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Enhanced scattering and absorption due to the presence of a particle close to an interface

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Abstract: We study the influence of the presence of an interface on the scattering by a Rayleigh scatterer. The influence of an interface on the spontaneous emission has been known for many years. Here, we study the influence on the extinction cross-section and absorption cross-section. We provide a detailed analysis of interference and near-field effects. We show that the presence of a Rayleigh scatterer may *enhance* the specular reflection or specular transmission under certain conditions. Finally, we analyze the enhancement of absorption in the bulk in the presence of a small scatterer.

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1. Introduction

The purpose of this paper is to report an analysis of the scattering and absorption by a small particle in medium 1 in the presence of an interface separating medium 1 from medium 2 (see Fig. 1). The study has been originally motivated by a number of recent works attempting to enhance absorption in photovoltaic cells by using particles deposited on interfaces [1–8]. For this particular application, two basic phenomena are expected to take place: i) an antireflection effect that favors the transmission of propagating waves into the absorbing substrate, ii) a local enhancement of the near-field producing a localized enhanced absorption in the substrate. While a number of numerical simulations have been used to study these effects, it appears that a theoretical analysis of the basic concepts involved would be useful in order to gain a deeper physical understanding of this system.

As a Rayleigh scatterer is a dipolar scatterer. The process studied here is very similar to the emission by a dipole in the vicinity of an interface [9–13]. This problem has been studied thoroughly since the pioneering work of Drexhage demonstrating the effect of an interface on the lifetime of quantum emitters [9]. It is now well understood that the local density of states varies with the distance to the interface. In the language of quantum physics, this entails that the lifetime varies according to Fermi golden rule. The classical electrodynamics point of view offers an alternative picture: the power radiated by a monochromatic dipole with fixed dipole moment depends on its distance to the interface. Within this picture, the dependence is viewed as an interference effect between the power radiated by the dipole directly into medium 1 or after reflection on the interface between medium 1 and medium 2. In this paper, we are not interested in spontaneous emission but in scattering. We will show that the scattering cross-section also depends on the distance between the particle and the interface.

An important issue when studying scattering by a particle is energy conservation. When

using particles to increase energy absorption, controlling the accuracy of energy conservation is of critical importance. It is well-known that the optical theorem is not satisfied by the simplest form of the polarizability for a spherical scatterer in a homogeneous medium. In order to ensure energy conservation, a correction needs to be included. This was first discussed by Draine [14] who pointed out that the effect of radiation reaction has to be included. This amounts to introduce an effective polarizability as discussed in refs. [14–18]. Here, we point out that the polarizability correction depends on the extinction cross-section of the particle. As the latter depends on the distance to the interface, the effective polarizability needs to be revisited.

Finally, an important specificity of the scattering in the presence of an interface is the existence of two different specular components, one in reflection and one in transmission. When dealing with a scatterer in a homogeneous medium, the light scattered in the forward direction interferes with the incident beam. This is always a destructive interference that translates into a decay of the power specularly transmitted. This decay balances the energy scattered or absorbed [19]. When placing a particle above an interface, the scattered field can interfere with both the specularly transmitted and reflected fields. The interference depends on the distance z_0 between the particle and the interface in a different way so that it can be constructive in transmission and destructive in reflection. It follows that the specularly reflected or transmitted fields can be increased due to the presence of a scatterer. As energy needs to be conserved, an enhancement of the reflected specular beam requires a decay of the specularly transmitted beam and vice versa. Hence, the particle allows transferring energy between specularly reflected and transmitted beams. This point will be analyzed in detail using the optical theorem.

The paper is organized as follows: in the next section, we introduce the notations used throughout the paper. In the third section, we derive the form of the scattered field by a particle illuminated by a plane wave of arbitrary angle of incidence and arbitrary polarization. The generalized optical theorem is introduced in section 4. Finally, we discuss the absorption induced in the substrate by the presence of a particle in section 5.

2. Geometry and definitions

The system that we are interested in, is scattering from a small particle above an interface. If the particle is small compared to the wavelength, it can be modeled as an electric dipole with a polarizability that depends on the particle and the environment characteristics. A schematic description is illustrated in Fig. 1. A time dependence $e^{-i\omega t}$ is assumed throughout the paper. In surface optics, it is convenient to introduce a wave vector parallel to the interface, i.e. in the (x, y) plane,

$$\mathbf{k}_{\parallel} = (k_x, k_y) \quad (1)$$

which is real, and a complex z-component

$$k_{zj} = \sqrt{k_j^2 - k_{\parallel}^2} \quad (2)$$

whose imaginary part is chosen non-negative with $k_j^2 = \epsilon_j(\omega/c)^2$ where ϵ_j is the dielectric constant in medium $j = 1, 2$. The scattered plane is defined by the direction of the scattered light and the $\hat{\mathbf{z}}$ direction. For waves propagating upward, the direction of propagation is given by

$$\hat{\mathbf{u}}_{j+} = (k_{\parallel} \cos \phi, k_{\parallel} \sin \phi, k_{zj})/k_j \quad (3)$$

whereas for waves propagating downward,

$$\hat{\mathbf{u}}_{j-} = (k_{\parallel} \cos \phi, k_{\parallel} \sin \phi, -k_{zj})/k_j. \quad (4)$$

To define the polarization, it is convenient to use a unit vector which is the component of $\hat{\mathbf{u}}$ parallel to the xy plane, $\hat{\mathbf{k}} = \mathbf{k}_{\parallel}/k_{\parallel}$. Accordingly, the direction of s polarization is defined by,

$$\hat{\mathbf{s}} = \hat{\mathbf{k}} \times \hat{\mathbf{z}} \quad (5)$$

and the direction of p polarization derived from the orthogonal triad $(\hat{\mathbf{s}}, \hat{\mathbf{k}}, \hat{\mathbf{z}})$

$$\hat{\mathbf{p}}_{j\pm} = \hat{\mathbf{s}} \times \hat{\mathbf{u}}_{j\pm}. \quad (6)$$

It should be emphasized that $\hat{\mathbf{k}}$ and $\hat{\mathbf{s}}$ are always real whereas $\hat{\mathbf{p}}$ can be complex, depending on the medium and whether the wave is propagating or evanescent. In any case, $\hat{\mathbf{p}}$ is calculated in the same manner such that $\hat{\mathbf{p}} \cdot \hat{\mathbf{p}} = 1$ [20].

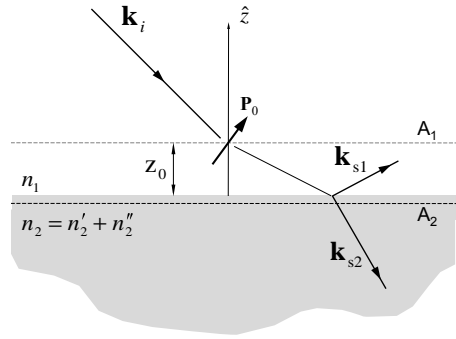


Fig. 1. Schematic set-up of scattering from a small particle – a dielectric sphere with refractive index $n_s = 2$ and radius $a = 50 \text{ nm}$ modeled by a dipole moment \mathbf{P}_0 .

3. Scattering by a particle above an interface

Scattering by a small particle above an interface bears some resemblance to fluorescence by a molecule close to an interface [10–12]. However, there are some key differences: in emission, the amplitude of the dipole moment is fixed whereas in scattering the magnitude and the direction of the dipole are determined by the interference between the incident and reflected fields. Furthermore, unlike fluorescence, the scattered field is coherent with the incident field. The resulting interference plays an important role in the extinction cross-section as we shall see.

The scattering process can be described as follows. First, an incident wave impinges on the particle directly and via reflection from the interface. The particle is excited at frequency ω of the incident field. In this step, the particle is modeled as a dipole with an effective polarizability that accounts for the multiple scattering between the particle and the interface. The scattered field is computed in a second step which is similar to the emission problem. While each of these steps is well-known [21–23], it appears that there is still a need for a comprehensive discussion of the influence of near-field effects and coherent effects on the extinction.

3.1. Excitation of a dipole

We begin with the polarization of the incident wave in medium 1,

$$\hat{\mathbf{e}}_i = \begin{cases} (0, -1, 0) & s - \text{polarization} \\ (k_{z1}, 0, k_{\parallel})/k_1 & p - \text{polarization} \end{cases} \quad (7)$$

The amplitude of the incident field at the location of the scatterer $\mathbf{r}_0 = (0, 0, z_0)$ is chosen such that it is unity at the plane $z = z_0$. It can be written as $\mathbf{E}_i = \hat{\mathbf{e}}_i \exp[ik_{z1}(z_0 - z)]$. The field at \mathbf{r}_0 in absence of the particle is composed of the incident and the reflected fields. It can be written in dyadic notation

$$\mathbf{E}_d(\mathbf{r}_0) = \left[\left(1 + r_{12}^s e^{2ik_{z1}z_0} \right) \hat{\mathbf{s}}\hat{\mathbf{s}} + \left(\hat{\mathbf{p}}_{1-}\hat{\mathbf{p}}_{1-} + r_{12}^p e^{2ik_{z1}z_0} \hat{\mathbf{p}}_{1+}\hat{\mathbf{p}}_{1-} \right) \right] \hat{\mathbf{e}}_i \quad (8)$$

where $r_{12}^{s,p}$ is the reflection coefficient on the interface for s and p polarization, respectively. For an interface between two semi-infinite layers, the Fresnel factors are given by

$$r_{ij}^{s,p} = \frac{Q_i - Q_j}{Q_i + Q_j}, \quad t_{ij}^s = \frac{2Q_i}{Q_i + Q_j}, \quad t_{ij}^p = \frac{n_i}{n_j} \frac{2Q_i}{Q_i + Q_j}, \quad (9)$$

where n is the refractive index and $Q_j = k_{zj}$ for s polarization or k_{zj}/ϵ_j for p polarization. This form can be generalized to any planar multilayer system by introducing the corresponding reflection factors.

3.2. Effective polarizability of a dipole

The dipole moment is related to the incident field by the polarizability α

$$\mathbf{p}_0 = \epsilon_1 \alpha \mathbf{E}_d. \quad (10)$$

Usually, a small particle is characterized by the quasi-static polarizability $\alpha_0 = 4\pi a^3(m^2 - 1)/(m^2 + 2)$ where $m^2 = \epsilon_{\text{sphere}}/\epsilon_1$, which is a good approximation for most purposes. It has been known for a long time that if the scatterer is in a given environment, then the multiple scattering between the dipole and the environment can be accounted for by introducing an effective polarizability. This effective polarizability has been first introduced by Efrima and Metiu in order to explain possible enhancements of the scattering by a molecule [24]. A similar discussion can also be found in refs. [16, 18] where the focus was on the enhancement of the scattering cross section due to a surface wave resonance of either the particle or the surface. Finally, the concept of effective polarizability has been discussed by Draine from a different perspective. He noted that a lossless Rayleigh scatterer does not satisfy energy conservation. Indeed, the extinction cross-section is proportional to $\text{Im}(\alpha_0)$ and therefore vanishes if m^2 is real. Draine pointed out that by including the radiation reaction, an effective polarizability can be obtained in the form

$$\alpha_{\text{eff}} = \frac{\alpha_0}{1 - \zeta_0 \alpha_0} \quad (11)$$

where ζ_0 is the radiation damping correction. In a homogeneous medium (herein, the host medium is medium 1), it is given by [14],

$$\zeta_0 = i \frac{k_1^3}{6\pi}. \quad (12)$$

This form does satisfy energy conservation so that he proposed to use it to improve the convergence of the discrete dipole approximation (DDA), a numerical technique introduced by Purcell and Pennypacker to study the scattering by particles [25]. This question was further discussed in refs. [17, 26]. A more general derivation can be found in ref. [15].

In the case of scattering by a particle located in the vicinity of an interface, a careful consideration of the effective polarizability is of critical importance. Indeed, it is well known that when a dipole is located at a subwavelength distance from a dielectric interface, it can radiate into modes with an angle of propagation beyond the critical angle so that more energy is radiated. Furthermore, if the underlying medium is lossy, non-radiative coupling can also increase

the power emitted by the dipole. It follows that energy conservation can only be obtained if these effects are properly accounted for by the correct effective polarizability, i.e. the radiation reaction is affected not only by the hosting medium but also by reflection from the interface. A detailed derivation of the effective polarizability accounting for these effects is given in Appendix A. The result can be cast in the form:

$$\overset{\leftrightarrow}{\alpha}_{\text{eff}} = \alpha_0 [\overset{\leftrightarrow}{\mathbf{I}} - \overset{\leftrightarrow}{\zeta} \alpha_0]^{-1}, \quad (13)$$

where $\overset{\leftrightarrow}{\mathbf{I}}$ is the unit dyadic and

$$\overset{\leftrightarrow}{\zeta} = \zeta_0 \overset{\leftrightarrow}{\mathbf{I}} + \varepsilon_1 \overset{\leftrightarrow}{G}_r(\mathbf{r}_0, \mathbf{r}_0) \quad (14)$$

where $\overset{\leftrightarrow}{G}_r(\mathbf{r}, \mathbf{r}_0)$ is the Green tensor that accounts for reflection at the interface and relates the dipole at \mathbf{r}_0 to the field at \mathbf{r} . In our formalism, it can be calculated by a Fourier transform,

$$\overset{\leftrightarrow}{G}_r(\mathbf{r}, \mathbf{r}_0) = \int_{-\infty}^{+\infty} \frac{d^2 \mathbf{k}_{\parallel}}{(2\pi)^2} \overset{\leftrightarrow}{G}_r(\mathbf{k}_{\parallel}; z - z_0) e^{i\mathbf{k}_{\parallel} \cdot \mathbf{R}} \quad (15)$$

where $\mathbf{R} = (x, y)$ and substituting $\mathbf{r} = \mathbf{r}_0$.

3.3. Scattering from a dipole above an interface

Here, we give the form of the field radiated by a point-like dipole located at \mathbf{r}_0 with an electric dipole moment \mathbf{p}_0 . The corresponding dipole polarization can be cast in the form:

$$\mathbf{P}(\mathbf{r}) = \mathbf{p}_0 \delta(\mathbf{r} - \mathbf{r}_0), \quad (16)$$

where δ is the Dirac delta function. It is useful to introduce the Fourier transform $\mathbf{F}(\mathbf{k}_{\parallel}; z)$ of the scattered field $\mathbf{E}(\mathbf{r})$:

$$\mathbf{E}(\mathbf{r}) = \int \frac{d^2 \mathbf{k}_{\parallel}}{(2\pi)^2} \mathbf{F}(\mathbf{k}_{\parallel}; z) e^{i\mathbf{k}_{\parallel} \cdot \mathbf{R}}. \quad (17)$$

The scattered field is related to the source by the dyadic Green tensor,

$$\mathbf{F}(\mathbf{k}_{\parallel}; z) = \int \overset{\leftrightarrow}{G}(\mathbf{k}_{\parallel}; z - z') \cdot \mathbf{P}(\mathbf{k}_{\parallel}; z') dz', \quad (18)$$

so that the scattered field is given by:

$$\mathbf{F}(\mathbf{k}_{\parallel}; z) = \overset{\leftrightarrow}{G}(\mathbf{k}_{\parallel}; z - z_0) \cdot \mathbf{p}_0. \quad (19)$$

The Green tensor for a source located at z_0 above an interface and an observation point into medium 1 is composed of the contribution of the free space radiation $\overset{\leftrightarrow}{G}_0$ and reflection from the interface $\overset{\leftrightarrow}{G}_r$,

$$\overset{\leftrightarrow}{G} = \overset{\leftrightarrow}{G}_0 + \overset{\leftrightarrow}{G}_r, \quad (20)$$

where we adopt Sipe's formalism to present the Green tensor [20],

$$\begin{aligned} \overset{\leftrightarrow}{G}_0(\mathbf{k}_{\parallel}; z - z_0) &= \frac{i}{2} \left(\frac{\omega}{c} \right)^2 \frac{e^{ik_{z1}(z-z_0)}}{k_{z1}} (\hat{\mathbf{s}}\hat{\mathbf{s}} + \hat{\mathbf{p}}_{1+}\hat{\mathbf{p}}_{1+}) \\ \overset{\leftrightarrow}{G}_r(\mathbf{k}_{\parallel}; z - z_0) &= \frac{i}{2} \left(\frac{\omega}{c} \right)^2 \frac{e^{ik_{z1}(z+z_0)}}{k_{z1}} (r_{12}^s \hat{\mathbf{s}}\hat{\mathbf{s}} + r_{12}^p \hat{\mathbf{p}}_{1+}\hat{\mathbf{p}}_{1-}) . \end{aligned} \quad (21)$$

Inserting Eq. (21) into Eq. (15) yields the Green tensor at \mathbf{r}_0 . The numerical integration requires some care. Note that at large parallel wave vector, $k_{z1} \sim ik_{\parallel}$ such that the exponent term in Eq. (21) decays. This attenuation ensures the convergence of the integral in Eq. (15) for positive values of z_0 . A detailed discussion on the numerical calculation is given in Appendix A of ref. [27]. Following Eq. (19), the electric field scattered in medium 1 (reflection) is given by

$$\mathbf{F}_{s1}(\mathbf{k}_{\parallel}; z > z_0) = \frac{i}{2} \left(\frac{\omega}{c} \right)^2 \frac{e^{ik_{z1}(z-z_0)}}{k_{z1}} \times \left[\left(1 + r_{12}^s e^{2ik_{z1}z_0} \right) \hat{\mathbf{s}}\hat{\mathbf{s}} + \left(\hat{\mathbf{p}}_{1+}\hat{\mathbf{p}}_{1+} + r_{12}^p e^{2ik_{z1}z_0} \hat{\mathbf{p}}_{1+}\hat{\mathbf{p}}_{1-} \right) \right] \cdot \mathbf{p}_0. \quad (22)$$

In the same manner, we calculate the field scattered into medium 2 (transmission),

$$\mathbf{F}_{s2}(\mathbf{k}_{\parallel}; z < 0) = \frac{i}{2} \left(\frac{\omega}{c} \right)^2 \frac{e^{ik_{z1}z_0}}{k_{z1}} (t_{12}^s \hat{\mathbf{s}}\hat{\mathbf{s}} + t_{12}^p \hat{\mathbf{p}}_{2-}\hat{\mathbf{p}}_{1-}) e^{-ik_{z2}z} \cdot \mathbf{p}_0. \quad (23)$$

So far, we have derived the scattered fields for the system of a small particle above an interface. We stress that all the equations above accounts for the general case of losses in the media and in the particle. An extension of the model to large particles can be done using the DDA [26, 27].

4. Energy Conservation and Optical Theorem

In this section, we investigate the energy conservation. For this purpose, we use the generalized optical theorem. Although the proof has been shown in various publications [28, 29], for the sake of completeness, we will show its derivation in Appendix B with the notation presented here. At this point, we would limit ourselves to the case of lossless particle hosted in a lossless medium. Under these conditions, there might be absorption only in the substrate and one obtains from energy conservation that

$$W_{ext}^{(r)} + W_{ext}^{(t)} = W_{sca}^{(r)} + W_{sca}^{(t)}, \quad (24)$$

where W_{sca} is the power scattered through surfaces A_1 (reflection) and A_2 (transmission), as shown in Fig. 1. We recall that energy conservation is fulfilled only when the effective polarizability, given in Eq. (13), is applied. Herein, W_{ext} stands for the extinction term which originates from the interference between the specularly reflected or transmitted beam with the scattered field. We see from the energy conservation equality that although the total extinction W_{ext} must be positive, $W_{ext}^{(r)}$ or $W_{ext}^{(t)}$ can be negative. In other words, the energy in the specularly reflected beam can be *increased* provided that the decay of the transmitted beam compensates the scattering power. This behavior is fundamentally different from the extinction of a particle in homogeneous medium which is always a decay term. In order to give a dimensionless number, it is common to introduce the scattering and extinction efficiencies, which are defined by

$$Q_{ext} = \frac{W_{ext}}{W_i(\pi a^2)}; \quad Q_{sca} = \frac{W_{sca}}{W_i(\pi a^2)}, \quad (25)$$

for spherical particles, and W_i is the incident power. The explicit form of the extinction efficiency is known as extinction theorem, given by Eq. (B.14). In case of lossless mediums, these expressions can be simplified to show explicitly the interference between the specular field (\mathbf{E}_r or \mathbf{E}_t) with the scattered field (\mathbf{F}_s)

$$Q_{ext}^{(r)} = -\frac{2}{\pi a^2} \text{Re} \{ \mathbf{E}_r \cdot \mathbf{F}_{s1}^* \}; \quad Q_{ext}^{(t)} = -\frac{2}{\pi a^2} \frac{k_{z2}}{k_{z1}} \text{Re} \{ \mathbf{E}_t \cdot \mathbf{F}_{s2}^* \}. \quad (26)$$

Note that in homogeneous medium, $\mathbf{E}_r = 0$, $\mathbf{E}_t = \mathbf{E}_i$, and if we write \mathbf{F}_{s2} as a function of \mathbf{E}_i using Eqs. (23), (10), and (8), and assuming $\mathbf{E}_i \cdot \mathbf{E}_i^* = 1$, we retrieve the extinction term $Q_{ext}^{(t)} \propto k \text{Im}(\alpha_{\text{eff}})$.

5. Results and discussion

To study the effect of the particle on the absorption in medium 2, we choose the system shown in Fig. 1 when the upper half space is vacuum and the substrate has a refractive index $n_2 = 2 + in_2''$. The particle is a small sphere of radius $a = 50 \text{ nm}$ with refractive index $n_s = 2$. The illumination wavelength (from the vacuum) is $\lambda = 1 \mu\text{m}$ such that the dipole approximation is satisfied. Since in this case there are no further losses in the virtual enclosure between A_1 and A_2 , the contribution of the particle on the absorption in medium 2 is easier to be discerned.

5.1. Absorption and extinction cross section

Here, we focus on the transmission to medium 2. Using Eq. (B.15), we plot the scattering efficiency into medium 2, under normal incident angle, as a function of the particle position z_0 . For lossy media with $n_2'' > 0$, all the power transmitted is eventually absorbed because we consider here a semi-infinite space. Hence,

$$Q_{abs,2} = Q_{sca}^{(t)}. \quad (27)$$

The results are shown in Fig. 2. In case medium 2 is transparent ($n_2'' = 0$), we see oscillations at $z_0 > \lambda/2$ and enhanced transmission at small distances. The oscillations are clearly due to the interference. The enhancement for small distances can be attributed to the excitation of plane waves propagating in the dielectric beyond the critical angle. Note that this enhancement cannot be recovered by using only the quasi-static polarizability α_0 , regardless of the particle size. As an example of lossy medium, we choose $n_2'' = 0.5$ which is a typical value for semiconductors in the visible range; e.g. Si, GaAs, Cu(InGa)Se₂, etc. For $z_0 > \lambda/2$ there are oscillations as in the transparent case but with a phase shift which results from the reflection factor phase. At small distances, the power absorbed in medium 2 is much larger and enhanced sharply at $z_0 < \lambda/10$. Here, the dipole excites not only modes propagating beyond the critical angle but also modes with larger parallel wave vectors. Finally, we consider a substrate with $n_2'' = 5$ so that the real part of the dielectric constant $\epsilon_2' = n_2'^2 - n_2''^2 < -1$. This value corresponds to the case of a metal such that a plasmon can propagate. When the particle is far from the surface, there is a small absorption due to the large reflection factor of the metal. However, in the very near field regime, the absorption increases dramatically.

The reason for the enhanced absorption as the particle approaches the surface stems from

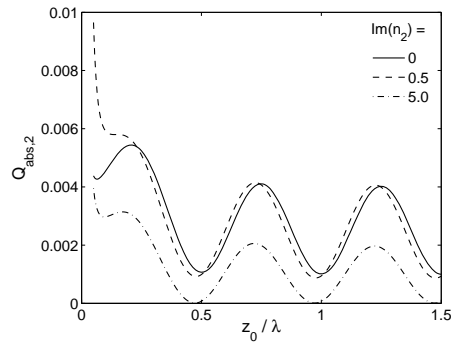


Fig. 2. Absorption efficiency depends on the particle height from the surface for transparent (line), lossy (dash), and metallic-like (dash-dot) substrates. Illumination in normal direction.

coupling of the dipole to non-radiative waves in medium 2. When the particle is far from the interface, the non-radiative waves decay and there is no contribution by the presence of the particle. At distances smaller than the decay length, additional radiative and non-radiative channels contribute to the energy transfer into medium 2. To better understand this mechanism, we calculate the absorption efficiency as a function of the parallel wave vector k_{\parallel} (cf. Fig. 3). The parallel wave vector can be understood (at least for lossless mediums) as $k_{\parallel} = k_1 \sin \theta_1 = k_2 \sin \theta_2$. Therefore, we can distinguish between three regions. First, propagating waves in mediums 1 and 2 ($0 < k_{\parallel} < k_1$). Assuming $n_2 > n_1$, in this representation it is evident to see the critical angle in medium 2 which corresponds to $k_{\parallel} = k_1$. Second region, evanescent waves in medium 1 and propagating waves in medium 2 in directions beyond the critical angle ($k_1 < k_{\parallel} < k_2$). Third region, non-radiative waves in mediums 1 and 2 ($k_2 < k_{\parallel}$).

In Fig. 3(a) we see the dependent of $Q_{abs}(k_{\parallel})$ when the particle touches the surface, i.e. $z_0 = a$, for the three substrates we have introduced above. For comparison, we show the absorption efficiency when the particle is at $z_0 = \lambda/2$ in Fig. 3(b). Without losses, there is only transmission of evanescent waves in medium 1 to propagating waves in medium 2 [part (a)]. If the distance increases [part (b)], the contribution of evanescent waves vanishes. For lossy substrate, the absorption increases due to further coupling of evanescent waves in medium 1 to non-radiative waves in medium 2 with arbitrary large values of the wave vector. Regarding the metallic-like substrate, we see the coupling to surface waves as a peak in the absorption at $k_{sw} = k_1[\epsilon_1\epsilon_2/(\epsilon_1 + \epsilon_2)]^{1/2} \approx k_1$, for the refractive index we used. The further contribution at larger wave vectors is not due to surface plasmon but to absorption in the medium. This is the *quenching effect* known in fluorescence of molecules near metallic films.

The metallic substrate may have a different effect on the absorption at a particular frequency coinciding with the horizontal asymptote of the dispersion relation. In metals such as silver or gold, the plasma frequency lies in the UV-visible range. Therefore, placing particles near

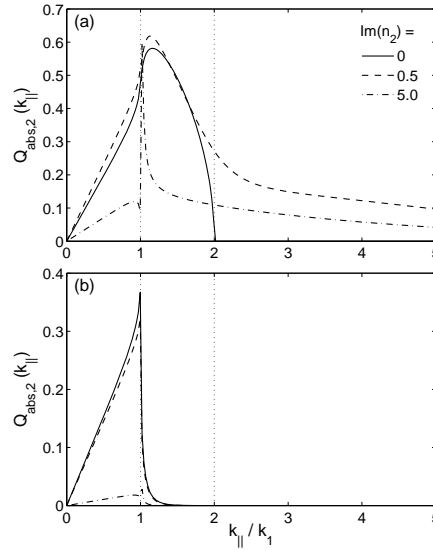


Fig. 3. Absorption efficiency as a function of the parallel wave vector, k_{\parallel} , in medium 2 due to (a) particle on the interface ($z_0 = a$), and (b) particle at $z_0 = \lambda/2$ for transparent (line), lossy (dash), and metallic-like (dash-dot) substrates. The dotted vertical lines present the wave vectors in medium 1 and 2.

these kind of metals will result in enhanced resonance absorption. To demonstrate this effect, we show in Fig. 4 the absorption efficiency in silver when a transparent particle ($n_s = 2$) is placed on the interface. The illumination frequency corresponds to the solar spectrum in normal incident direction. The loci of maximum absorption in the plane (k, ω) clearly reproduces the dispersion relation of the surface plasmons.

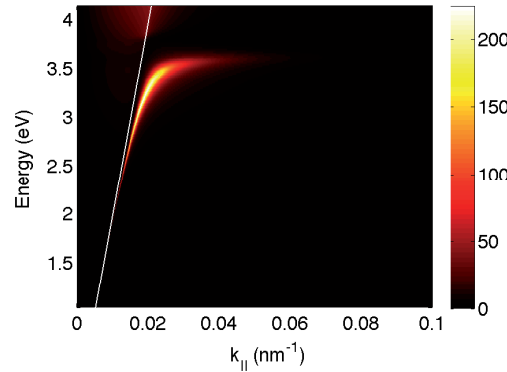


Fig. 4. Absorption efficiency in the (k, ω) plane when the substrate is silver and the particle is placed on the surface. Illumination in normal incident direction corresponds to wavelength between 300 nm and 1200 nm. The white line denotes the light-line in medium 1.

5.2. Modifying the specular fields

So far, we have discussed the effect of the particle on the absorption and scattering cross section. In the following, we discuss its effect on the extinction of the specular fields. For a particle located in a homogeneous medium, scattering always result in an attenuation of the specular field so that it is tempting to consider that scattering also reduces the specular terms both in reflection and in transmission. However, we will show that scattering can be used to enhance the specular terms in the presence of an interface. In this section, we limit the discussion to the case of a single particle above an interface. For a practical application, a system consisting in a number of N particles is more significant. Yet, we note that for specular directions, the phase of the scattered field does not depend on the scatterer position. Hence, the fields scattered by any number of particles located at the same height z_0 above the interface interfere constructively in the specular direction. It follows that the following discussion can be applied to any number of particles inasmuch as multiple scattering between particles is neglected.

We computed the extinction efficiency for reflection $Q_{ext}^{(r)}$, transmission $Q_{ext}^{(t)}$, and their sum denoted as total extinction efficiency Q_{ext} , as a function of the particle height (see Fig. 5). Without losses [part (a)], we see oscillations of the reflected and transmitted extinction. Note that the total extinction is relatively low (scale on the right). In addition, it is intriguing to see that the reflection and transmission change sign as a function of the particle height. A positive sign means that the power is attenuated with respect to reflection or transmission from a flat interface without particle, and a negative sign means that the power is enhanced. Thus, power is transferred between the reflected and transmitted beams. In particular, by positioning a particle on the interface ($z_0 = a$), the specular reflection is reduced and the specular transmission is increased. This behavior contributes to the antireflection effect mentioned in the Introduction section. Surprisingly, for a particle at $z_0 \sim 0.4\lambda$, the specular reflection is increased. Since the

total extinction cross section is an order of magnitude smaller than the peak oscillations of $Q_{ext}^{(r,t)}$, it means that most of the effect of small particles is due to modifying the specular factors rather than scattering light.

We now consider the effect of losses for a lossy dielectric [Fig. 5(b)]. By increasing the losses of the substrate, the amplitude of the extinction efficiency is not changed much except in the near field where it increases due to quenching. It is interesting to see that whatever the losses, the extinction is zero when the particle is at $z_0 \approx \lambda/2$. It can be explained more clearly in the metal case [Fig. 5(c)]. When light impinges on the surface at normal direction, the interference with the reflected field produces a minimum at $\lambda/2$. Therefore, placing the particle there will not affect the scattering nor the extinction. On the other hand, when the particle is placed at $z_0 \approx \lambda/4$, the amplitude is maximum and so is the extinction efficiency. This is similar to thin metal film above a mirror, known as *Salisbury screen* used in infrared absorbers and antireflection coatings [30].

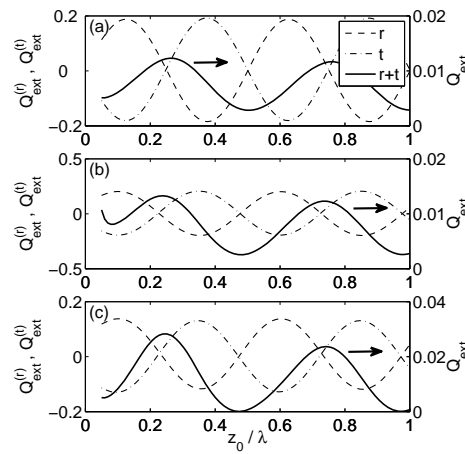


Fig. 5. Extinction efficiency of reflected (dash), transmitted (dash-dot) and total power (line, scale on the right) vs. the particle height from the surface for (a) transparent, (b) lossy, and (c) metallic-like substrates. Normal incident illumination.

In Fig. 6, we show the dependence of the extinction efficiency on the direction of the incident angle. The illumination is with unpolarized light (average of s and p polarization). As can be seen, there is a small variation in the extinction terms versus the incident angle. The maximum at the total extinction in the weak losses case [parts (a) and (b)] can be understood by the orientation of the dipole which is determined by interference of the incident and reflected fields. Clearly, the direction of s polarized fields does not depend on the angle of incidence. Regarding p polarization, the fields interfere constructively in the direction perpendicular to the interface for angles smaller than the Brewster's angle ($\theta_B = 64.43^\circ$) thereby enhancing the transmission. Similarly, at incident angles larger than the Brewster's angle, the transmission decreases since constructive interference is in direction parallel to the interface [11]. This behavior is advantageous for solar cells application since it means that the total extinction (and the scattering) does not depend on the incident angle up to ~ 60 degrees.

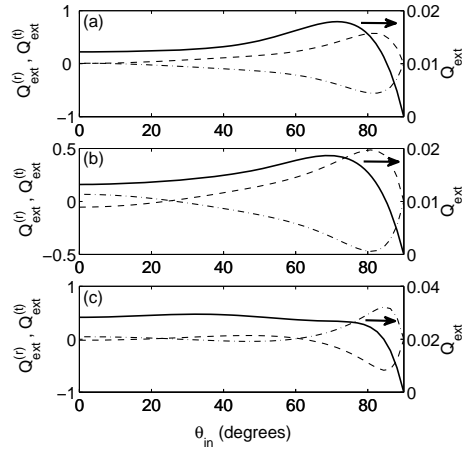


Fig. 6. Extinction efficiency of reflected (dash), transmitted (dash-dot) and total power (line, scale on the right) as a function of the angle of incidence for (a) transparent, (b) lossy, and (c) metallic-like substrates. Illuminating the particle on the interface ($z_0 = a$), by an unpolarized light.

6. Conclusion

We have studied the scattering from a particle near an interface in order to investigate quantitatively its effect on the absorption of the substrate. Our calculation included the effective polarizability of the particle, taking into account radiation damping correction in the presence of an interface. This contribution enables to satisfy energy conservation. As the particle approaches the interface, the absorption by the substrate is enhanced due to radiative and non-radiative modes. In addition, it was shown by the extinction theorem that the specular reflection (or transmission) can be attenuated or enhanced according to the particle position. This study can be utilized to increase the absorption within solar cell devices.

A. Effective polarizability

In this appendix, we derive Eq. (13) and (14), following the same procedure as in Appendix A of ref. [15]. Assume a particle of volume V with a dielectric constant ϵ_s embedded in a medium with a dielectric constant ϵ_h . The total field at a position \mathbf{r} is calculated by

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_{\text{exc}}(\mathbf{r}) + \int_V \overset{\leftrightarrow}{G}(\mathbf{r}, \mathbf{r}') (\epsilon_s - \epsilon_h) \mathbf{E}(\mathbf{r}') d^3 \mathbf{r}', \quad (\text{A.1})$$

where \mathbf{E}_{exc} is an external field exciting the scatterer. In the presence of an interface, the Green tensor $\overset{\leftrightarrow}{G}$ is the sum of the free space Green tensor $\overset{\leftrightarrow}{G}_0$ and a term $\overset{\leftrightarrow}{G}_r$ accounting for the reflection on the interface. Thus, Eq. (A.1) can be rewritten:

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_{\text{exc}}(\mathbf{r}) + \int_V [\overset{\leftrightarrow}{G}_0(\mathbf{r}, \mathbf{r}') + \overset{\leftrightarrow}{G}_r(\mathbf{r}, \mathbf{r}')] \epsilon_h (m^2 - 1) \mathbf{E}(\mathbf{r}') d^3 \mathbf{r}' \quad (\text{A.2})$$

where $m^2 = \epsilon_s / \epsilon_h$. The Green tensor in a homogeneous medium is well known

$$\overset{\leftrightarrow}{G}_0(\mathbf{r}, \mathbf{r}_0) = \text{PV} [k_h^2 \overset{\leftrightarrow}{\mathbf{I}} + \nabla \nabla] \frac{\exp(ik_h \rho)}{4\pi \epsilon_h \rho} - \frac{\overset{\leftrightarrow}{\mathbf{I}}}{3\epsilon_h} \delta(\mathbf{r} - \mathbf{r}_0), \quad (\text{A.3})$$

where PV denotes the principal value and $\rho = |\mathbf{r} - \mathbf{r}_0|$. We note that $\overset{\leftrightarrow}{G}_0$ is singular at $\rho = 0$. As discussed in ref. [15], when $\rho \rightarrow 0$, one obtains

$$\overset{\leftrightarrow}{G}_0(\mathbf{r}, \mathbf{r}_0) \approx -\frac{\overset{\leftrightarrow}{\mathbf{I}}}{3\epsilon_h} \delta(\mathbf{r} - \mathbf{r}_0) + i \frac{k_h^2}{6\pi\epsilon_h} \overset{\leftrightarrow}{\mathbf{I}}. \quad (\text{A.4})$$

Substituting the value of $\overset{\leftrightarrow}{G}_0(\mathbf{r}, \mathbf{r}_0)$ into Eq. (A.2) and taking the particle as a small sphere of radius a , we derive

$$\mathbf{E}(\mathbf{r}_0) = \mathbf{E}_{\text{exc}}(\mathbf{r}_0) + [i \frac{k_1^3}{6\pi\epsilon_h} \overset{\leftrightarrow}{\mathbf{I}} + \overset{\leftrightarrow}{G}_r(\mathbf{r}_0, \mathbf{r}_0)] V \epsilon_h (m^2 - 1) \mathbf{E}(\mathbf{r}_0) - \frac{\overset{\leftrightarrow}{\mathbf{I}}(m^2 - 1)}{3} \mathbf{E}(\mathbf{r}_0) \quad (\text{A.5})$$

Expressing the total field as a function of the excited field yields

$$\mathbf{E}(\mathbf{r}_0) = \frac{3\mathbf{E}_{\text{exc}}(\mathbf{r}_0)}{m^2 + 2} \left\{ \overset{\leftrightarrow}{\mathbf{I}} - 4\pi a^3 \frac{m^2 - 1}{m^2 + 2} \left[\frac{ik_h^3}{6\pi} \overset{\leftrightarrow}{\mathbf{I}} - \epsilon_h \overset{\leftrightarrow}{G}_r(\mathbf{r}_0, \mathbf{r}_0) \right] \right\}^{-1} \quad (\text{A.6})$$

The dipole moment can be described by

$$\mathbf{p} = V(\epsilon_s - \epsilon_h) \mathbf{E}(\mathbf{r}_0) = \epsilon_h \overset{\leftrightarrow}{\alpha}_{\text{eff}} \mathbf{E}_{\text{exc}}(\mathbf{r}_0) \quad (\text{A.7})$$

Using Eqs. (A.6) and (A.7), and noting that $\alpha_0 = 4\pi a^3(m^2 - 1)/(m^2 + 2)$, the effective polarizability can be derived as

$$\overset{\leftrightarrow}{\alpha}_{\text{eff}} = \alpha_0 \left\{ \overset{\leftrightarrow}{\mathbf{I}} - \left[\frac{ik_h^3}{6\pi} \overset{\leftrightarrow}{\mathbf{I}} + \epsilon_h \overset{\leftrightarrow}{G}_r(\mathbf{r}_0, \mathbf{r}_0) \right] \alpha_0 \right\}^{-1}. \quad (\text{A.8})$$

B. Generalized optical theorem

In this section, we show that energy conservation is fulfilled to the system of scattering by a single particle near an interface. In principle, the optical theorem can be formulated in the general case of lossy mediums and lossy particle. However, for the purpose of this paper, we discuss only the case where medium 2 absorbs. In the derivation of the optical theorem, we construct an imaginary enclosure that contains the dipole. It is advantageous to avoid losses within this enclosure. Hence, the surfaces of the enclosure are at $z = z_0^+$ and at $z = 0^-$, as depicted in Fig. 1 as A_1 and A_2 , respectively. Then, we integrate the Poynting vector over this enclosure. First, we consider scattering without particle, i.e. reflection and transmission from a flat interface through the enclosure. In the second step, we introduce the particle with the scattered fields that have been derived in Section 3.3 to find the relation between the scattering and the extinction powers.

B.1. Absence of particle

We begin in the case where there is no particle. In medium 1, the electric field is composed of the incident field and the specularly reflected field,

$$\mathbf{E}_1 = \mathbf{E}_i + \mathbf{E}_r \quad (\text{B.1})$$

with the corresponding magnetic fields. The electric field in medium 2 is only the specularly transmitted field

$$\mathbf{E}_2 = \mathbf{E}_t. \quad (\text{B.2})$$

The Poynting vector in each medium is

$$\begin{aligned}\mathbf{S}_1 &= \frac{1}{2} \text{Re}\{\mathbf{E}_1 \times \mathbf{H}_1^*\} = \frac{1}{2} \text{Re}\{(\mathbf{E}_i + \mathbf{E}_r) \times (\mathbf{H}_i^* + \mathbf{H}_r^*)\} \\ \mathbf{S}_2 &= \frac{1}{2} \text{Re}\{\mathbf{E}_2 \times \mathbf{H}_2^*\} = \frac{1}{2} \text{Re}\{\mathbf{E}_t \times \mathbf{H}_t^*\}.\end{aligned}\quad (\text{B.3})$$

Assuming there is no absorption in medium 1 nor by the particle, the Poynting vector over the enclosure equal zero,

$$\int_A \mathbf{S} \cdot d\mathbf{A} = \int_{A_1} \mathbf{S}_1 \cdot d\mathbf{A} + \int_{A_2} \mathbf{S}_2 \cdot d\mathbf{A} = 0 \quad (\text{B.4})$$

where $d\mathbf{A} = \pm \hat{\mathbf{z}} dx dy$ associated with A_1 and A_2 , respectively. For a flat surface we recover Fresnel's coefficients that satisfy energy conservation

$$-\frac{1}{2} \text{Re}\{\mathbf{E}_i \times \mathbf{H}_i^*\} + \frac{1}{2} \text{Re}\{\mathbf{E}_r \times \mathbf{H}_r^*\} + \frac{1}{2} \text{Re}\{\mathbf{E}_t \times \mathbf{H}_t^*\} = 0. \quad (\text{B.5})$$

B.2. Presence of particle

In the presence of a particle, the fields have also scattering components

$$\begin{aligned}\mathbf{E}_1 &= \mathbf{E}_i + \mathbf{E}_r + \mathbf{E}_{s1} \\ \mathbf{E}_2 &= \mathbf{E}_t + \mathbf{E}_{s2}\end{aligned}\quad (\text{B.6})$$

with the associated magnetic fields. The Poynting vector in each medium is then

$$\begin{aligned}\mathbf{S}_1 &= \frac{1}{2} \text{Re}\{(\mathbf{E}_i + \mathbf{E}_r + \mathbf{E}_{s1}) \times (\mathbf{H}_i^* + \mathbf{H}_r^* + \mathbf{H}_{s1}^*)\} \\ \mathbf{S}_2 &= \frac{1}{2} \text{Re}\{(\mathbf{E}_t + \mathbf{E}_{s2}) \times (\mathbf{H}_t^* + \mathbf{H}_{s2}^*)\}.\end{aligned}\quad (\text{B.7})$$

We rewrite the energy conservation as before, Eq. (B.4), when considering also the scattered fields and substitute the relations we obtained from flat surface without particle, Eq. (B.5). We then have

$$\begin{aligned}\int_{A_1} \frac{1}{2} \text{Re}\{(\mathbf{E}_i \times \mathbf{H}_{s1}^* + \mathbf{E}_{s1} \times \mathbf{H}_i^*) + (\mathbf{E}_r \times \mathbf{H}_{s1}^* + \mathbf{E}_{s1} \times \mathbf{H}_r^*) + (\mathbf{E}_{s1} \times \mathbf{H}_{s1}^*)\} \cdot d\mathbf{A} + \\ \int_{A_2} \frac{1}{2} \text{Re}\{(\mathbf{E}_t \times \mathbf{H}_{s2}^* + \mathbf{E}_{s2} \times \mathbf{H}_t^*) + (\mathbf{E}_{s2} \times \mathbf{H}_{s2}^*)\} \cdot d\mathbf{A} = 0.\end{aligned}\quad (\text{B.8})$$

We can decompose Eq. (B.8) to give each term its physical meaning,

$$\begin{aligned}W_{ext}^{(i)} &= - \int_{A_1} \frac{1}{2} \text{Re}\{\mathbf{E}_i \times \mathbf{H}_{s1}^* + \mathbf{E}_{s1} \times \mathbf{H}_i^*\} \cdot d\mathbf{A} \\ W_{ext}^{(r)} &= - \int_{A_1} \frac{1}{2} \text{Re}\{\mathbf{E}_r \times \mathbf{H}_{s1}^* + \mathbf{E}_{s1} \times \mathbf{H}_r^*\} \cdot d\mathbf{A} \\ W_{sca}^{(r)} &= \int_{A_1} \frac{1}{2} \text{Re}\{\mathbf{E}_{s1} \times \mathbf{H}_{s1}^*\} \cdot d\mathbf{A} \\ W_{ext}^{(t)} &= - \int_{A_2} \frac{1}{2} \text{Re}\{\mathbf{E}_t \times \mathbf{H}_{s2}^* + \mathbf{E}_{s2} \times \mathbf{H}_t^*\} \cdot d\mathbf{A} \\ W_{sca}^{(t)} &= \int_{A_2} \frac{1}{2} \text{Re}\{\mathbf{E}_{s2} \times \mathbf{H}_{s2}^*\} \cdot d\mathbf{A}\end{aligned}\quad (\text{B.9})$$

such that

$$W_{ext}^{(i)} + W_{ext}^{(r)} + W_{ext}^{(t)} = W_{sca}^{(r)} + W_{sca}^{(t)}. \quad (\text{B.10})$$

We distinguish between two terms: extinction and scattering. The extinction term is related to the energy removed from specular part (the sign minus is introduced to be consistent with the definition of extinction by Bohren and Huffman [19]). Note that in this notation a positive sign refers to attenuation of the radiated power in the specular direction (with respect to flat interface) whereas negative sign corresponds to enhancement. We will remark that in case the hosting medium of the particle is lossy, there is an additional absorption in the enclosure surface we defined above, $0^- < z < z_0^+$. In order to calculate the extinction terms, we recall that

$$\begin{aligned} \mathbf{E}_i &= \hat{\mathbf{e}}_i e^{i\mathbf{k}_i \cdot \mathbf{r}} \\ \mathbf{H}_i^* &= \frac{\mathbf{k}_i^*}{\omega\mu} \times \mathbf{E}_i^* \\ \mathbf{E}_s &= \int \frac{d^2\mathbf{k}_\parallel}{(2\pi)^2} \mathbf{F}_s e^{i\mathbf{k} \cdot \mathbf{r}} \\ \mathbf{H}_s^* &= \frac{\mathbf{k}_s^*}{\omega\mu} \times \mathbf{E}_s^* = \int \frac{d^2\mathbf{k}_\parallel}{(2\pi)^2} \frac{\mathbf{k}_s^*}{\omega\mu} \times \mathbf{F}_s^* e^{-i\mathbf{k} \cdot \mathbf{r}}. \end{aligned} \quad (\text{B.11})$$

As we have seen, the extinction terms have the form

$$W_{ext} = \int_A \frac{1}{2} \text{Re} \left\{ (\mathbf{E}_i \times \mathbf{H}_s^* + \mathbf{E}_s \times \mathbf{H}_i^*) \right\} \cdot d\mathbf{A}. \quad (\text{B.12})$$

Using the relation

$$\iint_{-\infty}^{\infty} e^{-i(\Delta k_x x + \Delta k_y y)} dx dy = (2\pi)^2 \delta(\Delta k_x) \delta(\Delta k_y), \quad (\text{B.13})$$

we derive for extinction the following expressions,

$$\begin{aligned} W_{ext}^{(i)} &= 0 \\ W_{ext}^{(r)} &= -\frac{1}{2\omega\mu} \text{Re} \left\{ \mathbf{E}_r \times \left[\mathbf{k}_r^* \times \mathbf{F}_{s1}^*(\hat{\mathbf{u}}_r) \right] + \mathbf{F}_{s1}(\hat{\mathbf{u}}_r) \times \left[\mathbf{k}_r^* \times \mathbf{E}_r^* \right] \right\} \cdot (+\hat{\mathbf{z}}) \\ W_{ext}^{(t)} &= -\frac{1}{2\omega\mu} \text{Re} \left\{ \mathbf{E}_t \times \left[\mathbf{k}_t^* \times \mathbf{F}_{s2}^*(\hat{\mathbf{u}}_t) \right] + \mathbf{F}_{s2}(\hat{\mathbf{u}}_t) \times \left[\mathbf{k}_t^* \times \mathbf{E}_t^* \right] \right\} \cdot (-\hat{\mathbf{z}}) \end{aligned} \quad (\text{B.14})$$

Similarly, the scattering powers into medium 1 (reflection) and medium 2 (transmission) are calculated

$$\begin{aligned} W_{sca}^{(r)} &= \frac{1}{2\omega\mu} \text{Re} \int \frac{d^2\mathbf{k}_\parallel}{(2\pi)^2} \left\{ \mathbf{F}_{s1}(\mathbf{k}_\parallel) \times \left[\mathbf{k}_s^* \times \mathbf{F}_{s1}^*(\mathbf{k}_\parallel) \right] \right\} \cdot (+\hat{\mathbf{z}}) \\ W_{sca}^{(t)} &= \frac{1}{2\omega\mu} \text{Re} \int \frac{d^2\mathbf{k}_\parallel}{(2\pi)^2} \left\{ \mathbf{F}_{s2}(\mathbf{k}_\parallel) \times \left[\mathbf{k}_s^* \times \mathbf{F}_{s2}^*(\mathbf{k}_\parallel) \right] \right\} \cdot (-\hat{\mathbf{z}}). \end{aligned} \quad (\text{B.15})$$

We emphasize that the integral is over all the spectrum of the parallel wave vectors. For $k_\parallel < k_j$, these are propagating waves in medium j and for $k_\parallel > k_j$ it corresponds to evanescent waves.

Acknowledgments

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