2(\mathbf{r}), w_3(\mathbf{r})\} = \sum_{\mu} w_{\mu}(\mathbf{r}) \otimes \hat{\mathbf{e}}_{\mu}, \quad (\text{E.20})$$

Then Eq. (E.19) becomes especially simple

$$\mathcal{W}^H \mathcal{A} \mathcal{W} = \iint_{\mathbb{R}^3} d^3 \mathbf{r} d^3 \mathbf{r}' \left[\overline{\mathbf{W}}(\mathbf{r}) \right]^H \cdot \overline{\mathbf{A}}(\mathbf{r}, \mathbf{r}') \cdot \overline{\mathbf{W}}(\mathbf{r}'). \quad (\text{E.21})$$

Replacing \mathcal{A} with the identity operator, we obtain that $\mathcal{W}^H \mathcal{W}$ is a dyadic with elements $\langle w_\mu, w_\nu \rangle$. This dyadic is diagonal if the basis is orthogonal. Importantly, Eq. (E.16) holds for linear maps as well, in particular:

$$\forall \mathbf{f}, \mathbf{g} : (\mathcal{W}^H \mathbf{f}) \otimes (\mathcal{W}^H \mathbf{g})^* = \mathcal{W}^H (\mathbf{f} \otimes \mathbf{g}^*) \mathcal{W}. \quad (\text{E.22})$$

Finally, we consider a specific case of \mathcal{W}_0 defined by the constant basis inside the scatterer volume:

$$\forall \mu : w_{0,\mu} \in \mathcal{H}_V, \quad \overline{\mathbf{W}}(\mathbf{r}) = \overline{\mathbf{I}}, \quad (\text{E.23})$$

which is a complete basis for constant vector functions. Then (E.18), (E.19) becomes:

$$\mathcal{W}_0^H \mathbf{f} = \int_V d^3 \mathbf{r} \mathbf{f}(\mathbf{r}), \quad (\text{E.24})$$

$$\mathcal{W}_0^H \mathcal{A} \mathcal{W}_0 = \iint_V d^3 \mathbf{r} d^3 \mathbf{r}' \overline{\mathbf{A}}(\mathbf{r}, \mathbf{r}'), \quad (\text{E.25})$$

i.e., \mathcal{W}_0 is the integration map, while $\mathcal{W}_0 \mathbf{a}$ is a constant function inside V . Another possible basis based on the VSWFs is discussed in Appendix D.

Appendix F. Relation between the static and dynamic transition operators

Starting from Eqs. (52), (54) we obtain:

$$(\mathcal{J} - \mathcal{U}) \mathcal{G}_{\text{st}} \mathcal{T}_{\text{st}} = \mathcal{T} \mathcal{G} \mathcal{U} \mathcal{G}_{\text{st}} \mathcal{T}_{\text{st}} = \mathcal{T} \mathcal{G} (\mathcal{T}_{\text{st}} - \mathcal{U}) \Rightarrow \mathcal{T} (\mathcal{G} - \mathcal{G}_{\text{st}}) \mathcal{T}_{\text{st}} = \mathcal{T} \mathcal{G} \mathcal{U} - \mathcal{U} \mathcal{G}_{\text{st}} \mathcal{T}_{\text{st}} = \mathcal{T} - \mathcal{T}_{\text{st}}, \quad (\text{F.1})$$

which stays valid if the static and dynamic quantities are interchanged. Thus,

$$\mathcal{T} - \mathcal{T}_{\text{st}} = \mathcal{T} (\mathcal{G} - \mathcal{G}_{\text{st}}) \mathcal{T}_{\text{st}} = \mathcal{T}_{\text{st}} (\mathcal{G} - \mathcal{G}_{\text{st}}) \mathcal{T}. \quad (\text{F.2})$$

Further we plan to express \mathcal{T}^I and analyze its order of magnitude. For that we prove the following general operator identity. Let \mathcal{A}, \mathcal{B} be arbitrary operators (the same holds for dyadics) and

$$\mathcal{C} = \mathcal{A} (\mathcal{J} - \mathcal{B} \mathcal{A})^{-1}, \quad (\text{F.3})$$

where the unique inverse of $(\mathcal{J} - \mathcal{B} \mathcal{A})$ is assumed to exist, and \mathcal{J} is the identity operator. We multiply the identity $\mathcal{J} - (\mathcal{J} - \mathcal{B} \mathcal{A}) = \mathcal{B} \mathcal{A}$ by \mathcal{C}^H and $(\mathcal{J} - \mathcal{B} \mathcal{A})^{-1}$ from the left and right, respectively, and obtain

$$\mathcal{C}^H (\mathcal{J} - \mathcal{B} \mathcal{A})^{-1} - \mathcal{C}^H = \mathcal{C}^H \mathcal{B} \mathcal{C}, \quad (\text{F.4})$$

which implies

$$\mathcal{C} = (\mathcal{J} - \mathcal{B} \mathcal{A})^{-H} \mathcal{A} (\mathcal{J} - \mathcal{B} \mathcal{A})^{-1} - \mathcal{C}^H \mathcal{B} \mathcal{C} = (\mathcal{J} - \mathcal{B} \mathcal{A})^{-H} (\mathcal{A} - \mathcal{A}^H \mathcal{B} \mathcal{A}) (\mathcal{J} - \mathcal{B} \mathcal{A})^{-1}. \quad (\text{F.5})$$

Taking the R - and I -parts of Eq. (F.5) results in

$$\mathcal{C}^R = (\mathcal{J} - \mathcal{B} \mathcal{A})^{-H} \mathcal{A}^R (\mathcal{J} - \mathcal{B} \mathcal{A})^{-1} - \mathcal{C}^H \mathcal{B}^R \mathcal{C} = (\mathcal{J} - \mathcal{B} \mathcal{A})^{-H} (\mathcal{A}^R - \mathcal{A}^H \mathcal{B}^R \mathcal{A}) (\mathcal{J} - \mathcal{B} \mathcal{A})^{-1}, \quad (\text{F.6})$$

$$\mathcal{C}^I = (\mathcal{J} - \mathcal{B} \mathcal{A})^{-H} \mathcal{A}^I (\mathcal{J} - \mathcal{B} \mathcal{A})^{-1} + \mathcal{C}^H \mathcal{B}^I \mathcal{C} = (\mathcal{J} - \mathcal{B} \mathcal{A})^{-H} (\mathcal{A}^I + \mathcal{A}^H \mathcal{B}^I \mathcal{A}) (\mathcal{J} - \mathcal{B} \mathcal{A})^{-1}, \quad (\text{F.7})$$

where we used Eq. (E.10). The above derivation is completely general, in particular, it does not require that \mathcal{A} and \mathcal{C} are invertible. However, if the latter holds, the same result can be obtained even simpler through [cf. Eq. (11)]:

$$\mathcal{C}^{-1} = \mathcal{A}^{-1} - \mathcal{B}, \quad (\text{F.8})$$

by directly taking its R - and I -parts and using Eq. (E.12).

Applying Eq. (F.7) to Eq. (F.2) (substituting \mathcal{A}, \mathcal{B} , and \mathcal{C} by $\mathcal{T}_{\text{st}}, \mathcal{G} - \mathcal{G}_{\text{st}}$, and \mathcal{T} , respectively) we obtain:

$$\begin{aligned} \mathcal{T}^I &= [\mathcal{J} - (\mathcal{G} - \mathcal{G}_{\text{st}}) \mathcal{T}_{\text{st}}]^{-H} \mathcal{T}_{\text{st}}^I [\mathcal{J} - (\mathcal{G} - \mathcal{G}_{\text{st}}) \mathcal{T}_{\text{st}}]^{-1} + \mathcal{T}^H \mathcal{G}^I \mathcal{T} \\ &= [\mathcal{J} - (\mathcal{G} - \mathcal{G}_{\text{st}}) \mathcal{T}_{\text{st}}]^{-H} (\mathcal{T}_{\text{st}}^I + \mathcal{T}_{\text{st}}^H \mathcal{G}^I \mathcal{T}_{\text{st}}) [\mathcal{J} - (\mathcal{G} - \mathcal{G}_{\text{st}}) \mathcal{T}_{\text{st}}]^{-1}, \end{aligned} \quad (\text{F.9})$$

i.e., \mathcal{T}^I is the sum of the term proportional to \mathcal{G}^I , which cannot be neglected for small absorption, and the term proportional (and asymptotically equivalent) to $\mathcal{T}_{\text{st}}^I$. Operator \mathcal{G}^R affects only the multiplicative factor for both these terms, more specifically, the small difference between this coefficient and \mathcal{J} .

Operators \mathcal{U}, \mathcal{T} , and \mathcal{T}_{st} are definitely not invertible when considered from \mathcal{H} to \mathcal{H} , since their kernels are non-zero only inside V ; however, in many cases they are invertible if considered from \mathcal{H}_V to \mathcal{H}_V . In particular, if $\overline{\mathbf{U}}(\mathbf{r})$ is non-singular for all $\mathbf{r} \in V$, let us define $\mathcal{U}^{-1} : \mathcal{H}_V \rightarrow \mathcal{H}_V$ with the kernel $\overline{\mathbf{U}}^{-1}(\mathbf{r}) \delta(\mathbf{r} - \mathbf{r}')$. It obviously satisfies $\mathcal{U}^{-1} \mathcal{U} = \mathcal{U} \mathcal{U}^{-1} = \mathcal{J}_V$. One may also call it pseudo-inverse,

but abusing the notation of superscript -1 should not cause any confusion. Then \mathcal{T}^{-1} and $\mathcal{T}_{\text{st}}^{-1}$ exist in the same sense, since Eqs. (52), (54) imply

$$\mathcal{T}^{-1} = \mathcal{U}^{-1} - \mathcal{J}_V \mathcal{G} \mathcal{J}_V, \quad \mathcal{T}_{\text{st}}^{-1} = \mathcal{U}_{\text{st}}^{-1} - \mathcal{J}_V \mathcal{G}_{\text{st}} \mathcal{J}_V, \quad (\text{F.10})$$

In other words, the operators \mathcal{T}^{-1} and $\mathcal{T}_{\text{st}}^{-1}$ are known analytically, and the expressions are especially simple for their I -parts. Moreover, Eq. (F.9) can be simplified to

$$\mathcal{T}^I - \mathcal{T}^H \mathcal{G}^I \mathcal{T} = -\mathcal{T}^H (\mathcal{T}_{\text{st}}^{-1})^I \mathcal{T}. \quad (\text{F.11})$$

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