

Subwavelength Multilayer Dielectrics: Ultrasensitive Transmission and Breakdown of Effective-Medium Theory

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We show that a purely dielectric structure made of alternating layers of deep subwavelength thicknesses exhibits novel transmission effects which completely contradict conventional effective medium theories exactly in the regime in which those theories are commonly used. We study waves incident at the vicinity of the effective medium's critical angle for total internal reflection and show that the transmission through the multilayer structure depends strongly on nanoscale variations even at layer thicknesses smaller than $\lambda/50$. In such deep subwavelength structures, we demonstrate dramatic changes in the transmission for variations in properties such as periodicity, order of the layers, and their parity. In addition to its conceptual importance, such sensitivity has important potential applications in sensing and switching.

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In electromagnetism, homogenization is the conceptual process of replacing a complex structure of subwavelength sized components with an “effective medium” with uniform properties. It is a fundamentally important notion which can be traced back to the earliest days of electromagnetic theory, to the Lorentz-Lorenz and Maxwell-Garnet effective medium models [1–3]. More modern approaches to homogenization calculate the effective parameters in greater precision and for a variety of geometries, either analytically or by extraction from a numerical or physical experiment [4]. Importantly, the effective medium parameters can be very different from those ordinarily found in nature. The effective medium of properly designed metamaterials can exhibit a huge birefringence [5–7], a negative refractive index [8–12], or a near-zero electric permittivity [13,14].

While effective medium theories have become ubiquitous, the homogenization approach becomes particularly intricate and fails altogether in periodic metal-dielectric structures involving wave vectors significantly larger than the vacuum wave vector [15,16], especially in the presence of gain [17] or involving strong interaction with surface waves [18–22]. In such cases, the transmission of metal-dielectric multilayer systems can be sensitive to details (e.g., the permittivity of a single layer) which are typically averaged out in the effective medium description [22]. On the other hand, all-dielectric systems, which fundamentally do not support extremely large wave vectors or surface wave resonances, are believed to tightly obey homogenization. Consider the case of a multilayer structure composed of layers with permittivity ϵ_a, ϵ_b and thicknesses $d_a, d_b \ll \lambda$, where λ is the vacuum wavelength [23]. Naturally, we assume that all layer thicknesses are much larger than the molecular dimensions (or lattice constant in crystalline media). For this case, the basic predictions of the homogenization approach are as follows.

(1) When $d_{a,b} \rightarrow 0$, the permittivity of the entire multilayer layer approaches the effective medium value, which under TE illumination is the simple average $\bar{\epsilon} = ((\epsilon_a d_a + \epsilon_b d_b)/(d_a + d_b))$ [23]. As a consequence, the subwavelength structural features are smoothed out and it is hard to tell apart the two similar structures that differ only on a subwavelength scale [3].

(2) The behavior of the structure is independent of the number of layers and their order. Only the total size of the structure matters. Adding 1–2 additional layers should, therefore, not have any noticeable effect on the transmission.

(3) The effective permittivity and permeability does not depend on nearby materials that border the multilayer structure. One can analyze the transmission of (and reflection from) the multilayer structure by consistently replacing it with an effective medium (which can be isotropic or anisotropic), regardless of what materials border the structure.

Here, we examine a simple stratified dielectric structure that defies all these basic predictions of effective medium theory and displays boundary effects that cannot be understood from the conventional effective medium perspective. Specifically, we show that even when the layers are 0.02 λ thin, impedance matching with the exterior medium [from which the electromagnetic (EM) waves enter or leave the multilayer structure] is not always possible. The phase of the observed reflection depends on the properties of the last layer, and as a result, the transmission of the entire structure can change significantly (of order unity) due to the addition of a single 10 nm thin layer or when the order of the layers is reversed. In addition, we examine the extreme sensitivity of this system to nanometric variations in layer thickness and discuss its potential applications in sensing and switching. Finally, we consider the nature of propagation and power conservation in such multilayers and show that it

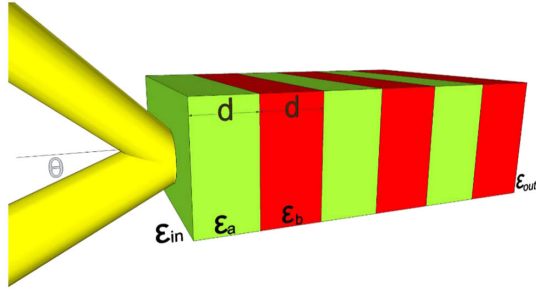


FIG. 1 (color online). Schematics of a multilayer structure ($N = 3$ periods), surrounded by homogeneous materials with permittivities ϵ_{in} and ϵ_{out} . A plane wave is incident at angle θ , at the proximity of the critical angle for total internal reflection.

is fundamentally different from the effective model prediction, even in the $d/\lambda \rightarrow 0$ limit.

Consider the dielectric multilayer stack displayed in Fig. 1, made of N pairs of dielectric layers with a permittivity of ϵ_a or ϵ_b and a thickness of d_a or d_b , respectively. The structure is surrounded by homogeneous mediums $\epsilon_{\text{in,out}}$. Assuming $\epsilon_{\text{in}} > \bar{\epsilon}$, we can define the angle $\theta_c = \arcsin(\sqrt{\bar{\epsilon}/\epsilon_{\text{in}}})$, which is the critical angle for total internal reflection for a wave incident from ϵ_{in} into $\bar{\epsilon}$.

The effects described in this Letter are reasonably robust—they persist over an angular range of $\sim 5\%$ below θ_c , assuming the difference between ϵ_a, ϵ_b is order unity. Observation of the effects should be possible over a reasonably broad bandwidth, and appear for both TE and TM polarization. However, in order to simplify the presentation, we restrict the discussion below to the case where the effects are most easily observed: illumination by a TE-polarized CW wave, incident at precisely $\theta = \theta_c$, with a beam wide enough to justify its approximation to a plane wave. Under these conditions, the wave is an evanescent wave in the lower permittivity (ϵ_b) layer and a propagating wave in the higher permittivity (ϵ_a) layer, and

the magnitude of the wave vector in both layers is equal: $k_{z,a} = ik_{z,b}$, where $k_{z,j} = \sqrt{\epsilon_j k_0^2 - k_x^2}$ for $j = a, b$ and $k_0 = 2\pi/\lambda$.

As a concrete example, we take $\lambda = 500$ nm, $\epsilon_a = 5, \epsilon_b = 1$, and $d_a = d_b = d = 10$ nm (i.e., 0.02λ), which is deeply subwavelength, and consider the case where $\epsilon_{\text{in}} = \epsilon_{\text{out}} = 4$. We also assume constant layer thicknesses (no disorder), but the main results remain valid and observable even in the presence of weak disorder ($< 5\%$ width variation). The results presented in this Letter are calculated using the transfer-matrix formalism, as described in detail in Ref. [24]. Importantly, we emphasize that this formalism is exact, accounting for all the forward and backward reflections in the structure [28]. To obtain the effective permittivity ϵ_{eff} for such a structure, we consider the dispersion relation of a multilayer structure [29] and assume deeply subwavelength layers (we neglect a term which is small when $k_z d \ll 1$) to get

$$\cos[k_{z,\text{eff}}(d_a + d_b)] = \cos(k_{z,a}d_a) \cosh(ik_{z,b}d_b). \quad (1)$$

From this relation we can extract $\epsilon_{\text{eff}} = \bar{\epsilon} + \Delta(d)$, where Δ is a small correction term (on the order of 5×10^{-3} for our choice of parameters), and almost independent of θ . At normal incidence, this correction term is practically negligible and one can safely approximate $\epsilon_{\text{eff}} = \bar{\epsilon}$, as expected from the effective medium approach. However, for incidence near the critical angle, the z component of the wave vector is $k_{z,\text{eff}} = \sqrt{(\bar{\epsilon} + \Delta)k_0^2 - k_x^2} = k_0\sqrt{\Delta}$ and the correction term Δ becomes crucial. We calculate the transmission of the multilayer structure and find that small variations in the layer width (and accordingly small changes in Δ) translate to significant transmission modulations. As Fig. 2(a) shows very clearly, the transmission through the multilayer structure for incidence at the critical angle is strongly affected by minute (1 nm) variations in the layer thickness. Hence, it is easy to tell apart two structures identical in all aspects other than their layer thicknesses.

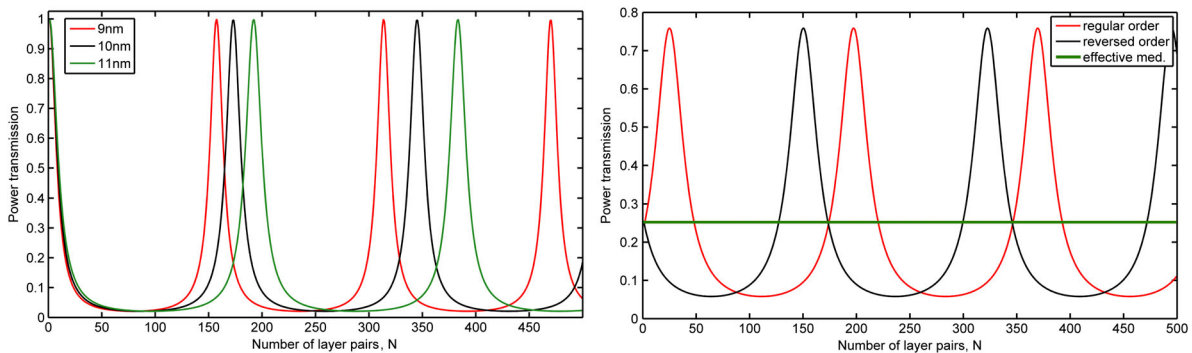


FIG. 2 (color online). Transmission as a function of structure width ($= Nd$) for several structures. (a) For layer thicknesses $d = 9, 10, 11$ nm, $\epsilon_{\text{in}} = \epsilon_{\text{out}} = 4$. (b) For $d = 10$ nm and $\epsilon_{\text{in}} = 4, \epsilon_{\text{out}} = \epsilon_{\text{eff}}$ with a regular layer order (ϵ_a followed by ϵ_b) or reversed (ϵ_b followed by ϵ_a). The prediction of the effective medium (EM) theory is marked by the yellow horizontal lines. All plots in (a) and (b), apart from the yellow line, are calculated via transfer-matrix formalism.

Evidently, the sensitivity observed in Fig. 2(a) contradicts the intuitive notion (prediction 1, above) that the transmission through a dielectric stack can be changed only by thickness variations on the optical wavelength scale.

To further demonstrate the breakdown of the effective medium description of the multilayer stack, we consider the case where the output permittivity ϵ_{out} is equal to ϵ_{eff} while $\epsilon_{\text{in}} = 4$, as before. We compare the transmission through two structures that are identical except that the order of layers is reversed in one with respect to the other. Clearly, effective medium theory should yield the same transmission for both structures, because ϵ_{eff} is not affected by the order of the layers. However, this seemingly trivial variation of the structure completely changes the transmission through this deep subwavelength multilayer stack. Because $\epsilon_{\text{out}} = \epsilon_{\text{eff}}$, the interface of the multilayer with ϵ_{out} should be impedance matched in the effective medium description and the boundary between the multilayer. Moreover the exterior ϵ_{out} should, from the effective medium perspective, be no boundary at all. Therefore, in the effective medium model, we expect to have a constant transmission through the multilayer—independent of the number of layers or their order (note that this transmission is not zero, since θ is the critical angle relative to $\bar{\epsilon}$ and we already found that $\epsilon_{\text{eff}} > \bar{\epsilon}$). However, in reality, the transmission through the multilayer is not constant. As the red and black lines in Fig. 2(b) show, the transmission strongly depends on both the *number* and the *order* of the layers. For example, when “regular ordering” is at a transmission resonance (transmission is 0.75), “reverse ordering” is off resonance (transmission is 0.1).

These results highlight the crucial role of the layer adjacent to the impedance matched exterior (ϵ_{out}). The effective medium model can be phenomenologically extended to account for these effects, by treating the stack as a slab with homogeneous permittivity ϵ_{eff} but assuming a nontrivial reflection coefficient of $r = r_0 e^{i\phi}$ at the impedance matched boundary. The value of this coefficient

cannot be found through the logic of effective medium concepts. Rather, it is extracted from Fig. 2(b), which is the result of the transfer-matrix calculation. For our choice of parameters $r_0 = 0.76$ (instead of $r_0 = 0$, the effective medium prediction), but the phase ϕ depends on the identity of the last layer. It is $\phi = -1.2$ rad when the structure terminates with an ϵ_b layer (regular layer order) and $+1.2$ rad when it terminates with an ϵ_a layer (reversed order). Changing the order of the layers effectively changes the identity of the last layer and drives the system from high to low transmission. A simpler way to change the identity of the last layer is to simply add a single ϵ_a layer to the structure while keeping the layer order fixed. Doing so results in a plot almost identical to Fig. 2(b), with the same large jump in transmission, even though the two structures only differ by *one* additional 10 nm thick layer.

This remarkable sensitivity to subwavelength variations in the structure can lead to many potential applications. First, as shown in Fig. 2(a), it can be used to measure the layer thickness d . More importantly, it can also be used to measure the properties of a *single* extremely thin layer. To highlight this, Fig. 3(b) shows the dependence of the total transmission of the stack as a function of the width of the terminating layer, which is a strong dependence when $\epsilon_{\text{out}} = \epsilon_{\text{eff}}$ but insignificant when $\epsilon_{\text{out}} = 4$. In addition to its potential application to sensing, extreme sensitivity is attractive for electro-optical manipulation of light—miniscule electro-optical modifications of light (for example, by a weak control beam) can induce large and significant transmission variations. The critical angle θ_c in this scheme does not change and the multilayer remains virtually loss-free.

Before proceeding further, it is important to note that this boundary effect does not appear just at the singular case where $\epsilon_{\text{out}} = \epsilon_{\text{eff}}$ precisely; rather, it occurs in a range of parameters around that point. To show this, consider the case where the exterior permittivity ϵ_{out} is varied continuously. For every value of ϵ_{out} , we can deduce the reflection

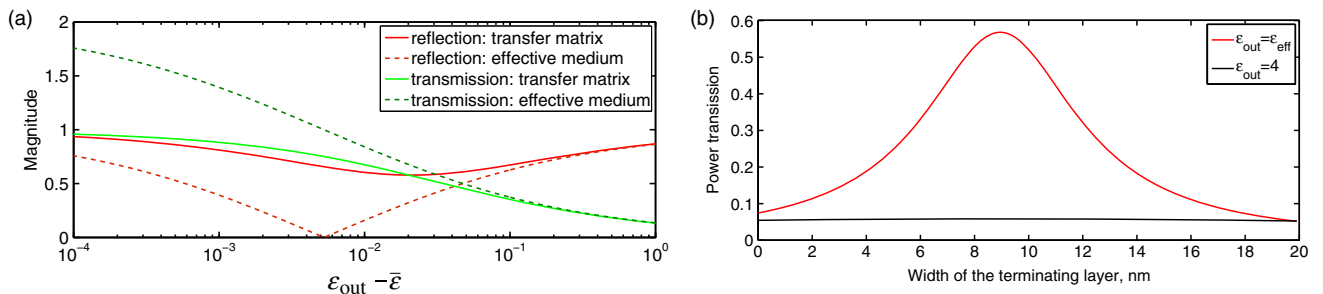


FIG. 3 (color online). (a) Amplitude of the reflection and transmission coefficients from the output boundary of the multilayer stack versus $\epsilon_{\text{out}} - \bar{\epsilon}$ deduced from the transfer-matrix calculation (dashed line) compared against the values found from naive effective medium theory (solid line). The results from both methods coincide for $\epsilon_{\text{out}} - \bar{\epsilon} > 0.1$, but clearly when the permittivity difference is small, effective medium yields erroneous results. (b) Transmission as a function of the width of the last layer in the structure, for two values of ϵ_{out} (red and black lines). Near impedance matching, the transmission is extremely sensitive to nanometric changes in the width of the last layer. Here, $N = 34$ and the layers are in reversed order (ϵ_b before ϵ_a).

and transmission coefficients from the transfer-matrix calculation and compare it against the value obtained from the effective medium approach. Figure 3(a) shows the magnitude of the coefficients obtained from both approaches for various values of $\epsilon_{\text{out}} - \bar{\epsilon}$. The effective medium result is reasonably accurate when $\epsilon_{\text{out}} - \bar{\epsilon} > 0.1$, but fails for a range of ϵ_{out} permittivities closer to $\bar{\epsilon}$. Also, we can see from this figure that there is no value of ϵ_{out} for which the reflection disappears ($r_0 > 0.6$ always). It is important to note that for a structure with thinner layers (or illumination with larger wavelengths) this boundary effect would appear on a narrower range of ϵ_{out} permittivities, and that in the $\lambda/d \rightarrow \infty$ limit, the existence regime of the effects vanishes. Nevertheless, as we have shown here, it is a significant effect at a broad range of λ/d ratios in which the effective medium approach is otherwise justified.

So far, we have considered only the macroscopic properties of the multilayer, such as the transmission of power, but close examination of the field reveals that even when the effective model is seemingly accurate, it fails to predict the actual distribution of the field. Since the layers we consider are extremely thin, the wave does not accumulate phase (or decay) considerably across one layer. However, it can accumulate a considerable phase on reflection (the Goos-Hanchen phase shift) because the Fresnel reflection coefficients are $r_{ab} = -r_{ba} = ((k_{z,a} - k_{z,b}) / (k_{z,a} + k_{z,b}))$, with $|r_{ab}| = 1$ and incidence exactly at θ_c yields $k_{z,a} = -ik_{z,b}$, and $r = \pm i$. Therefore, the way waves accumulate phase inside the structure (through Fresnel transmission and reflection) is fundamentally different from the “usual” case, where phase is accumulated through propagation only.

A magnitude unity reflection coefficient also implies that the reflected components of the field are strongly coupled and that we must account for all the orders of reflection to get a full physical picture. This is done implicitly in

the transfer-matrix formalism, in which we decompose the field into its forward propagating component E_+ and the backward propagating component E_- . When k_z is imaginary, E_+ decays in the forward direction and E_- decays in the backward direction. Using the same transfer-matrix formalism, we can also calculate the field distribution inside the structure and Fig. 4(a) shows that the $E_{\text{tot}}(z)$ resultant of this calculation is almost identical in the effective medium and layered descriptions. However, the components E_+ and E_- behave completely differently: In the effective model, in which the structure is essentially a Fabry-Perot etalon, E_+ and E_- have constant amplitudes and counterpropagating phases, whereas our analysis shows that their amplitudes depend on z with the same period as $E_{\text{tot}}(z)$, as shown in Fig. 4(a).

The phase of the two components also deviates from the effective medium prediction [Fig. 4(c)]. To understand why, we turn to observe the way energy is conserved in this system. Inside the higher permittivity (ϵ_a) layers (where $k_z \in \mathbb{R}$), the power carried in the z direction is simply the difference between the power carried by the forward and backward components $P_a = (\epsilon_a/2)[E_+^2 - E_-^2]$. Therefore, energy conservation implies that inside the ϵ_a layers [Fig. 4(b)] E_+ is always slightly larger than E_- so that P_a is conserved [30].

The situation is different in the lower permittivity (ϵ_b) layers. The power carried in the z direction in these layers is $P_b = \epsilon_b E_+ E_- \sin(\phi)$, where ϕ is the relative phase between E_+ and E_- (for the same reason a single evanescent wave cannot carry power) [30]. Since E_+ and E_- are z dependent, we can expect that the relative phase ϕ will also change accordingly, so that P_b is z independent in the ϵ_b layers. As shown in Fig. 4(c), the simulations corroborate this prediction: the peaks of $\sin(\phi)$ correspond to points where E_+ and E_- are small [the nodes in Fig. 4(a)], and $\sin(\phi)$ approaches 0 when E_+ and E_- are large. Calculating P_b explicitly shows that it is indeed

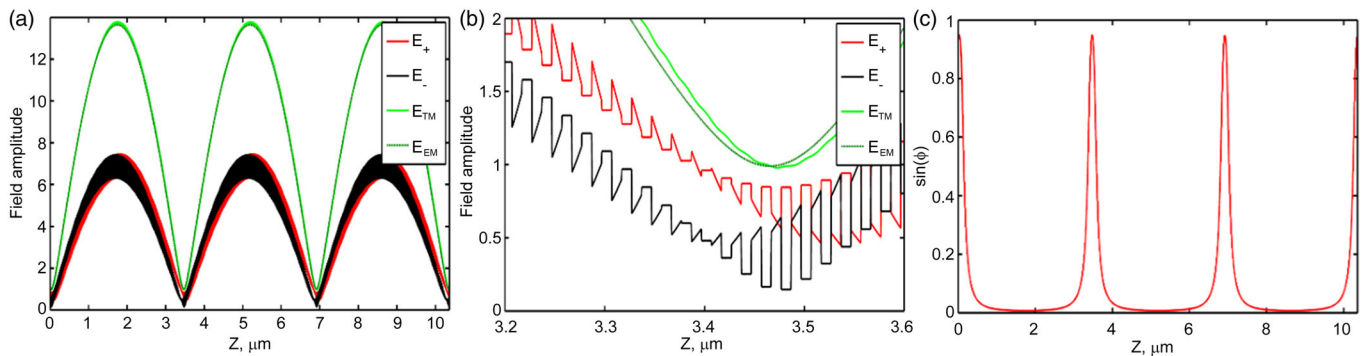


FIG. 4 (color online). (a) Amplitude of E_+ , E_- , the forward and backward propagating components, and $E_{\text{TM}} = E_+ + E_-$, the total field, as found through the transfer-matrix calculation exactly on the transmission resonance ($N = 518$) and the field E_{EM} found from the effective model. The total field found in both methods agrees closely, but the components do not. (b) A magnified portion of (a), showing the field amplitude inside the layers. (c) The sine of the relative phase between E_+ and E_- . The peaks of (c) correspond to the minima in (a).

constant in the ϵ_b layers. It is equal to P_a found in the higher permittivity layers.

In conclusion, we analyzed the transmission of light through a dielectric stack of alternating layers with thicknesses much smaller than the optical wavelength. We showed that, despite its simplicity, this structure displays intricate and counterintuitive effects at the vicinity of the critical angle for total internal reflection. Crucially, the waves in such a structure are alternately evanescent and propagating, while the phase is mostly accumulated via multiple Fresnel reflections. We showed that transmission through the multilayer can change dramatically when the layer thickness is varied by as little as 1 nm (relative to $\lambda = 500$ nm wavelength). Furthermore, we have shown that this type of structure cannot be impedance matched in the usual sense. That is, reflection always occurs at the output boundary, regardless of the permittivity of this exterior (even when the exterior is impedance matched with the effective permittivity of the structure). Since the phase of this reflection depends on the identity of the boundary layer, varying the order of the layers or adding a single 10 nm layer can change the transmission dramatically. All of these results constitute a clear breakdown of the effective medium description of our structure.

This work raises a variety of intriguing questions that are left for future research. For example, what happens in the intermediate regime, where the layer thickness is $d \sim \lambda/10$? From an applications-oriented perspective, our preliminary results indicate that such “intermediate” structures would be advantageous, because they display similar effects, but at the same time they are much more resistant to disorder and loss than those discussed here. Perhaps even more interesting is the issue of a disordered multilayer structure of subwavelength thicknesses. Would Anderson localization effects [31] be considerably altered in such a system? Answers to this question cannot rely on known asymptotic analyses of the Anderson localization, because the phases of electromagnetic waves, accumulated in these structures, arise from Fresnel reflections rather than propagation effects. Finally, many of the ideas presented here (with TE polarized EM waves) are general and carry over to other types of wave systems, such as quantum physics or acoustics.

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