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Energy balance in the coherent scattering of radiation by a system of many particles

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Using a unified description of the fields of incident and scattered waves, the energy balance conditions for time-averaged energy fluxes during the scattering of a monochromatic wave created by an arbitrary radiation source on a system of particles interacting through scattered fields are considered. A „duality lemma“ is obtained for local values of energy fluxes, similar to Lorentz lemma for fields from two sources and determining the redistribution of energy fluxes between scatterers and the source. The total energy flow is divided into „energy“ and „interference“ parts, each of which has its own source function localized on particles, and which are preserved during propagation in free space. The variants of the optical theorem corresponding to various subsystems (clusters) are described, as well as their relationship to the Purcell factor. The result is a detailed picture of energy exchange for arbitrarily chosen clusters of interacting particles.

Keywords: multiple scattering, coherent radiation source, energy conservation, optical theorem, Purcell effect, radiation losses.

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Introduction

Interaction of electromagnetic radiation with scattering and absorbing particles covers a large variety of practical applications that include both classic problems of optics, radiolocation, atmosphere physics, etc. and modern problems related to development of nanotechnologies. This sphere of the problems is described based on the Maxwell equations in a frequency area [1,2] or their ensuing system of the integral equation like the Lippmann-Schwinger quantum-mechanical equation [3]. In addition to scattering on single particles, these equations can also encompass various effects of multiple scattering, which correspond to interaction of particles through the scattered electromagnetic field, such as formation of photon band gaps in metamaterials, Anderson localization in systems of random scatters, effects of backscattering amplification (also known as weak localization), etc. [2].

Unlike a phenomenological theory of radiation transfer, which uses the transfer equation with an heuristic notion of an elementary scattering volume [4], the Maxwell equations provide rigorous description of specific models of the particles with an arbitrary form and an internal structure and make it possible to obtain statistics-wave justification of the transfer theory [5]. However, these rigorous models as a rule allow an accurate solution only in an extremely limited number of cases and usually require application of numerical calculations.

Relationships related to an energy balance are important for understanding general mechanisms of absorption and scattering of radiation. They particularly include a so-

called optical theorem and the Purcell factor related to variation of emitter power due to presence of a scatterer. These relationships make it possible to simplify finding of such important integral characteristics of the considered system as complete sections of extinction, scattering and absorption. They are used to check the quality of the used approximate solutions and to obtain a general physical picture in the scattering problems. In recent years, the energy relationships have been also used in extensive new applications related to obtaining fundamental boundaries of variation of the various physical parameters of the scattering system. Finding these boundaries makes it possible, in particular, to judge potential capabilities when creating new metamaterials and designing engineering devices in nanophotonics and plasmonics (see the studies [6–8] and many sources referred to therein). This approach is based on methods of convex optimization [9], which mean that extremums of desired physical magnitudes are found by using various integral limitations like the energy balance conditions. At the same time, it considers a wider class of functions than rigorous solutions of the wave equations, thereby making it possible to find the desired maximums without solving the exact wave equations. Extension of the class of the additional conditions results in tightening of these boundaries, which in certain cases turn out to be achievable, thereby acquiring a direct physical meaning.

Traditionally, the conditions of the energy balance are considered as applicable to a single scatterer irradiated by a flat wave, thereby resulting in a classic formulation of the optical theorem that relates extinction to an amplitude of scattering „forward“ [10,11]. The description of the energy

balance was extended in the studies [12–14] for the system that consists of both the scatterer and the emitter. Unlike a common approach of considering the balance of total flows outgoing through a surface of the selected volumes, the previous studies of the authors [15,16] have also used local conditions of the energy balance, which express divergence of the flows through the respective sources in each point of space. At the same time, it considered a model of the scalar wave equation and the single scatterer. The present study is aimed at extending the results [15,16] to the case of multiple scattering of electromagnetic radiation on the system of the absorbing scatterers. The obtained results are aimed at improving understanding of the mechanisms absorption and scattering of light in the systems of interacting particles, which are of primary importance in optics, photonics and plasmonics.

Below is described the case of coherent scattering by the system of fixed scattering particles, for which a phase of radiation does not vary in individual acts of scattering. Situations that are related to noncoherent scattering and require use of the statistical approach [5] are not considered below. At this, for the sake of simplicity, we limit ourselves to the case of one pre-defined (i.e. foreign) current $\mathbf{j}_0(\mathbf{r})$ localized outside the scatterer system, although the below-obtained results can be easily extended to the case of an assembly of radiating and absorbing particles as well by introducing a set of particles with external currents pre-defined thereon instead of one external current.

Section 1 considers a general formulation of the problem that results in a system of differential equations of multiple scattering of monochromatic radiation on the system of particles with pre-defined arbitrary distribution of permittivity inside each particle. Section 2 provides the main result of the present study, namely a „duality lemma“ (12), which expresses a local form of the law of conservation of averaged energy flows from two arbitrary sources that are pre-defined or induced. As known to the authors, this form of the law of conservation of energy was not previously written explicitly in literature on multiple scattering (some references to similar results, but non-identical to (12) are given in Conclusion, a simple derivation of this lemma is provided in Appendix). After that, the same Section describes local and integral forms of the laws of conservation of the time-averaged power flows, which are divided into energy flow and interference flows, which are relation to superposition of the scattered fields and the incident wave field. The next sections are used to illustrate applicabilities of the said lemma. Division of the powers and the flows into partial components is given in Section 3. Section 4 describes various variants for selecting auxiliary volumes, which allow considering, when based on (12), the energy balance for the arbitrary clusters inside the system of particles that interact through the scattered fields. A relationship of the partial powers to an operator of free propagation and field-creating currents is briefly described in Section 5. Conclusion formulates the main conclusions.

1. Problem formulation

Let us consider scattering of monochromatic radiation created by the pre-defined external current $\mathbf{j}_0(\mathbf{r})$ that is distribute inside the volume V_0 . Radiation is scattered by the system (cluster) of absorbing particles (Fig. 1).

The positions of the particles are described by some internal points of each of them $\mathbf{r}_i$, $i = 1, 2, \dots, N$, $\mathbf{r}_0$ is the internal point V_0 , wherein the volumes of the particles V_i and the sources V_0 are considered to be non-overlapping. The time dependence is proportional to a multiplier $\exp(-i\omega t)$ that is omitted below, so are the dependences on the angular frequency ω . Taking for sake of simplicity that medium permittivity is unity, we will assume that each of the particles is described by its own distribution of complex permittivity $\varepsilon_i(\mathbf{r})$ (as known, it also includes the case of nanoparticles [2]). Then, complete permittivity is written as

$$\begin{aligned}\varepsilon(\mathbf{r}) &= \sum_{i=1}^N \varepsilon_i(\mathbf{r})\theta_i(\mathbf{r}) + \left(1 - \sum_{i=1}^N \theta_i(\mathbf{r})\right) \\ &\equiv 1 + \sum_{i=1}^N (\varepsilon_i(\mathbf{r}) - 1)\theta_i(\mathbf{r}).\end{aligned}\quad (1)$$

Hereinafter, $\theta_i = \theta_i(\mathbf{r})$ is a characteristic function that is equal to unity inside the volume V_i and zero outside it, $i = 0, 1, 2, \dots, N$. At the same time, $\mathbf{j}_0(\mathbf{r}) = \theta_0(\mathbf{r})\mathbf{j}_0(\mathbf{r})$, since the pre-defined current $\mathbf{j}_0(\mathbf{r})$ is assumingly localized inside the volume V_0 .

The electric field strength $\mathbf{E}(\mathbf{r})$ is determined by the equation

$$[\nabla \times \nabla \times - k_0^2 \varepsilon(\mathbf{r})]\mathbf{E}(\mathbf{r}) = i\omega\mu_0\mathbf{j}_0(\mathbf{r}), \quad (2)$$

where $k_0 = \omega/c$ is a wave number, while μ_0 is magnetic permeability of vacuum. The equation (2) is supplemented by the Sommerfeld infinity radiation conditions, which can be fulfilled by introducing an infinitely small imaginary part to the wave number k_0 , which is omitted for sake

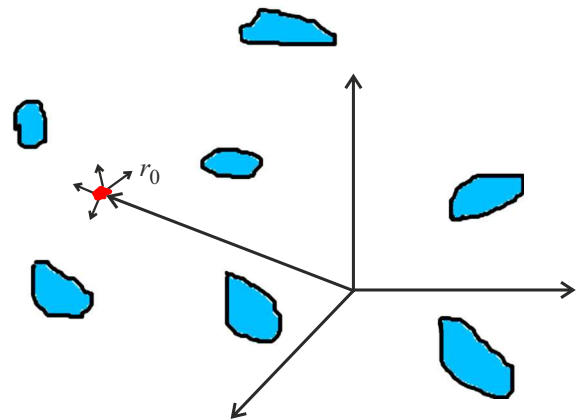


Figure 1. Radiation of the pre-defined external current outside the cluster of scattering and absorbing particles.

of simplicity hereinafter (see, for example, [1,2]). The solution (2) makes it possible to find the magnetic field strength $\mathbf{H}(\mathbf{r})$ as well, which is

$$\mathbf{H}(\mathbf{r}) = \nabla \times \mathbf{E}(\mathbf{r}) / (i\omega\mu_0). \quad (3)$$

By substituting (1) into (2) and omitting spatial arguments for sake of simplicity, we write (2) in an abbreviated form as

$$L_0 \mathbf{E}_t = \mathbf{q}_t. \quad (4)$$

Here, for uniformity, the index $\mathbf{E} \equiv \mathbf{E}_t$ is assigned to the full field $\mathbf{E}$ and the following designations are introduced

$$L_0 = \nabla \times \nabla \times -k_0^2, \quad (5)$$

$$\mathbf{q}_t = \mathbf{q}_0 + \mathbf{q}_s \equiv \sum_{i=0}^N \mathbf{q}_i, \quad (6)$$

$$\mathbf{q}_0 = i\omega\mu_0 \mathbf{j}_0(\mathbf{r}), \quad \mathbf{q}_s = k_0^2(\varepsilon(\mathbf{r}) - 1)\mathbf{E}_t = \sum_{i=1}^N U_i \mathbf{E}_t \equiv \sum_{i=1}^N \mathbf{q}_i, \quad (7)$$

where

$$\mathbf{q}_i = U_i \mathbf{E}_t, \quad U_i = k_0^2(\varepsilon_i - 1)\theta_i. \quad (8)$$

Unlike the external current $\mathbf{q}_0$, which is assumed to be pre-defined, the values of „scattering potentials“ U_i (8) describe the secondary sources $\mathbf{q}_i$, i.e. the currents related to polarization of the i -th scatterer. At the same time, the source $\mathbf{q}_s$ corresponds to the full scattered field or, in other words, to description of all the scattering particles as a single combined scatterer with permittivity $\varepsilon(\mathbf{r})$.

2. Local and integral laws of conservation

2.1. Differential equations for the partial fields

Let us write the full field $\mathbf{E}_t$ as a sum of the „partial fields“, namely, the field of the incident wave $\mathbf{E}_0$ and the waves scattered by each of the scatterers $\mathbf{E}_i$:

$$\mathbf{E}_t = \mathbf{E}_0 + \mathbf{E}_s \equiv \sum_{i=0}^N \mathbf{E}_i, \quad (9)$$

$$\mathbf{E}_s = \sum_{i=1}^N \mathbf{E}_i. \quad (10)$$

Here, $\mathbf{E}_0$ corresponds to the field created by the pre-defined (foreign) current $\mathbf{q}_0$ without the scatterers, while the full scattered field $\mathbf{E}_s$ is created by the source $\mathbf{q}_s$ and expressed as a sum of the fields $\mathbf{E}_i$ scattered by each of the scatterers with the sources $\mathbf{q}_i$, $i = 1, 2, \dots, N$. Finding an explicit form of these sources as well as the scattered fields requires the full solution of the scattering problem (Section 5.2).

Due to linearity of the problem, all the strengths $\mathbf{E}_i$, $i = t, s, 0, 1, 2, \dots, N$ are described by the equations that coincide with (4) in a form

$$L_0 \mathbf{E}_i = \mathbf{q}_i, \quad (11)$$

but with different functions of the sources (currents) $\mathbf{q}_i$, $i = t, s, 0, 1, 2, \dots, N$.

The equations (11) when $i = 0, 1, 2, \dots, N$ form a system that is equivalent to the equation (4) and describes the full field $\mathbf{E}_t$ (9). Summing both the parts of (11) over $i = 0, 1, 2, \dots, N$ transforms the system of equations (11) for the incident wave and the particle-scattered waves into an initial equation for the full field $\mathbf{E}_t$ (4). At the same time, each of the fields $\mathbf{E}_i$ is matched with a corresponding magnetic field $\mathbf{H}_i$ of the form (3).

The sources of the incident wave $\mathbf{E}_0$ and the scattered waves $\mathbf{E}_i$, $i = 1, 2, \dots, N$ are „local“ in the sense that they are spatially localized in an area of the pre-defined currents $\mathbf{q}_0$ and each of the scatterers $\mathbf{q}_i$, respectively. We will call these waves the „the partial components of the full field $\mathbf{E}_t$ “. Unlike it, the sources $\mathbf{q}_t$ (6) and $\mathbf{q}_s$ (7) that correspond to the full field $\mathbf{E}_t$ and the scattered fields $\mathbf{E}_s$ are „non-local“, i.e. distributed between the external current $\mathbf{q}_0$ and all the scatterers. Assumingly, all the local sources are not overlapped.

The only source of energy in the considered problem is the pre-defined external current $\mathbf{q}_0$, while the secondary induced currents $\mathbf{q}_i$, $i = 1, 2, \dots, N$ correspond to the scatterers that not only scatter the field energy, but absorb it as well, i.e. function as drains. It will be reflected in selecting signs related to currents of powers and their respective flows (see below). Despite these differences, the general properties of the local currents $\mathbf{q}_0$ and $\mathbf{q}_i$ and the energy relationships related thereto can be considered uniformly.

2.2. Local and integral laws of conservation of the partial flows

It is easy to obtain the following relationship from the Maxwell equations (11) and (3) (see Appendix)

$$\nabla(\mathbf{s}_{ij} + \mathbf{s}_{ji}) = (w_{ij} + w_{ji}), \quad (12)$$

which relate cross flows

$$\mathbf{s}_{ij} = (1/2)\text{Re}(\mathbf{E}_i \times \mathbf{H}_j^*) \quad (13)$$

with the respective powers

$$w_{ij} = (1/2)\text{Re}[\mathbf{E}_i \mathbf{q}_j^* / (i\omega\mu_0)], \quad (14)$$

where $i, j = t, s, 0, 1, 2, \dots, N$. Similar to the Lorentz lemma, we will call the relationship (12) a „duality lemma“ for the average values of the local energy flows. Let us describe simple consequences of the relationships (12)–(14).

We will call the vectors $\mathbf{s}_{ij} + \mathbf{s}_{ji}$, when $i \neq j$, „interference flows“, since they are related to interference of the

fields created by the two different sources $\mathbf{q}_i$ and $\mathbf{q}_j$. In accordance with (12), each of the sources of the considered pair gives its own contribution (w_{ji} and w_{ij}) to divergence of the interference flow $\mathbf{s}_{ij} + \mathbf{s}_{ji}$. When $i = j$, i.e. for the field $\mathbf{E}_i$ from one source $\mathbf{q}_i$, the vectors $\mathbf{s}_{ij} + \mathbf{s}_{ji}$ transit to double common Poynting vectors $2\mathbf{s}_{ii}$, while (12) gives the law of conservation of energy

$$\nabla \mathbf{s}_{ii} = w_{ii} \quad (15)$$

for the time-average values of the flow $\mathbf{s}_{ii}$ and the power w_{ii} . The traditional derivation of the law of conservation (15) for the energy flow $\mathbf{s}_{tt}$ of the full field $\mathbf{E}_t$ from the Maxwell equations is provided in many monographs (see, for example, [1,2]).

Let us note that the really observed flows of the energy and the power also have, in addition to the average values, fluctuating parts that oscillate with a double frequency and omitted below (the condition of conservation of the fluctuating power is discussed in detail in [17]). We also note that the physical meaning of the continuity condition (15) is clearer when using a definition of divergence as a local measure of productivity of the sources in the considered point of space, i.e. a surface integral of the energy flow $\mathbf{s}_{ii}$ for the infinitely small volume dv , which is normalized to the value of this volume: the positive value of w_{ii} corresponds to the field energy sources (i.e. a flow outgoing from dv outwardly), while the negative value corresponds to drains (the dv -absorbed flow, see, for example, [18]).

Since the scattered fields are assumed to be non-zero, generally speaking, in the whole space spatial localization of the powers $w_{ij} \equiv w_{ij}(\mathbf{r})$ (14) coincides with localization of the sources $\mathbf{q}_j \equiv \mathbf{q}_j(\mathbf{r})$. In other words, each of the powers w_{ij} determined according to (14) can be non-zero only in the area of the respective sources $\mathbf{q}_j$ and zero outside this area. Consequently, the powers $w_{ij}(\mathbf{r})$ are „local“ for the incident wave and the partial scattered waves (i.e. when $j = 0, 1, 2, \dots, N$) and „non-local“ for the full field and the scattered fields (i.e. when $j = t, s$; here, „locality“ is understood in the same sense, in which „locality“ of the sources $\mathbf{q}_j$ was spoken of above).

Let us consider some simple consequences of the conditions (12) for the local sources $\mathbf{q}_j$, to which the indices $i, j = 0, 1, 2, \dots, N$ correspond. According to (12), for each selection of the pair (i, j) a source of divergence of the flow $\mathbf{s}_{ij} + \mathbf{s}_{ji}$ is the power $w_{ij} + w_{ji}$. At the same time, each power w_{ij} is localized in the area of localization of the respective field source $\mathbf{q}_j$. Consequently, the full flows that correspond to $\mathbf{s}_{ij} + \mathbf{s}_{ji}$ are preserved with free propagation of radiation in the area between the sources where $w_{ij} + w_{ji} = 0$.

Integration of the both parts of (12) by the arbitrary volume v using the Gauss-Ostrogradsky theorem makes it possible to transit from the local flows $\mathbf{s}_{ij}$ and the powers w_{ij} to integral characteristics that are related to selecting v , which we designate with capital letters S_{ij} and W_{ij} with

the same indices:

$$\begin{aligned} \Sigma_{ij} &\equiv (S_{ij} + S_{ji}) \equiv \oint_{Dv} (\mathbf{s}_{ij} + \mathbf{s}_{ji}) d\Sigma \\ &= \int_v (w_{ij} + w_{ji}) d\mathbf{r} \equiv (W_{ij} + W_{ji}). \end{aligned} \quad (16)$$

If the considered volume v has not at least one of the sources $\mathbf{q}_i$ or $\mathbf{q}_j$, then $w_{ij} = w_{ji} = 0$ and the full flow Σ_{ij} vanishes, i.e. a part of this flow, which enters v , is equal to an exiting one. Thus, for each pair of the local sources $\mathbf{q}_i$ and $\mathbf{q}_j$, the flow Σ_{ij} that corresponds to them does not depend directly on availability of other local sources in the considered volume (it certainly does not mean that the scattered field $\mathbf{E}_i$ does not depend on other scatterers: here we are talking about a balance of the time-average powers).

When $i = j$, after being divided by 2, the flow (16) transits to the condition of energy conservation:

$$S_{ii} \equiv \oint_{Dv} \mathbf{s}_{ii} d\Sigma = \int_v \mathbf{w}_{ii} d\mathbf{r} \equiv W_{ii}, \quad (17)$$

where S_{ii} is the full energy flow that outgoes outwardly through a surface of the considered volume, while W_{ii} is the full power of the sources included in it. In accordance with (17), the flow S_{ii} vanishes when there is no respective source $\mathbf{q}_i$ inside the volume, which corresponds to conservation of the magnitude S_{ii} in a process of propagation of the waves outside this source.

If the considered volume v has only one of the currents, let us say, $\mathbf{q}_i$, then $W_{ij} = 0$ and according to (16), $\Sigma_{ij} = W_{ji}$. With increase of the volume v , the interference flow Σ_{ij} does not vary until this volume touches the second source of the considered pair $\mathbf{q}_j$ (Fig. 1). After this, with increase of v , as this volume absorbs the area of the source $\mathbf{q}_j$, the flow Σ_{ij} starts changing and goes to a new constant level after $\mathbf{q}_j$ is completely enclosed by the considered volume. At the same time, other sources do not directly affect the value of the interference flow that characterizes this pair $\mathbf{q}_i$ and $\mathbf{q}_j$. Outside the volume that includes both the sources, the flow Σ_{ij} propagates unchanged. This pattern is preserved for the case of a single source as well, i.e. at $i = j$, when the interference flow Σ_{ij} transits to the double energy flow.

3. Partial components of the flows and their sources

The local relationships (12) associate the values relating to each point of space of the flows $\mathbf{s}_{ij}$ with the respective powers w_{ij} . The conditions of the energy balance are given by relationships between various full powers W_{ij} (or relationships between the full flows S_{ij} , which are equivalent to them) at special selections of the volumes, to which these integral magnitudes belong. For the emitter and the single

scatterer, in addition to the law of conservation of energy, these integral characteristics are directly related to the optical theorem (scattering „forward“, the energy balance between the scattered field and the incident wave) as well as description of the Purcell factor (scattering „backwards“, the effect of the scattered wave on the emitter power) [16]. Let us consider the specific features of these effects for our system of the scatterers.

Let us write the expressions (13) and (14) for the flow s_{tt} and the power w_{tt} of the full field $\mathbf{E}_t$ by substituting $\mathbf{q}_t$ (6) and $\mathbf{E}_t$ (9) into them:

$$w_{tt} = \Sigma w_{ij} \equiv \Sigma w_{ii} + \Sigma' w_{ij}, \quad (18)$$

$$s_{tt} = \Sigma s_{ij} \equiv \Sigma s_{ii} + \Sigma' s_{ij}. \quad (19)$$

Here, the sums are taken over all the values of the indices $i, j = 0, 1, 2, \dots, N$, which correspond to the local sources, while the sum with a prime corresponds to summation over $i \neq j$.

By integrating both the parts of (18) and (19) by the arbitrary volume v in accordance with (16), we obtain similar relationships for the integral magnitudes:

$$W_{tt} = \Sigma W_{ij} \equiv \Sigma W_{ii} + \Sigma' W_{ij}, \quad (20)$$

$$S_{tt} = \Sigma S_{ij} \equiv \Sigma S_{ii} + \Sigma' S_{ij}. \quad (21)$$

Unlike the differential relationships (18) and (19), here, now all the integral magnitudes S_{ij} and W_{ij} depend on selection of the volume v .

The relationships (20) and (21) make it possible to describe all the integral characteristics of the field, which are related to the energy balance. In order to obtain these characteristics, it is sufficient to select the volume of interest to us and take into account a various energy character of the foreign source $\mathbf{q}_0$ that transfers energy to the electromagnetic field, and the induced currents $\mathbf{q}_i$, $i = 1, 2, \dots, N$ that function as drains.

After this, we will consider only the volumes that fully enclose the respective local sources $\mathbf{q}_j$, so that the following magnitudes used below when $i, j = 0, 1, \dots, N$

$$W_{ij} = \int w_{ij} d\mathbf{r} \quad (22)$$

are the full powers that correspond to w_{ij} and are related to full absorption or radiation of the source $\mathbf{q}_j$.

4. Energy balance and the optical theorem for the system of the scatterers

4.1. Foreign current and the Purcell factor

First we consider the arbitrary volume V_0^* that fully encloses V_0 (i.e. the external current $\mathbf{q}_0$), but does not affect the volumes of the scattering particles (the induced currents $\mathbf{q}_i$, $i = 1, 2, \dots, N$ (Fig. 2)). According to (14), for this

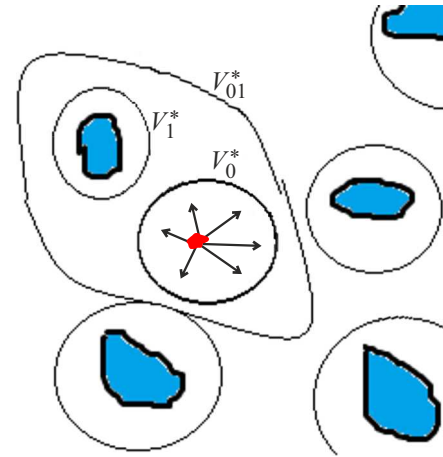


Figure 2. Selection of the volumes that enclose the scatterer q_1 and the source q_0 .

volume, all the values of the local w_{ij} and, consequently, full W_{ij} powers vanish when $j \neq 0$. Taking it into account, the expression for the full power (20) transits into

$$W_{em,0} \equiv W_{tt} = W_{00} + W_{int,0}, \quad (23)$$

where the sum of the interference summands is

$$W_{int,0} = \sum_{i=1}^N W_{i0}. \quad (24)$$

With this selection of the volume, the magnitude $W_{em,0}$ has a meaning of the full source radiation power that is a sum of a power of radiation in the free volume $W_{00} \geq 0$ and (generally, sign-variable) a power $W_{int,0}$ that describes the effect of all the scatterers on the power of radiation of the pre-defined current $\mathbf{q}_0$, i.e. the Purcell effect. For the single scatterer (i.e. when $N = 1$), this relationship was considered in detail [13]. The magnitude $\gamma = W_{int,0}/W_{00}$ is called a factor of amplification. In accordance with (24), each of the scatterers makes its own contribution to this factor (naturally, these factors are not independent due to mutual influence of the fields scattered by the particles).

In accordance with (12), each summand in (24) creates the flow $\Sigma_{i0} = S_{i0} + S_{0i}$, so that the emission flow (21) takes the following form

$$S_{em,0} \equiv S_{tt} = S_{00} + \sum_{i=1}^N (S_{i0} + S_{0i}). \quad (25)$$

This expression determines the energy flow that outgoes from the source $\mathbf{q}_0$ outwardly through the surface of the considered volume and is equal to the emission power (23).

We note that in our formulation we consider the external current that does not depend on the scatterers, which is a standard technique in classic electrodynamics [17]. More complex processes related to a quantum nature of the emitter can be described in a quasi-classical approximation

when taking into account a possible effect of the scattered fields on the foreign source q_0 (see, for example, the study [19], which considers a simple model of classic description of the dependence of fluorescence of an excited molecule on plasmon resonance of a metal nanoparticle nearby). This problem formulation can be easily included into the considered diagram, but it is outside the scope of the present study.

4.2. Single scatterer and the optical theorem

The similar relationships are obtained when considering any of the scatterers $\mathbf{q}_i$ instead of $\mathbf{q}_0$ and taking into account their absorbing nature. Selection of the volume V_1^* that has only one of the scatterer $\mathbf{q}_1$ (Fig. 3, *a*) corresponds to vanishing of all the powers W_{ij} with $j \neq 1$, so that instead of (24) we obtain the relationship

$$W_{tt} = W_{11} + \Sigma' W_{i1}, \quad (26)$$

where the sum with the prime is taken when $i \neq 1$, i.e. when $i = 0, 2, 3, \dots, N$.

Since the positive value of the power W_{tt} by definition corresponds to predominance of the flow that outgoes outwardly from the considered volume, here W_{tt} provides the full power W_{abs1} taken with a minus sign and absorbed by the scatterer $W_{tt} = W_{t1} = -W_{abs1}$. Similarly, the sum of (26) provides the extinction power $W_{ext,1}$ that is taken with the minus sign and corresponds to the interference flow directed inward the considered flow:

$$W_{ext,1} \equiv -\sum_{i \neq 1} W_{i1} \equiv -W_{01} - \sum_{i=2}^N W_{i1}. \quad (27)$$

In (26), the magnitude $W_{11} \geq 0$ gives the full power of scattering, i.e. the full flow outgoing through the surface of the volume V_1^* , which encloses the first scatterer, but does not affect other scatterers and the source q_0 . The part of this power is spent for excitation of currents induced in adjacent scatterers and contributes to variation of the emitter power

q_0 , i.e. the Purcell effect. The rest part corresponds to radiation losses.

As follows from determination of the powers w_{ij} (14), the right-hand part of (27) is matched with the local power

$$w_{ext,1} \equiv -\left(w_{01} + \sum_{i=2}^N w_{i1}\right) = -\left(1/2\right)\text{Re}\left[\left(\mathbf{E}_0 + \sum_{i=2}^N \mathbf{E}_i\right)\mathbf{q}_1^*/(i\omega\mu_0)\right]. \quad (28)$$

Whence, it is clear that the extinction power $W_{ext,1}$ describes a full work of the induced-in-particle current q_1 , which is done to the field that is external to the particle, i.e. the field of the incident wave and the waves scattered by all other particles (Fig. 3, *a*):

$$\mathbf{E}_{out1} \equiv \sum_{i \neq 1}^N \mathbf{E}_i = \mathbf{E}_0 + \sum_{i=2}^N \mathbf{E}_i. \quad (29)$$

As a result, (26) takes the form of the common optical theorem for the single scatterer, which says that the extinction power is a sum of the power of scattering $W_{sc,1} \equiv W_{11}$ and absorption:

$$W_{ext,1} = W_{sc,1} + W_{abs1}. \quad (30)$$

The powers in the right-hand part here are nonnegative, so that the following inequalities are fulfilled

$$W_{sc,1} \geq 0, \quad W_{abs,1} \geq 0, \quad W_{ext,1} \geq 0, \quad (31)$$

while a part of them is used when obtaining the fundamental boundaries [6–8], which were discussed above in Introduction. The optical system (30) is matched with the condition of the flow balance

$$S_{ext,1} \equiv -\sum_{i \neq 1} (S_{i1} + S_{1i}) = S_{11} + S_{abs1}, \quad (32)$$

where the particle-absorbed flow

$$S_{abs1} = -(S_{t1} + S_{1t}). \quad (33)$$

4.3. Optical theorem for the group of the scatterers

Obtaining the optical theorem for the arbitrary volume V_{12}^* that encloses only the two scattering particles q_1 and q_2 (Fig. 3, *b*) is formally equivalent to termwise summation of the optical theorems (30) written for each of them:

$$\begin{aligned} W_{ext,1} + W_{ext,2} &\equiv -W_{01} - W_{02} - \sum_{i=2}^N W_{i1} - \sum_{\substack{i=1 \\ i \neq 2}}^N W_{i2} \\ &= W_{11} + W_{22} + W_{abs2} + W_{abs1}. \end{aligned} \quad (34)$$

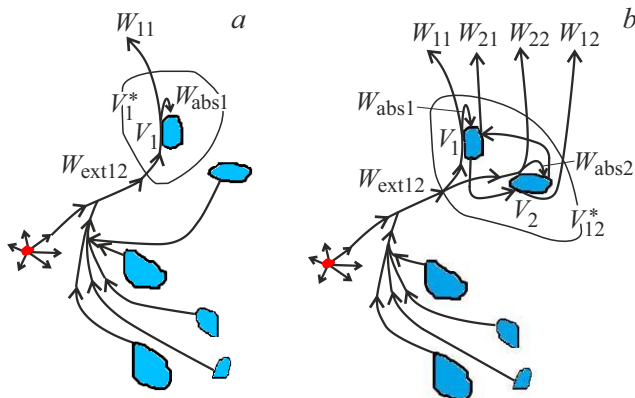


Figure 3. Diagrams of distribution of radiation from the average powers for the volumes V_1^* and V_{12}^* that enclose one scatterer (*a*) and two scatterers (*b*).

However, in order to transit to the common form of the optical theorem, which interprets a content of the considered volume V_{12}^* as one combined scatterer, it is necessary to regroup the summands included herein, by distinguishing an action of the scatterers that are external in relation to V_{12}^* . As a result, (34) will be written as

$$W_{\text{ext},1,2} = W_{\text{sc},1,2} + W_{\text{abs},1,2}, \quad (35)$$

where

$$W_{\text{ext},1,2} = - \sum_{\substack{i=0 \\ i \neq 1,2}}^N W_{i1} - \sum_{\substack{i=0 \\ i \neq 1,2}}^N W_{i2} \quad (36)$$

— the extinction power related to the action of all the sources that are external in relation to q_1 and q_2 (including q_0),

$$W_{\text{sc},1,2} = \sum_{i,j=1,2} W_{ij} \quad (37)$$

— the scattering power on the two scatterers that interact through the scattered fields, while

$$W_{\text{abs},1,2} = W_{\text{abs}2} + W_{\text{abs}1} \quad (38)$$

— the power of absorption by the two particles.

At the same time, the extinction power (36) is not reduced to the sum of the similar magnitudes (27) for each of the scatterers separately $W_{\text{ext},1,2} \neq W_{\text{ext},1} + W_{\text{ext},2}$, since a part of this sum now functions as the interference part of scattering on the two particles. All the powers included in the optical theorem (35) are nonnegative:

$$W_{\text{sc},1,2} \geq 0, \quad W_{\text{abs},1,2} \geq 0, \quad W_{\text{ext},1,2} \geq 0. \quad (39)$$

A pattern of conversion of the source energy for the two selected scatterers is illustrated in Fig. 3, *b*.

For the full cluster that encloses all the particles, only the field of the source u_0 is external in relation to it. Here, now the extinction power transits into a simple sum of the extinction powers for separate particles:

$$W_{\text{ext}1,2,\dots,N} \equiv - \sum_{i=1}^N W_{0i}, \quad (40)$$

while the optical theorem takes a conventional form

$$W_{\text{ext}1,2,\dots,N} = W_{\text{sc}1,2,\dots,N} + W_{\text{abs}1,2,\dots,N}, \quad (41)$$

where

$$W_{\text{sc}1,2,\dots,N} = \sum_{i,j=1}^N W_{ij}, \quad W_{\text{abs}1,2,\dots,N} = - \sum_{i=1}^N W_{ii}. \quad (42)$$

4.4. Scatterer and the source

The obtained relationships can be easily extended to the volume that encloses an arbitrary group of the scatterers by including the foreign source q_0 into it as well. Thus, for

the volume V_{01}^* that encloses only the source q_0 and the scatterer q_1 (Fig. 2), in (20) nonzero are the powers W_{i0} and W_{i1} that correspond to these sources. As a result, (20) takes the form of the expression for the emission power that outgoes from the considered pair

$$W_{em0,1} \equiv W_{tt} = W_{00} + \sum_{i \neq 0}^N W_{i0} + W_{11} + \sum_{i \neq 1}^N W_{j1}. \quad (43)$$

Taking into account (23) and the optical theorem (30), this relationship can be also written as

$$W_{em0,1} = W_{em0} - W_{abs1}. \quad (44)$$

This relationship expresses the law of conservation of energy for the pair q_0 and q_1 : the power that creates the emission flow from the considered volume is a difference of the power W_{em0} of the emitter q_0 (expressed with taking into account the Purcell effect) and the power W_{abs1} absorbed by the considered scatterer q_1 . Since the absorbed power can not exceed the emitted one, the inequality follows from (44)

$$W_{em0,1} \geq 0. \quad (45)$$

Similarly, it is possible to consider the flows that correspond to the arbitrary groups of the scatterers that interact through the scattered fields. At the same time, inclusion of the new scattering particles into the considered volume results in supplementing the right-hand side of (44) with the powers W_{absi} that describe absorption in each of them. In particular, for the volume that encloses the entire considered system, the full emission power is

$$W_{em0,1,2,\dots,N} = W_{em0} - \sum_{i=1}^N W_{absi}. \quad (46)$$

This power describes radiation into external space, i.e. radiation losses of the entire system as a whole. By taking into account the above-given relationships, it is easy to write also an explicit form of the power flow that corresponds to (44).

5. Partial powers and the operator of free propagation

Let us consider a relation of the above-obtained functions of the sources (powers) W_{ij} with the currents q_i that create the incident fields and the scattered fields. In accordance with (5), each of the scattered fields E_i , $i = 1, 2, \dots, N$ will be written as follows

$$E_i = G_0 q_i, \quad (47)$$

where

$$\begin{aligned} G_0 &= (L_0)^{-1} \equiv \nabla \times \nabla \times (-k^2 2_0)^{-1} \\ &\equiv -(1 + \nabla \nabla / k_0^2)(\Delta + k_0^2)^{-1} \end{aligned} \quad (48)$$

— the operator of free propagation, whose explicit form is known [1,2], is not written out here, too. Let us determine the scalar product for vectors of field strengths $\mathbf{E}_j$ a

$$\mathbf{E}_j^\dagger \mathbf{E}_i \equiv \int \mathbf{E}_j^*(\mathbf{r}) \mathbf{E}_i(\mathbf{r}) d\mathbf{r}, \quad (49)$$

where the integral is taken across the entire space. This unitary scalar product specifies a metric in the space of the three-component vectors $\mathbf{E}(\mathbf{r})$, which can be considered as the field $u = u(\alpha, \mathbf{r})$ that depends on the complex argument $(\alpha, \mathbf{r})$. Then, the operator G_0 is matched with the kernel $G_0(\alpha, \mathbf{r}; \alpha_0, \mathbf{r}_0)$ that includes tensor arguments (α, α_0) , wherein the action G_0 includes integration by $\mathbf{r}_0$ and summation over the discrete argument α_0 .

As it is a linear operator in the unitary space, G_0 is expressed as a sum the Hermitian G_0^h and the anti-Hermitian $i G_0^a$ parts¹, $G_0 \equiv G_0^h + i G_0^a$, where $G_0^h = (G_0 + G_0^\dagger)/2$, while $G_0^a = (G_0 - G_0^\dagger)/2i$, wherein the symbol „†“ designates the Hermitian conjugation (see, for example, [20]). In these notation, taking into account (14), the expression for the energy power W_{ii} (17) provides

$$\begin{aligned} W_{ii} &= \int_v w_{ii} d\mathbf{r} = (1/2\omega\mu_0) \text{Im} \int_v \mathbf{q}_i^* G_0 \mathbf{q}_i d\mathbf{r} \\ &= (1/2\omega\mu_0) \mathbf{q}_i^\dagger G_0^a \mathbf{q}_i, \end{aligned} \quad (50)$$

which is, in accuracy of an multiplier, a diagonal matrix element of the anti-Hermitian part of the operator of free propagation G_0^a , which corresponds to the source $\mathbf{q}_i$. Thus, the energy powers W_{ii} depend only on the anti-Hermitian part G_0^a of the operator of free propagation G_0 and do not depend on its Hermitian part. This property is retained for a symmetrized sum $W_{ij} + W_{ji}$ with $i \neq j$, which occurs when considering interaction of the pair of the sources $\mathbf{q}_i$ and $\mathbf{q}_j$ using the basic relationship (12). Generally, when using the off-diagonal magnitudes W_{ij} with $i \neq j$, the extinction sources in them are presented by the full Green's operator G_0 , rather than its anti-Hermitian part, so that

$$W_{ij} = (1/2\omega\mu_0) \text{Im} \mathbf{q}_j^\dagger G_0 \mathbf{q}_i. \quad (51)$$

Unlike the kernel G_0 , the matrix kernel $G_0^a(\mathbf{r}, \mathbf{r}_0)$ of the operator G_0^a has not specific features in zero, i.e. when $\mathbf{r} = \mathbf{r}_0$, wherein $G_0^a(\mathbf{r}_0, \mathbf{r}_0) = 1k_0/(6\pi)$, where 1 is a unity matrix [21]. Thus, it is possible to avoid regularization for calculating the powers in case of point scatterers [21] in contrast to describing the field that uses the full wave operator G_0 .

The operator G_0^a is nonnegative, i.e. all its diagonal matrix elements are nonnegative [20], $\mathbf{q}^\dagger G_0^a \mathbf{q} \geq 0$. This condition follows from physical considerations, reflects (nonnegative)

¹ The literature has not commonly-accepted designations for selecting the Hermitian and the anti-Hermitian components of the operator A (see discussion of the terminology in Appendix F in the paper [23]). In our notation, the anti-Hermitian part A^a is an Hermitian operator (like the imaginary part $\text{Im } z$ of the complex number z is a real number).

radiation losses in the system and provides non-negativity of all the energy powers W_{ii} (unlike the sign-variable interference powers W_{ij} , $i \neq j$). Thus, if considering a single dipole with the dipole moment $\mathbf{p}$ as a pre-defined source, by assuming that $\mathbf{q}_0 = \omega^2 \mu_0 \mathbf{p} \delta(\mathbf{r})$, then the power (50) takes a conventional form of the radiation power of the point dipole with the pre-defined dipole moment $\mathbf{p}$, $W_{00} = (\omega^4 \mu_0 |\mathbf{p}|^2 / 12\pi c)$ (this expression is usually derived by integration of the power flow over the infinite-radius sphere that encloses the scatterer).

Conclusion

The study has considered local and integral conditions of the energy balance for the arbitrary system of the scatterers that interact with each other through the scattered fields. Description of the partial components of the energy flows that are preserved in the process of propagation between the scatterers essentially uses the „duality lemma“ (12) which is obtained in the study to determine a relation of the time-average flows with the sources with taking into account interference of the scattered fields. It included consideration of the forms of the optical theorem for the clusters included in the considered system of the scatterers. It is shown, in particular, that the flow of extinction and scattering for the cluster depends on selection of the particles included in it and unlike the absorbed flow, it is not reduced to a simple sum of the similar extinction flows for its components.

The obtained clear picture of the energy balance is meant to improve understanding of the energy exchange processes during coherent scattering on the system of scattering particles, thereby making it possible to consider the energy exchange between the system clusters in contrast to the traditional approach that describes the energy balance for the system as a whole. The approach that uses the local conditions (12) can be also extended to other problems of the scattering theory, which are related to the more complex geometries of a problem. It is also applicable for the widespread model of the point scatterers and sources [21,23], when instead of continuous distributions of permittivity and the currents, the respective delta functions are used.

We note that the problem formulation that describes one monochromatic component of the field corresponds to a steady-state and does not use division of the currents into a „cause“ (the field sources $\mathbf{q}_0$) and „consequences“ (the induced sources $\mathbf{q}_j$), so that the above-described diagram features the currents $\mathbf{q}_0$ and $\mathbf{q}_j$ quite symmetrically (if only neglecting signs of the full flows of the energies outgoing from them). We also note that many studies in the literature have considered the similar relationships, but non-identical to (12) (thus, for example, when considering the case of the single scatterer, a similar conclusion was given in the study [13] as applicable to a specific choice of the fields of the incident and scattered waves as $\mathbf{E}_1$ and $\mathbf{E}_2$ and in the article [24] when constructing the optical theorem in the

time domain). However, the authors believe that it is use of the simple relationship (12) that makes it possible to obtain the most concise description of the considered problem.

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Conflict of interest

The authors declare that they have no conflict of interest.

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Appendix

„Duality lemma“ for the cross flows

When discussing the reciprocity conditions for the Maxwell equations in electrodynamic as well as in various applications, widely used is a classic Lorentz lemma that relates the values of the fields that are created in a fixed medium by two different external currents $\mathbf{j}_1$ and $\mathbf{j}_2$ [17]:

$$\nabla(\mathbf{E}_1 \times \mathbf{H}_2 - \mathbf{E}_2 \times \mathbf{H}_1) = (-\mathbf{E}_1 \mathbf{j}_2 + \mathbf{E}_2 \mathbf{j}_1)/(i\omega\mu_0). \quad (\text{A1})$$

When there is anisotropy, this relationship is fulfilled only for reciprocal media, for which the tensors of permittivity and permeability satisfy certain symmetry conditions, and it is violated, for example, in case of gyrotropy (nonreciprocal media are described in detail in the recent review [22]). Let us consider a derivation of the similar condition that is associated with the time-average values of the energy flows.

As in derivation of the Lorentz lemma, we will consider the Maxwell equations (2) and (3) for the two fields that are created by the two different pre-defined external currents $\mathbf{j}_1$ and $\mathbf{j}_2$. We write these equations as follows

$$\nabla \times \mathbf{E}_\alpha = (i\omega\mu_0)\mathbf{H}_\alpha, \quad (\text{A2})$$

$$\nabla \times \mathbf{H}_\alpha = (q_\alpha + k_0^2\mathbf{E}_\alpha)/(i\omega\mu_0), \quad (\text{A3})$$

where $\alpha = 1, 2$, while the sources $\mathbf{q}_1$ and $\mathbf{q}_2$ will be regarded by us as arbitrary ones (for the full $\mathbf{E}_t$ field $\mathbf{q}_\alpha = i\omega\mu_0\mathbf{j}_0(\mathbf{r}) + k_0^2(\varepsilon(\mathbf{r}) - 1)\mathbf{E}_\alpha$, the incident wave $\mathbf{E}_0$ corresponds to the case when $\varepsilon(\mathbf{r}) = 1$, while the partial scattered waves correspond to the case when $q_\alpha = (\varepsilon_i(\mathbf{r}) - 1)\theta_\alpha(\mathbf{r})\mathbf{E}_t$, where finding of the field $\mathbf{E}_t$ requires the full solution of the scattering problem).

Let us consider the expression $\nabla(\mathbf{E}_1 \times \mathbf{H}_2^* + \mathbf{E}_2 \times \mathbf{H}_1^*)$. Using the vector relationship $\nabla(\mathbf{A} \times \mathbf{B}) = \mathbf{B}\nabla \times \mathbf{A} - \mathbf{A}\nabla \times \mathbf{B}$ and taking into account (A2) and (A3), we have

$$\begin{aligned} \nabla(\mathbf{E}_1 \times \mathbf{H}_2^* + \mathbf{E}_2 \times \mathbf{H}_1^*) &= \mathbf{H}_2^* \nabla \times \mathbf{E}_1 \\ &\quad - \mathbf{E}_1 \nabla \times \mathbf{H}_2^* + \mathbf{H}_1^* \nabla \times \mathbf{E}_2 - \mathbf{E}_2 \nabla \times \mathbf{H}_1^*. \end{aligned} \quad (\text{A4})$$

Let us express rotors included in the right-hand side of this expression according to (A2) and (A3):

$$\begin{aligned} \nabla(\mathbf{E}_1 \times \mathbf{H}_2^* + \mathbf{E}_2 \times \mathbf{H}_1^*) &= (i\omega\mu_0)(\mathbf{H}_2^* \mathbf{H}_1 + \mathbf{H}_1^* \mathbf{H}_2) \\ &\quad + (k_0^2/(i\omega\mu_0))(\mathbf{E}_1 \mathbf{E}_2^* + \mathbf{E}_2 \mathbf{E}_1^*) - (\mathbf{E}_1 \mathbf{q}_2^* + \mathbf{E}_2 \mathbf{q}_1^*)/(i\omega\mu_0). \end{aligned}$$

By taking now the real part from both the parts of this equation, we obtain the relationship, which is equivalent

to (12):

$$\nabla \text{Re}(\mathbf{E}_1 \times \mathbf{H}_2^* + \mathbf{E}_2 \times \mathbf{H}_1^*) = \text{Re}[(\mathbf{E}_1 \mathbf{q}_2^* + \mathbf{E}_2 \mathbf{q}_1^*) / (i\omega\mu_0)], \quad (\text{A5})$$

which we will call the „duality lemma“ for the cross flows.

Unlike the Lorentz lemma (A1), instead of the external currents $\mathbf{j}_\alpha$ the right-hand side (A5) has the full currents $\mathbf{q}_\alpha$ that take into account dissipation inside the medium (when permittivity has an imaginary part). Another difference from the Lorentz lemma related to reciprocity of the fields is that the condition (A5) that generalizes the condition of preservation of the average flows (15) is fulfilled in the case of nonreciprocal media, too.