

# Non additivity of Poynting vector within opaque media

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The energy flux within an opaque medium near an interface is not the sum of an incident plus a reflected term, as there is a synergistic contribution to the time averaged Poynting vector which involves simultaneously both the incident and reflected fields. Therefore, the well known formula  $R + T = 1$ , where  $R$  is the reflectance and  $T$  the transmittance, does not hold, and furthermore,  $R$  and  $T$  lose their accepted meaning. We illustrate the perils of assuming energy flux additivity by calculating the transmission and reflection spectrum of a film over a substrate normally illuminated by incoherent light at frequencies in the neighborhood of an optical resonance. We also show that the usual relation between the scattering, absorption, and extinction cross sections for particles immersed within a dissipative host have to be modified to account for the non additivity.

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*OCIS codes:* 290.5850,310.6860,260.0260,240.0310,030.1640

## 1. Introduction

Fizeau fringes, Newton rings, and the colors of butterfly's wings, soap bubbles, duck's plumage, oil stains, etc., are familiar optical phenomena. They are due the interference between several outgoing waves which emerge from thin films after having transversed their widths several times, being subject to multiple reflections at their interfaces, and they illustrate the fact that the total outgoing energy flux is not simply the sum of the irradiances of each multiply reflected wave. It is well known that the Poynting vector is quadratic in the field amplitude, and therefore it is in general a non-additive quantity. However, there is a common situation in which additivity of the energy flux is usually assumed and where interference is seldomly discussed, namely, the reflection of light at an interface separating two media. When light impinges on a surface, it is partially reflected and partially transmitted. Energy conservation seems to imply that the power that is brought to the surface by the incident wave is balanced by the power that is taken away by the reflected and the transmitted waves. Thus,  $R + T + A = 1$  where  $R$  and  $T$  are the ratios of the reflected and transmitted power to the incident power per unit area of the interface and  $A$  corresponds to the power absorbed by the interface itself, which may be disregarded for sharp, infinitely thin interfaces, even if the bordering media are absorptive.<sup>1</sup> Hidden in this statement is the assumption that the Poynting vector at the interface may be divided into an incoming contribution which is a property of the incident wave alone, as if no reflected wave were present, and an outgoing contribution which corresponds to the reflected wave alone, as if there were no incident wave. The possible interference between the incident and reflected fields is therefore neglected in the calculation of the energy flux. In this paper we show that although this is frequently correct, it is not so in general. Whenever the medium where the incoming wave propagates is opaque, there is a contribution to the energy flux which arises from both the the incident and reflected fields and which cannot be ascribed to any one of them individually. A similar situation arises even within transparent materials whenever the kinematical conditions are such that the incident and reflected waves are evanescent.

The effect that interference between incident and reflected waves has on the electric and magnetic fields is very well known,<sup>2</sup> and has an historical significance, as it was used to establish that the electric and not the magnetic field is responsible for blackening photographic emulsions and for inducing fluorescence.<sup>3</sup> However, we have not found any textbook where its effect on *the energy flux* were discussed and it is only recently<sup>4</sup> that it has been

discussed in the literature within the context of the coupling of energy into a waveguide. In standard books of Optics<sup>5-8</sup> incident and reflected contributions to the energy density flux are considered separately. In a typical optical experiment, light is produced by a distant source and measured by a distant detector. Thus, it has to propagate through transparent media within which additivity holds. However, it does not hold for propagation through opaque films and assuming it does uncritically might lead to wrong and absurd results.

In this paper we discuss the conditions under which non-additivity manifests itself and some of its consequences. In section 2 we calculate the reflectance of an opaque film illuminated by quasi-monochromatic incoherent light in order to illustrate the errors in which we may incur by the nonchalant assumption of additivity. These errors and their origin are analyzed in detail in section 3 where we study the propagation of energy near the surface of an opaque medium illuminated by monochromatic waves. The case of quasi-monochromatic partially coherent light is analyzed in detail in section 4. In section 5 we discuss the extinction, absorption, and scattering cross section of particles immersed within dissipative media, and we show that the usual relation linking extinction to the sum of absorption plus scattering has to be modified to account for the non additivity of the energy flux. Finally, we devote section 6 to conclusions.

## 2. Reflectance of a film

As a simple example which yields unphysical results when non-additivity is ignored, we consider the optical response of a film of thickness  $d$  as illustrated in Fig. 1. The film is characterized by a dielectric function

$$\epsilon_2(\omega) = 1 + \frac{\omega_p^2}{\omega_T^2 - \omega^2 - i\omega\gamma}, \quad (1)$$

with a single resonance at frequency  $\omega_T$ , a dissipation factor  $\gamma$  and a strength  $\omega_p^2 = \omega_L^2 - \omega_T^2$ , where  $\omega_L$  is the root of  $\epsilon_2(\omega)$  when  $\gamma \rightarrow 0$ . It has a complex refraction index  $n_2(\omega) = \sqrt{\epsilon_2(\omega)}$

and lies over a transparent substrate with a constant real refraction index  $n_3$ . Consider a normally incident monochromatic wave of frequency  $\omega$  which illuminates the system from a transparent medium with a constant real refraction index  $n_1$ . The reflection amplitude  $r(\omega)$ , defined as the quotient between the incident and reflected fields at the front surface (1-2) of the film, may be calculated as a sum of multiple reflected waves,<sup>9</sup>

$$r = r_{12} + t_{12}r_{23}t_{21} \exp(2ik_2d) \sum_{\ell=1}^{\infty} [r_{21}r_{23} \exp(2ik_2d)]^{\ell-1}, \quad (2)$$

where

$$r_{ij} = \frac{n_i - n_j}{n_i + n_j} \quad (3)$$

and

$$t_{ij} = \frac{2n_i}{n_i + n_j} \quad (4)$$

are the Fresnel reflection and transmission amplitudes<sup>10</sup> corresponding to a single scattering of light that reaches the interface ( $i - j$ ) while moving from medium  $i$  towards the adjacent medium  $j$ , and  $\exp(ik_2d)$  contains the change in the phase ( $\exp(ik_2'd)$ ) and in the amplitude ( $\exp(-k_2''d)$ ) of the electromagnetic wave due to a single one-way transversal of the film, where  $k_2 = k_2' + ik_2''$  is the complex wavenumber,

$$k_2 = n_2 \frac{\omega}{c}. \quad (5)$$

The first term in the RHS of Eq. (2) accounts for the field reflected from the front surface of the film. The second term accounts for the field that is transmitted into the film, crosses the film to and fro  $\ell = 1 \dots \infty$  times, being repeatedly reflected at its back and front, finally being transmitted backwards into the first medium. The geometrical sum in Eq. (2) can be readily performed<sup>9</sup> and yields

$$r = r_{12} + \frac{t_{12}t_{21}r_{23} \exp(2ik_2d)}{1 - r_{21}r_{23} \exp(2ik_2d)}. \quad (6)$$

Similarly, the transmission amplitude  $t(\omega)$ , defined as the quotient between the transmitted field at the back surface (23) and the incident field at the front surface (12) of the film, is<sup>9</sup>

$$t = \frac{t_{12}t_{23} \exp(ik_2d)}{1 - r_{21}r_{23} \exp(i2k_2d)}. \quad (7)$$

From Eqs. (6) and (7) we obtain the reflectance  $R_M = |r|^2$  and transmittance  $T_M = (n_3/n_1)|t|^2$  of the film for monochromatic light, defined as the ratios of the reflected and transmitted energy fluxes, respectively, to the incident energy flux.

Fig. 2 illustrates the typical behavior of the reflectance of an opaque film as obtained from Eq. (6). Its width was chosen as  $d = \lambda_T/2$ , where  $\lambda_T = 2\pi c/\omega_T$  is the free space wavelength at resonance. The reflectance is relatively low below the resonance frequency  $\omega_T$  and after the longitudinal frequency  $\omega_L$ , while it is relatively high at resonance, where the dielectric function is very large, and within the stop gap, between  $\omega_T$  and  $\omega_L$ , where  $\epsilon_2$  is negative and  $n_2$  imaginary. Furthermore, the reflectance shows a series of oscillations due to the interference among the multiple reflected waves, which alternate between partially constructive to partially destructive as the frequency is varied. These oscillations become more closely spaced as the width of the film is increased.

We might expect that if the film is illuminated with incoherent instead of monochromatic light, the interference ought to be washed away due to the introduction of randomness in the phase. Assuming there is no fixed phase relation between the multiple reflected waves, we perform a random phase approximation and obtain the reflectance by adding incoherently the *intensities* of the multiply reflected waves, instead of adding their amplitudes. Thus, we write the reflectance  $R_I$  for incoherent illumination<sup>11</sup> as

$$R_I = R_{12} + T_{12}R_{23}T_{21} \exp(-4k_2''d) \sum_{\ell=1}^{\infty} [R_{21}R_{23} \exp(-4k_2''d)]^{\ell-1}, \quad (8)$$

in analogy to Eq. (2), where

$$R_{ij} = |r_{ij}|^2 = \frac{|n_i - n_j|^2}{|n_i + n_j|^2} \quad (9)$$

and

$$T_{ij} = (n'_j/n'_i)|t_{ij}|^2 = 4 \frac{n'_j (n'_i)^2 + (n''_i)^2}{n'_i |n_i + n_j|^2} \quad (10)$$

are the reflectance and transmittance of the  $ij$  interface, defined in analogy to  $r_{ij}$  and  $t_{ij}$  above but dividing wave intensities instead of amplitudes. Each term of the series (8) is the incoherent counterpart of the corresponding term in the series (2). There is a factor  $\exp(-2k''_2 d)$  corresponding to each traversal of the film to account for the intensity decay due to its opacity. Summing the ensuing geometrical series, we obtain

$$R_I = R_{12} + \frac{T_{12}T_{21}R_{23} \exp(-4k''_2 d)}{1 - R_{12}R_{23} \exp(-4k''_2 d)}. \quad (11)$$

Similarly, we calculate the incoherent transmittance<sup>11</sup>

$$T_I = \frac{T_{12}T_{23} \exp(-2k''_2 d)}{1 - R_{12}R_{23} \exp(-4k''_2 d)}. \quad (12)$$

These expressions for  $R_I$  and  $T_I$  are analogous to Eqs. (6) and (7) for  $r$  and  $t$ , and may be obtained by simply replacing  $r_{ij} \rightarrow R_{ij}$ ,  $t_{ij} \rightarrow T_{ij}$  and  $\exp(ik_2 d) \rightarrow \exp(-2k''_2 d)$ . Fig. 2 shows that  $R_I$  as given by Eq. (11) effectively removes the interference oscillations from  $R_M$  yielding the expected result, halfway between their maxima and minima. Thus, our procedure of adding intensities instead of amplitudes *seems reasonable* for incoherent illumination.

However, there are problems with Eq. (11) as becomes evident when thinner films are considered. In Fig. 3 we show the coherent and incoherent reflectance calculated for a similar but narrower film of width  $\lambda_T/10$ . Obviously, the incoherent reflectance displayed in Fig. 3 is wrong as it violates the principle of energy conservation, which requires that

for any passive media, the outgoing power should be smaller or equal than the incoming power, the difference being accounted for by the power absorbed within the film. As the absorptance is positive,  $R_I$  and  $T_I$  should obey

$$0 \leq R_I \leq 1, \quad 0 \leq T_I \leq 1, \quad 0 \leq R_I + T_I \leq 1. \quad (13)$$

Eqs. (11) and (12) violate these conditions within the stop band.

It could be correctly argued<sup>11</sup> that our naive derivation of Eqs. (11) and (12) is flawed, as it must be obvious that the interference between the  $n$ -th multiply reflected contribution to the reflected field (Eq. (2)) and the  $n + 1$ -th and  $n - 1$ -th contributions cannot be washed away by incoherence effects if the film is too thin. In particular, it is unreasonable to expect an incoherent calculation to yield the correct result for films much thinner than a wavelength. We silently and uncritically assumed that the incoherent field has a small bandwidth  $\Delta\omega \ll \omega$  so that the frequency,  $n_2$ ,  $R_{21}$ ,  $R_{23}$ , etc. are well defined quantities and we can plot the incoherent response as a function of  $\omega$ . However, we also assumed that  $\Delta\omega$  is large enough so that the band encompasses many, or at the very least, one full interference-originated oscillation, if coherent effects are to be washed away. It is impossible to satisfy both requirements simultaneously for very thin films. Thus, it might not be surprising that Eqs. (11) and (12) lead to wrong results.

Although pertinent, this criticism does not fully resolve the unphysical situation displayed in Fig. 3. On one hand, even for very wide films it is always possible to find sets of parameters for which  $R_I$ , as given by Eq. (11), leads to unphysical results. For example, Fig. 4 shows that  $R_I > 1$  within a narrow frequency range close to  $\omega_L$  for relatively wide ( $d \approx 10\lambda_T$ ) free-standing films ( $n_1 = n_3 = 1$ ) if the dissipation factor is small enough ( $\gamma \approx 10^{-8}\omega_T$ ). It also illustrates the strong dependence of the region where this unphysical behavior manifests itself on the width of the film and on its dissipation factor. On the other

hand, we could always conceive a hypothetical system in which instead of electromagnetic waves or photons, classical particles are reflected with probability  $R_{ij}$  and transmitted with probability  $T_{ij}$  whenever they reach the  $ij$ -th interface, and are absorbed with probability  $2k_2''$  per unit length as they travel across the film. *For this classical system, the total probability of being reflected or transmitted would be given exactly<sup>12</sup> by Eqs. (11) and (12).* Thus, if Eqs. (11) and (12) produce not only wrong, but also absurd results, *it must be because their ingredients, i.e.,  $R_{ij}$  and  $T_{ij}$  are themselves unphysical.* In particular, it can be easily shown that if  $R_{ij}$  and  $T_{ij}$  were constrained by the usual conditions  $0 \leq R_{ij} \leq 1$  and  $0 \leq T_{ij} \leq 1$ ,  $R_{ij} + T_{ij} = 1$ , and  $R_{ij} = R_{ji}$ , as would happen for transparent films, then  $R_I$  and  $T_I$  as given by Eqs. (11) and (12) would obey the expected constraints (13);  $0 \leq R_I, T_I, R_I + T_I \leq 1$  for transparent films, regardless of their thickness, even if they are so thin as to invalidate Eqs. (11) and (12).

That the optical coefficients of opaque media are not so constrained is demonstrated by Fig. 5, which shows the transmittance  $T_{21}$  of a single interface separating the opaque medium 2 from vacuum, corresponding to illumination from within the opaque medium, as calculated directly from Fresnel's coefficients (Eq. (10)). For frequencies in the stop gap,  $T_{21}$  may exceed unity by several orders of magnitude, confirming that *the usual interpretation of  $T_{21}$  as the fraction of the incident power that is transmitted, or as the probability that a photon is transmitted across the interface, is wrong when the incident medium is opaque.* We remark that  $T_{21} \neq T_{12}$  unless both media are transparent.

### 3. Energy flux within opaque systems

To understand the meaning of  $R_{21}$  and  $T_{21}$  we calculate the energy flux within the opaque film and close to its interface. We write the electric field for a monochromatic wave as the

real part of  $\mathbf{E}(z) \exp(-i\omega t)$ , with

$$\mathbf{E}(z) = \mathbf{E}_i \exp(-ik_2 z) + \mathbf{E}_r \exp(ik_2 z) \quad (14)$$

where  $\mathbf{E}_i$  is the amplitude of the incoming wave which propagates in the  $-z$  direction with complex wavenumber  $k_2$ . It is reflected at the (2-1) interface, situated for convenience at  $z = 0$ , giving rise to a reflected wave of amplitude  $\mathbf{E}_r = r_{21} \mathbf{E}_i$  propagating in the  $z$  direction, and C.C denotes the complex conjugate of the previous terms. The time averaged Poynting vector,  $\mathbf{S}(\mathbf{r}) = (0, 0, S_z(z))$  within the film is then

$$S_z(z) = -I_i(z) + I_r(z) + I_{\text{int}}(z), \quad (15)$$

where  $I_i(z) = I_i \exp(2k_2'' z)$  with  $I_i = (c/8\pi)n_2'|E_i|^2$  is the intensity of the incident wave, which decays towards the surface due to the medium's opacity, while  $I_r(z) = I_r \exp(-2k_2'' z)$ , with  $I_r = (c/8\pi)n_2'|E_r|^2 = R_{21}I_i$  and  $R_{21} = |r_{21}|^2$ , is the intensity of the reflected wave, which decays away from the surface. However, Eq. (15) contains an additional interference term,

$$I_{\text{int}}(z) = -2 \frac{n_2''}{n_2'} \text{Im}(r_{21} \exp(2ik_2' z)) I_i = -2 \frac{n_2''}{n_2'} [r_{21}' \sin(2k_2' z) + r_{21}'' \cos(2k_2' z)] I_i, \quad (16)$$

which oscillates with  $z$  and which does not decay in either direction. This term has contributions from both the incident and reflected waves and it originates in the products of the incoming electric and outgoing magnetic fields, as well as in the product of the incoming magnetic and the outgoing electric fields. As the Poynting vector is quadratic in the fields, this interference term, arising from the cross products of incoming and outgoing fields, could have been expected. Nevertheless, as it is proportional to the imaginary part of the index of refraction, it is not present within transparent media. It may be for this reason that this term is commonly disregarded in optics textbooks.

Notice that the term (16) is required to satisfy Poynting's theorem,  $\nabla \cdot \mathbf{S} = -\mathcal{P}$ , within

the dissipative medium, as the power  $\mathcal{P}$  dissipated per unit volume

$$\mathcal{P}(z) = \omega \frac{\epsilon_2''}{8\pi} |E(z)|^2 \quad (17)$$

contains an interference term

$$\mathcal{P}_{\text{int}}(z) = 2\mathcal{P}_i \text{Re} [r_{21} \exp(2ik_2'z)], \quad (18)$$

besides the power taken from the incident wave  $\mathcal{P}_i(z) = \mathcal{P}_i \exp(2k_2''z)$  and from the reflected wave  $\mathcal{P}_r(z) = \mathcal{P}_r \exp(-2k_2''z)$ , where  $\mathcal{P}_r = R_{21}\mathcal{P}_i$  and  $\mathcal{P}_i = (\omega\epsilon_2''/8\pi)|E_i|^2$ . Actually, it is precisely this interference term the one responsible for the interference fringes observed in Wiener's experiments, which established in 1890 that electromagnetic fields interacted with matter mainly through their electrical action.<sup>2</sup>

The Poynting vector corresponding to the wave transmitted to the transparent medium is given by

$$S_z(z) = -I_t = -\frac{c}{8\pi} n_1 |E_t|^2 = -T_{21} I_i. \quad (19)$$

Energy conservation requires  $S_z(z)$  to be continuous at the interface  $z = 0$ , which implies that

$$T_{21} = 1 - R_{21} + 2\frac{n_2''}{n_2'} r_{21}'', \quad (20)$$

as may be simply verified by substituting Eqs. (3), (9), and (10). Thus,  $R_{21} + T_{21} \neq 1$  within absorptive media. We remark that  $T_{21} \neq T_{12}$  as  $n_2$  is complex (Eq. (10)) and that indeed  $R_{12} + T_{12} = 1$  since  $n_1$  is real.

It can be simply shown that a similar situation holds for non-normal incidence and for both  $s$  and  $p$  polarizations and it is even present within transparent films whenever the kinematic conditions are such that the field within them is evanescent. A very illustrative case is that of the air or vacuum gap within two glass prisms in the geometry of frustrated

total internal reflection (FTIR).<sup>13</sup> Assuming the interfaces are parallel to the  $xy$  plane and that the system is illuminated by a plane wave with  $xz$  as the plane of incidence with an angle of incidence  $\theta$  larger than the critical angle  $\theta_c$ , the field that reaches the second prism after being transmitted from the first prism into the vacuum carries no energy whatsoever in the  $z$  direction; that is the origin of the phenomenon of total internal reflection. Similarly, the evanescent field reflected by the second prism carries no energy along  $\pm z$  by itself. Thus, the interference term between the incident and reflected wave is responsible for *all* of the energy transported across the gap and is responsible for the attenuation of the total reflection when both prisms are close to each other. As in the previous case, the amplitude of the interference term is independent of position across the gap. However, unlike the previous case,  $k_{\perp} = \sqrt{\omega^2/c^2 - k_{\parallel}^2} = i\kappa$  is purely imaginary, so that there are no oscillations such as those in Eq. (16). Here, we wrote the wave-vector of the evanescent waves in the gap as  $\mathbf{k}_{\pm} = (k_{\parallel}, 0, \pm k_{\perp})$  where  $k_{\parallel} = n(\omega/c) \sin \theta$  and  $n$  is the index of refraction of the prism. Within the vacuum gap  $S_z$  is constant, while  $S_x$  has three contributions: an exponentially decaying term due to the transmitted evanescent wave, a relatively small exponentially increasing term due to the evanescent wave reflected by the second prism and a constant interference term. Thus, the direction of  $\mathbf{S}$  depends on  $z$  and rotates towards the surface normal as we move from the first towards the second interface, where it reaches a finite angle  $\theta_g = \tan^{-1}(k_{\parallel}|t|^2/2\kappa t'')$ , where  $t$  is the transmission amplitude of the evanescent wave from vacuum into the glass. Curiously, for  $s$  polarization  $\theta_g = \theta$ .<sup>14</sup> As  $\theta_g \neq 0$ , our analysis above for a single plane wave is insufficient to account for the asymptotic value observed in the lateral displacement of the transmitted rays in FTIR systems illuminated by finite beams.<sup>14-18</sup>

Another example is the coupling of light into a waveguide from a nearby source localized

within a transparent ambient. It has been shown recently<sup>4</sup> that the evanescent part of the field produced by the source is responsible for all of the energy carried away by the waveguide, and that all of the injected energy is accounted for by a term similar to our Eq. (16), describing the interference between the field produced by the source and the field reflected by the waveguide. A final example is given by the propagation through an amplifying layer,<sup>19-21</sup> which far from saturation may be described by a complex index of refraction, as in an absorbing medium, but changing the sign of its imaginary part.

As in the above systems the energy flux cannot be analyzed solely into an incoming and an outgoing contribution, it is no longer necessary to balance the energy flux of the incident wave with those of the reflected and transmitted waves. More energy may reach the surface than that carried by the incident wave alone, and therefore,  $R$  and  $T$  are no longer bounded, i.e., it is possible that  $R > 1$  and  $T > 1$ , as exemplified for  $T_{21}$  by Fig. 5. Furthermore,  $R$  and  $T$  can no longer be interpreted as the probability of reflecting or transmitting a photon. Thus, our derivation of Eqs. (11) and (12) is not valid.

#### 4. Partially Coherent Light

We have shown that additivity of the energy flux does not hold within opaque media, so that  $R$  and  $T$  lose their usual meaning, invalidating our derivation of Eqs. (11) and (12). Nevertheless, these equations seem to yield reasonable results for films that are not too thin, as shown in Fig. 2. To explore their physical soundness, we study in more detail the optical properties of a surface when illuminated from within an opaque medium by partially coherent, quasi-monochromatic light. We model the incoming wave as the superposition of a random succession of independent identical finite pulses with random phases and centered at random times. As a given pulse is uncorrelated with the other pulses of the train, the time autocorrelation function  $c(\tau) = \langle E(t + \tau)E(t) \rangle = \beta c_1(\tau)$  of the partially coherent wave

is proportional to the autocorrelation  $c_1(\tau)$  of a single pulse. Here,  $\beta$  is the average number of pulses per unit time and we denote by  $\langle(\dots)\rangle$  the time and ensemble average of any quantity  $(\dots)$ . Correspondingly, the correlation time  $\tau_0$  is equal to the correlation time of a single pulse, which we identify with its duration. We identify its spatial extension with the longitudinal correlation length,

$$2L = v_g \tau_0, \quad (21)$$

where  $v_g = (dk'_2/d\omega)^{-1}$  is the group velocity of the pulse.

The interference contribution (16) to the Poynting vector for monochromatic waves oscillates as a function of  $z$  with a spatial period of a half a wavelength and with an amplitude which is independent of the distance to the interface. However, it is also an oscillatory function of  $\omega$  with oscillations that become faster as we move away from the interface. As a pulse has a finite bandwidth, we expect contributions with slightly different frequencies to cancel each other at large enough distances. We verify this by calculating the total energy  $\mathcal{E}(z) = \int dt S_z(z, t)$  that crosses a unit area situated at position  $z$  after a single pulse with field

$$\mathbf{E}(z, t) = \int_0^\infty d\omega / (2\pi) \mathbf{A}(\omega) [\exp(-ik_2 z) + r_{21} \exp(ik_2 z)] \exp(-i\omega t) + C.C. \quad (22)$$

goes through it on its way to the surface (2-1) and back after being reflected. This can be obtained by integrating Eq. (14) with a weight function  $|A(\omega)|^2$ .

For definitiveness we choose a Gaussian amplitude  $\mathbf{A}(\omega) = \mathbf{E}_0 G(\omega)$ , where

$$G^2(\omega) = \frac{1}{\sqrt{2\pi}\Delta\omega} \exp\{[-(\omega - \omega_0)^2 / 2\Delta\omega^2]\} \quad (23)$$

is a normalized Gaussian function centered at  $\omega_0$  and with a width  $\Delta\omega$  which we assume much smaller than  $\omega_0$ . We approximate

$$k_2(\omega) \approx k_2(\omega_0) + (\omega - \omega_0) \left. \frac{dk_2}{d\omega} \right|_{\omega_0}, \quad (24)$$

and assume that  $z$  is close enough to the interface,  $\Delta\omega \frac{dk''}{d\omega} z \ll 1$ , so that although the pulse is attenuated, its shape is almost undeformed. In this case we obtain<sup>14</sup>

$$\mathcal{E}(z) = -\mathcal{E}_i(z) + \mathcal{E}_r(z) + \mathcal{E}_{\text{int}}(z) \quad (25)$$

where  $\mathcal{E}_i(z) = \mathcal{E}_i \exp(2k''_2 z)$  is the energy of the incident pulse, which decays towards the surface due to the opacity,  $\mathcal{E}_r(z) = R_{21} \mathcal{E}_i \exp(-2k''_2 z)$  is the energy of the reflected pulse, which decays away from the surface, and

$$\mathcal{E}_{\text{int}} = -2 \frac{n''_2}{n'_2} \exp(-z^2/2L^2) [r'_{21} \sin(2k'_2 z) + r''_{21} \cos(2k'_2 z)] \mathcal{E}_i \quad (26)$$

is the interference term. Here, we introduced the total energy  $\mathcal{E}_i = cE_0^2 n'_2 / (4\pi^2)$  that would cross a unit area at  $z = 0$  if there were no reflected wave, we identified  $\tau_0$  with  $1/\Delta\omega$ , and we evaluate all frequency dependent terms at the central frequency  $\omega_0$ . We may recover the energy flux  $S_z(z)$  in the partially coherent case simply by multiplying by the pulse rate  $\beta$ ,  $S_z(z) = \beta \mathcal{E}(z)$ . We note that Eq. (25) is similar to Eq. (15). However, the interference term displays now an exponential decay with a characteristic length  $L$  equal to half of the correlation length of the incident pulse.

The exponential decay of the interference term in Eq. (26) is easily understood by noticing that the elapsed time between the passage through  $z$  of the incident pulse and the passage of the reflected pulse is about  $T_d \approx 2z/v_g$  as  $2z$  is the distance to the surface and back. Interference between the reflected and the incident wave is only possible if the leading edge of a reflected pulse arrives before the trailing edge of the incident pulse has completely passed through  $z$ , i.e., if the delay is smaller than the correlation time,  $T_d < \tau_0$  (Fig. 6). Thus, at large enough distances from the interface,  $z \geq L$ , there is no interference term, additivity holds, and we may separate the energy flow into an incident  $S_i(z) = S_i \exp(2k''_2 z)$  and a reflected  $S_r(z) = S_r \exp(-2k''_2 z)$  contribution. This allows us to introduce a pseudointerface

with a given finite width  $w \geq L$  and define its reflectance and transmittance as

$$\tilde{R}_{21} \equiv \frac{S_r(w)}{S_i(w)} = R_{21} \exp(-4k_2''w), \quad (27)$$

and

$$\tilde{T}_{21} \equiv \frac{S_t(0)}{S_i(w)} = T_{21} \exp(-2k_2''w). \quad (28)$$

As could be expected from the discussion above, it may be shown<sup>14</sup> that the optical coefficients of the pseudo-interface do obey the constraints  $0 \leq \tilde{R}_{21} \leq 1$ ,  $0 \leq \tilde{T}_{21} \leq 1$ , and  $\tilde{R}_{21} + \tilde{T}_{21} + \tilde{A}_{21} = 1$ , where the absorptance  $\tilde{A}_{21}$  of the pseudo-interface obeys  $0 \leq \tilde{A}_{21} \leq 1$ . Defining similar quantities for the interface (2-3) we can write a series analogous to (8), but replacing the width  $d$  of the film by the distance  $\tilde{d} = d - 2w$  between pseudointerfaces, yielding

$$R_I = R_{12} + \frac{\tilde{T}_{12}\tilde{T}_{21}\tilde{R}_{23} \exp(-4k_2''\tilde{d})}{1 - \tilde{R}_{12}\tilde{R}_{23} \exp(-4k_2''\tilde{d})} \quad (29)$$

and

$$T_I = \frac{\tilde{T}_{12}\tilde{T}_{23} \exp(-2k_2''\tilde{d})}{1 - \tilde{R}_{12}\tilde{R}_{23} \exp(-4k_2''\tilde{d})}. \quad (30)$$

It is easy to show that Eqs. (29) and (30) are exactly equivalent to (11) and (12), as if  $R_{21}$ ,  $T_{21}$ , etc. had their simple-minded meaning. However, our derivation is only valid for films that are wide enough,

$$d \geq 2L, \quad (31)$$

so that the two pseudo-interfaces are non-overlapping and  $\tilde{d} \geq 0$ . Thus, the optical properties of an opaque film calculated as in Sec. 2 for the case of completely incoherent illumination are correct for the more realistic case of partially coherent light with a finite bandwidth, *provided that the film is wider than the longitudinal correlation distance*. The conditions  $d \geq 2L = v_g/\Delta\omega$  and  $\Delta\omega \ll \omega_0$  can only be satisfied simultaneously in films such that  $d \gg v_g/\omega_0$ . Thus, the case of completely incoherent but quasi-monochromatic illumination

is not realizable for very thin films. In this situation, the optical properties of the film have to be calculated<sup>14</sup> by averaging the full mono-chromatic result (6) and (7) with the spectral weight of the field and we can not short-cut this procedure as in Sec. 2.

We remark that besides the interference discussed above between incident and reflected waves within opaque media, there are additional well known effects due to interference between multiply reflected waves, and these are present even for transparent films whenever the delay  $T'_d \approx 2d/v_g$  between the  $n$ -th and the  $n + 1$ -th multiply reflected pulses is smaller than their correlation time,  $T'_d < \tau_0$ . Therefore, it would be absent for films wider than half a longitudinal correlation length  $d \geq L$ . Notice that this condition is similar but slightly less restrictive than the condition (31) pertaining to opaque films.

## 5. Extinction within dissipative media

In the previous sections we showed that the interference of the incident and reflected waves close to an interface yields a finite contribution to the energy flow within opaque films. Similar contributions are to be expected when an object placed within an absorbing medium is illuminated: the interference of the incoming wave with the scattered waves might yield an additional contribution to the energy flow, which should have to be properly accounted for in order to understand the scattering, absorption and extinction cross sections of the object.

The problem of scattering, absorption and extinction by particles in media with absorption was addressed many years ago.<sup>22-24</sup> When the media is non absorbing, it is well known that the extinction is due to the energy removed by absorption within the particle and by scattering. The *optical theorem* shows that the extinction may be simply obtained from the forward scattering amplitude, as only in the immediate neighborhood of the forward direction does interference between incident and scattered fields produce a finite contribution to

the energy flow. The main questions that have to be addressed for the case of particles embedded within absorbing media are the appropriate definition of the extinction and absorption cross sections, their calculation, and the validity or the correct generalization of the optical theorem. One problem when defining extinction and absorption coefficients within dissipative media is that the host contributes to both processes; there is a contribution from the far fields as well as from the near fields, and there would also be a contribution in the absence of the scatterer. Some definitions have been proposed<sup>22,23</sup> in which the absorption and scattering cross sections are identified by integrating the energy flux through conceptual large integration surfaces that surround the particle and for which the far field approximation may be employed. However, the resulting coefficients depend on the rather arbitrary size of the surface and are therefore not an intrinsic property of the scatterers. By calculating numerically the near fields for the special case of spherical particles, the cross sections have also been defined and calculated in terms of the energy flux through the actual boundary of the particle.<sup>25,26</sup> The various definitions do not agree among themselves within dissipative hosts. In the following, we present an alternative approach to extinction, absorption and scattering cross sections for a particle embedded in an absorbing medium. First we will use two special simple cases in order to illustrate our main ideas. Afterwards, we will generalize our results to the case of particles with arbitrary shape.

#### A. *Thin film*

Consider a system similar to that depicted in Fig 1, consisting of a thin film with an arbitrary dielectric function  $\epsilon_2$  and index of refraction  $n_2 = \sqrt{\epsilon_2}$  situated within an absorbing medium with a complex index of refraction  $n_1 = n_3 = n = n' + in'' = \sqrt{\epsilon}$ . The film plays the role of a scatterer located at the origin and for simplicity we have considered a planar geometry and we further assume that the width  $d$  is much smaller than the wavelength,  $d \ll \lambda$ , and

perform the calculations below to linear order in  $d$ . In the next subsections we will relax this assumptions. When normally illuminated by an incident field  $\mathbf{E}_i(z) = \mathbf{E}_i \exp(ikz)$ , with  $k = n\omega/c$ , an electric current  $\mathbf{j} = \mathbf{j}_0 + \Delta\mathbf{j}$  is established, where  $\mathbf{j}_0 = -i\omega(\epsilon - 1)\mathbf{E}_i/4\pi$  is the current density in the host and

$$\Delta\mathbf{j} = -i\omega\frac{\epsilon_2 - \epsilon}{4\pi}\mathbf{E}_i \quad (32)$$

is the change in the current density due to the presence of the scatterer. Acting on this extra current with the Green's operator  $\hat{G}$  of the host, represented by the Green's function<sup>27</sup>

$$G(z, z') = i\frac{\exp(ik|z - z'|)}{2k}, \quad (33)$$

we obtain the scattered potentials  $\mathbf{A}_\pm = 4\pi\hat{G}\Delta\mathbf{j}/c$  and fields

$$\mathbf{E}_\pm(z) = i\frac{\omega}{c}\mathbf{A}_\pm(z) = s_\pm\mathbf{E}_i \exp(\pm ikz), \quad (34)$$

where we introduced the *scattering amplitudes*

$$s_+ = s_- = i\frac{\omega d}{2nc}(\epsilon_2 - \epsilon) \quad (35)$$

in both the forward (+) and backward (-) directions, to lowest order in  $d/\lambda$  in the long wavelength approximation. As the scattered fields are linear in  $d$ , the scattered intensity  $I_s = I_+ + I_- \approx 0$  up to linear order in  $d$ . The extra power absorbed per unit area by the film may be obtained by integrating  $\text{Re}\Delta\mathbf{j} \cdot \mathbf{E}_i^*$ ,

$$I_a \approx \omega d \frac{\epsilon_2'' - \epsilon''}{8\pi} |E_i|^2 = \frac{\omega d}{n'c} (\epsilon_2'' - \epsilon'') I_i, \quad (36)$$

where  $I_i(z) = (c/8\pi)n'|E_i|^2 \exp(-2k''z)$  is the incident intensity at  $z$  and  $I_i \equiv I_i(0)$ . The total transmitted intensity is

$$I_t(z) = \frac{c}{8\pi} n' |E_i(z) + E_+(z)|^2 \approx (1 + 2s'_+) I_i \exp(-2k''z) = I_i(z) - I_e(z), \quad (z > 0) \quad (37)$$

from which we can identify the power

$$I_e(z) = -2s'_+ I_i \exp(-2k''z), \quad (38)$$

extinguished from the incoming wave per unit area. One might then define scattering

$$\sigma_s/\mathcal{A} = 0, \quad (39)$$

absorption

$$\sigma_a/\mathcal{A} = \frac{I_a}{I_i} = \frac{\omega d}{n'c}(\epsilon_2'' - \epsilon''), \quad (40)$$

and extinction

$$\sigma_e/\mathcal{A} = I_e(0)/I_i = -2s'_+ \quad (41)$$

coefficients per unit area  $\mathcal{A}$ . Eq. (41) is the 1D formulation of the optical theorem.<sup>28</sup>

However, the naive expectation that extinction is accounted for by scattering and absorption,

$$\sigma_e = \sigma_s + \sigma_a \quad (\text{wrong}) \quad (42)$$

does not hold whenever the host is opaque, as

$$(\sigma_e - \sigma_s - \sigma_a)/\mathcal{A} = \frac{\omega}{c}d \left[ \text{Im} \left( \frac{\epsilon_2 - \epsilon}{n} \right) - \frac{\epsilon_2'' - \epsilon''}{n'} \right] \neq 0, \quad (43)$$

unless  $n'' = 0$ . The reason for this discrepancy with the usual scattering result within transparent media is that the total incoming intensity

$$\begin{aligned} I_t(z) &= \frac{c}{8\pi} n' |E_i(z) + E_-(z)|^2 \approx \left\{ \exp(-2k''z) + 2\frac{n''}{n'} \text{Im}[s_- \exp(-2ik'z)] \right\} I_i \\ &= I_i(z) - I_{\text{int}}(z), \quad (z < 0) \end{aligned} \quad (44)$$

has an additional oscillating contribution

$$I_{\text{int}}(z) = -2\frac{n''}{n'} \text{Im}(s_- \exp(-2ik'z)) I_i \quad (45)$$

to the energy flow, due to the interference between the backscattered wave and the incident wave. Defining an *interference cross section*

$$\sigma_{\text{int}}/\mathcal{A} = I_{\text{int}}(0)/I_i = -2\frac{n''}{n'}s_-'' , \quad (46)$$

we may account for the additional contribution to the incoming energy flow, by modifying Eq. (42),

$$\sigma_e = \sigma_a + \sigma_s + \sigma_{\text{int}} . \quad (47)$$

Eq. (47) is simply verified by using Eqs. (39), (40), (41), (46), and (35).

### B. Arbitrary film

We can generalize the results above to films of an arbitrary width taking care that not only the scatterer but also the host absorb energy from the incident wave. We identify the transmitted wave  $t \exp[ik(z-d)]E_i$  with the sum of the incident plus forward-scattered wave, so that

$$s_+ = t \exp(-ikd) - 1, \quad (48)$$

where  $t$  is given by Eq. (7). The backward scattered wave is simply the reflected wave

$$s_- = r, \quad (49)$$

where  $r$  is given by Eq. (6). The scattered intensity is therefore

$$I_s(z) = \begin{cases} I_{s+}(z) = |s_+|^2 \exp(-2k''z)I_i, & (z \geq d) \\ I_{s-}(z) = |s_-|^2 \exp(2k''z)I_i. & (z \leq 0) \end{cases} \quad (50)$$

The power absorbed per unit area  $I_a^0(z_-, z_+) + I_a(z_-, z_+)$  within a volume delimited by two positions  $z_- < 0$  and  $z_+ > d$  is simply obtained by applying Poynting's theorem,

$$I_a(z_-, z_+) = -\{|s_-|^2 \exp(2k''z_-) - 2\frac{n''}{n'}\text{Im}[s_- \exp(-2ik'z_-)] + (|s_+|^2 + 2s_+'') \exp(-2k''z_+)\}I_i, \quad (51)$$

where  $I_a^0(z_-, z_+) = [\exp(-2k''z_-) - \exp(-2k''z_+)]I_i$  corresponds to the energy that would be absorbed in the absence of the film. The transmitted intensity may be written as

$$I_t(z) = I_i(z) + I_{s+}(z) - I_e(z), \quad (z > d) \quad (52)$$

where we identify the extinction intensity

$$I_e(z) = -2s'_+ \exp(-2k''z)I_i. \quad (z > d) \quad (53)$$

Extrapolating the results above to  $z, z_-, z_+ \rightarrow 0$ , we may define a scattering cross section

$$\sigma_s/\mathcal{A} = [I_{s-}(0) + I_{s+}(0)]/I_i = |s_+|^2 + |s_-|^2, \quad (54)$$

an absorption cross section

$$\sigma_a/\mathcal{A} = I_a(0,0)/I_i = -(|s_-|^2 + |s_+|^2) + 2\frac{n''}{n'}s''_- - 2s'_+, \quad (55)$$

and an extinction cross section

$$\sigma_e/\mathcal{A} = I_e(0)/I_i = -2s'_+. \quad (56)$$

As in the previous case, Eq. (42) is violated and the extinction, scattering, and absorption coefficients are related by Eq. (47) ( $\sigma_e = \sigma_a + \sigma_s + \sigma_{\text{int}}$ ), where the interference intensity and cross section are given anew by Eqs. (45) and (46), the only difference being the values (48), (49) of the forward ( $s_+$ ) and backward ( $s_-$ ) scattering matrix elements, given in terms of  $r$  (6) and  $t$  (7), and the finite scattering cross section (54).

### C. Particles

We can further generalize the results above to the case of scattering by arbitrary particles within opaque media. In this case there is a semantic problem related to the definition of absorption, scattering, and extinction cross sections, as both the particle and the host may

absorb energy. Thus there is energy absorption even in the absence of the particle, and the intensity of the scattered waves is dissipated by the host. Furthermore, the particle induces near fields which excite additional currents within the host, producing additional energy dissipation within the neighborhood of the particle, but on its outside. This near field contribution is absent in the cases of planar geometries discussed previously. Thus, several definitions of absorption and extinction have been proposed which, unlike the case within transparent hosts, are not equivalent to each other.<sup>22–26</sup>

Consider a small particle within a weakly absorbing host and illuminated by a linearly polarized monochromatic wave. We center the particle at the origin of the coordinate system and orient its axes so that the incoming wave is polarized along the  $x$  direction and propagates along the  $z$  direction,

$$\mathbf{E}_i(\mathbf{r}) = E_i \hat{x} \exp(ikz). \quad (57)$$

The scattered field far from the particle is given by

$$\mathbf{E}_s(\mathbf{r}) = i\mathbf{s}(\Omega) E_i \frac{\exp(ikr)}{kr}, \quad (58)$$

which defines the scattering amplitude  $\mathbf{s}(\Omega)$  in the direction  $\hat{\Omega} = \mathbf{r}/r$ . The corresponding magnetic fields may be obtained from Eqs. (57) and (58) through Faraday's law. The Poynting vector  $\mathbf{S} = \mathbf{S}_i + \mathbf{S}_s + \mathbf{S}_{\text{int}}$  has a contribution  $\mathbf{S}_i$  from the incident field, a contribution  $\mathbf{S}_s$  from the scattered fields, and a contribution  $\mathbf{S}_{\text{int}}$  due to their interference with the incident field. As the latter is proportional to  $\exp[i(kr - k^*z)] = \exp\{i[k - k^* \cos(\theta)]r\}$ , it is a very rapidly oscillating function of the polar angle  $\theta$ , except in the neighborhood of the stationary phase values  $\theta = 0, \pi$ . Thus, it does not contribute to the energy flow across a finite area detector (i.e., with a transverse size  $> \sqrt{r\lambda}$  or equivalently, an angular aperture  $> \sqrt{\lambda/r}$ ), unless it is situated along the forward or backward directions. The intensity close to  $\theta = 0$

is therefore

$$I(\mathbf{r}) = S_z(\mathbf{r}) = \left\{ 1 - 2\text{Im} \left[ s_x(0) \frac{\exp(ik^*r\theta^2/2)}{kr} \right] + \frac{|s(0)|^2}{|k|^2 r^2} \right\} I_i \exp(-2k''r), \quad (59)$$

which integrated within the aperture of a detector yields the detected power

$$\mathcal{P}_d(r) = \left[ \mathcal{A}_d - \frac{4\pi}{|k|^2} s'_x(0) \right] I_i \exp(-2k''r), \quad (60)$$

where  $\mathcal{A}_d = r^2 \int d\Omega f(\theta)$  is the effective collection area of the detector whose sensitivity  $f(\theta)$  is a function that interpolates smoothly from  $f(0) = 1$  at its center to  $f(\theta \gg \sqrt{\lambda/r}) = 0$  at its edge. To obtain Eq. (60) we assumed that the solid angular aperture  $\Omega_d = \mathcal{A}_d/r^2$  of the detector is very small, so that we may neglect the power  $|S(0)|^2 I_i \Omega_d / |k|^2$  scattered into it (i.e., the integral of the third term of Eq. (59)). Furthermore, we assumed that the collection surface is spherical and centered on the particle, so that  $r$  remains constant throughout the integration. Eq. (60) may be interpreted as a removal of all the energy that falls on an obstacle situated at the origin with a transverse area given by the *extinction* cross section

$$\sigma_e = \frac{4\pi}{|k|^2} s'_x(0), \quad (61)$$

followed by propagation for a distance  $r$  within the opaque medium, where additional energy is removed, yielding the extinction power

$$\mathcal{P}_e(r) = \frac{4\pi}{|k|^2} s'_x(0) I_i \exp(-2k''r). \quad (62)$$

Eq. (61) is a statement of the optical theorem<sup>24</sup> within opaque media, the only difference with the transparent case being the replacement of the wavenumber  $k$  by its magnitude  $|k|$ . We remark that integrating Eq. (59) over a flat surface, with constant  $z$  instead of a spherical surface with constant  $r$  we would have obtained a different expression,

$$\sigma_e(z) = 4\pi \text{Re} \left[ \frac{s_x(0)}{k^2} \right], \quad (\text{flat detector}) \quad (63)$$

for the extinction cross section.<sup>24</sup>

Similarly, we calculate the incoming intensity

$$I(\mathbf{r}) = S_z(\mathbf{r}) = \left\{ \exp(2k''r) + 2\frac{n''}{n'} \operatorname{Re} \left[ s_x(\pi) \frac{\exp(2ik'r) \exp(-ik^*r\xi^2/2)}{kr} \right] - \frac{|s(\pi)|^2}{|k|^2 r^2} \exp(-2k''r) \right\} I_i \quad (64)$$

with  $\mathbf{r}$  close to the backwards direction  $\theta = \pi - \xi \approx \pi$ . The first and the last terms in Eq. (64) correspond to the intensity of the incident wave and the intensity scattered along the backwards direction, while the middle term is an oscillating function of  $r$  analogous to that in Eqs. (45), due to the interference between the incident and the backscattered wave. Due to its rapid oscillations as a function of  $\xi$ , it only contributes to the energy flux close to  $\xi = 0$ , where it yields the outgoing power

$$\mathcal{P}_{\text{int}}(r) = -\frac{4\pi}{|k|^2} \frac{n''}{n'} \operatorname{Im} [s_x(\pi) \exp(2ik'r)] I_i. \quad (65)$$

Extrapolating to  $r \rightarrow 0$ , we define an interference cross section

$$\sigma_{\text{int}} \equiv \mathcal{P}_{\text{int}}(r \rightarrow 0)/I_i = -\frac{4\pi}{|k|^2} \frac{n''}{n'} s_x''(\pi). \quad (66)$$

The total power  $\mathcal{P}_a^0(r) + \mathcal{P}_a(r)$  absorbed within a sphere of radius  $r$  may be obtained through Poynting's theorem. Thus, it has a contribution  $\mathcal{P}_a^0(r) = \pi I_i (2k''r \cosh 2k''r - \sinh 2k''r)/(k'')^2$  corresponding to the power that would have been dissipated by the host in the absence of the scatterer. The other contributions  $\mathcal{P}_a = -\mathcal{P}_s + \mathcal{P}_e - \mathcal{P}_{\text{int}}$  include the scattered outflowing energy

$$\mathcal{P}_s(r) = \int d\Omega \frac{|s(\Omega)|^2}{|k|^2} \exp(-2k''r), \quad (67)$$

integrated over all the solid angle  $\Omega$ . Finally, there are contributions  $\mathcal{P}_e(r)$  and  $\mathcal{P}_{\text{int}}(r)$  (Eqs. (62) and (65)) due to the interference between incident and scattered fields along the forward

and backwards directions. Finally, extrapolating Eq. (67) towards  $r \rightarrow 0$  we get rid of the far field contributions to the energy absorbed by the host and arrive finally at Eq. (47)

( $\sigma_e = \sigma_a + \sigma_s + \sigma_{\text{int}}$ ), where we define the scattering cross section

$$\sigma_s \equiv \mathcal{P}_s(r \rightarrow 0)/I_i = \int d\Omega \frac{|s(\Omega)|^2}{|k|^2}, \quad (68)$$

and the absorption cross section

$$\sigma_a \equiv \mathcal{P}_a(r \rightarrow 0)/I_i \quad (69)$$

In summary, when a particle within an absorbing medium is illuminated, its extinction cross section, defined through its contribution to the removal of energy from the incoming wave, is not accounted for by the scattered and absorbed power, as in the case of transparent hosts. There is an additional contribution due to the interference between the counterpropagating incident and backscattered waves.

## 6. Conclusions

We have shown that the energy flux is not an additive quantity when the surface of an opaque medium is illuminated from within with an electromagnetic wave, as there is a spatially oscillating contribution to the Poynting vector whose amplitude does not decay away from the surface and which arises from the interference between the incoming and outgoing waves. In this situation, the reflectance and transmittance cannot be interpreted as the fraction of the incoming power that is reflected or transmitted, nor as the probability for an incoming photon to be reflected or transmitted. Thus, theoretical calculations which depend on these interpretations are bound to fail. We illustrated the dangers of an uncritical use of the concepts of reflectance and transmittance through the calculation of the optical properties of thin opaque supported films illuminated by incoherent quasi-monochromatic light, obtaining

unphysical results such as a reflectance that is larger than unity. A careful analysis for the case of partially coherent illumination showed that the interference contribution to the energy flux does decay away from the surface in this case and becomes negligible at distances of the order of half a longitudinal correlation distance. Thus, the simple minded approach assuming additivity might yield the correct results, but only for films that are wider than the correlation length  $d > 2L$ . This condition is more restrictive than that applicable to transparent media. For films that are too thin, it may prove impossible to satisfy the condition of quasi-monochromaticity  $\Delta\omega \ll \omega$  together with the condition of a relatively small correlation length  $2L < d$ , so a calculation of their optical properties would have to account in detail for the partial coherence.<sup>14</sup> Similar considerations apply to evanescent waves within transparent systems, as in the FTIR geometry, and for waves within media with gain, where the amplitude increases instead of decreasing exponentially as each wave propagates.

We analyzed the energy flow for the problem of scattering by particles immersed within dissipative systems, for both planar and for arbitrary geometries. We have proposed definitions of the absorption, scattering, and extinction cross sections in such a way that they are an intrinsic property of the particles although their calculation requires only a knowledge of the asymptotic scattered far field. We have shown that the usual relation between absorption, scattering, and extinction cross sections does not hold within dissipative media, as there is a finite contribution to the energy flow due to the interference between the incident and the backscattered waves. However, by introducing a new *interference* cross section, we obtained a generalization:  $\sigma_e = \sigma_a + \sigma_s + \sigma_{\text{int}}$ .

## Acknowledgments

We acknowledge useful discussions with H. Larralde, R. Barrera, E. R. Méndez and M. A. Alonso. GPO is grateful for the advice received from E. Vargas. This work was supported by Dirección General de Apoyo Académico - Universidad Nacional Autónoma de México (grant 117402) and Centro de Investigación en Polímeros-Comercial Mexicana de Pinturas, Sociedad Anónima de Capital Variable.

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## 7. Figure Captions

1. Film of thickness  $d$  on which monochromatic light is normally incident from the left. There are two interfaces between the three media (vacuum (1), film (2), and substrate (3)), with refraction indexes ( $n_i$ ). The wave vectors ( $\pm k_i$ ,  $i = 1 \dots 3$ ) for waves traveling towards the right and left within each media are shown. The coordinate system is also indicated.
2. Reflectance  $R_M$  (solid) calculated for a film normally illuminated with monochromatic light of frequency  $\omega$  and its incoherent counterpart  $R_I$  (dashed) as a function of frequency  $\omega$ . Light is incident from vacuum ( $n_1 = 1$ ) and the film lies on a substrate with  $n_3 = 2$ . The thickness of the film is  $d = \lambda_T/2$ , and it has a dissipation factor  $\gamma = 0.01\omega_T$ , and effective weight  $\omega_p^2 = \omega_T^2$ . The longitudinal and transverse frequencies  $\omega_T$  and  $\omega_L$  are indicated.
3. Reflectance  $R_M$  (solid) calculated for a film as in Fig. 2 but five times thinner, of width  $d = \lambda_T/10$ , normally illuminated by monochromatic light of frequency  $\omega$ , and its incoherent counterpart  $R_I$  (dashed).
4. Incoherent reflectance  $R_I$  of free-standing films ( $n_1 = n_3 = 1$ ) of widths  $d = 10\lambda_p, 11\lambda_p$ , dissipation factors  $\gamma = 10^{-8}, 2 \times 10^{-8}$ , and transition weight  $\omega_p^2 = \omega_T^2$ .
5. Transmittance  $T_{21}$  of a single interface 2 – 1 for a wave incident from a medium with dielectric response given by Eq. (1) on the interface that separates it from vacuum, as a function of frequency. We took  $\omega_p^2 = \omega_T^2$  and  $\gamma = 0.01\omega_T$ , as in Figs. 2 and 3.
6. Pulse of length  $2L = v_g\tau_0$  incident (solid) and reflected (dashed) from an interface (wide solid line) at different times increasing from the bottom towards the top. Beyond

the pseudointerface of width  $w \geq L$  (gray) additivity holds, as there is no overlap and therefore no interference between the incident and the reflected pulses. The directions of motion are indicated (arrows).

## 8. Figures

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Fig.1











