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Negative refraction, gain and nonlinear effects in hyperbolic metamaterials

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Negative refraction, gain and nonlinear effects in hyperbolic metamaterials

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Abstract: The negative refraction and evanescent-wave canalization effects supported by a layered metamaterial structure obtained by alternating dielectric and plasmonic layers is theoretically analyzed. By using a transmission-line analysis, we formulate a way to rapidly analyze the negative refraction operation for given available materials over a broad range of frequencies and design parameters, and we apply it to broaden the bandwidth of negative refraction. Our analytical model is also applied to explore the possibility of employing active layers for loss compensation. Nonlinear dielectrics can also be considered within this approach, and they are explored in order to add tunability to the optical response, realizing positive-to-zero-to-negative refraction at the same frequency, as a function of the input intensity. Our findings may lead to a better physical understanding and improvement of the performance of negative refraction and subwavelength imaging in layered metamaterials, paving the way towards the design of gain-assisted hyperlenses and tunable nonlinear imaging devices.

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1. Introduction

Hyperbolic metamaterials [1–7] hold great promise to enable a wide range of novel, metamaterial-inspired electromagnetic devices that may become essential components of future microwave, infrared (IR) and optical circuits. These devices may realize lenses overcoming the diffraction limit [8–10], exhibit negative refraction [11,12], achieve perfect absorption [13,14] and broadband super-Planckian thermal emission [15], and increase the spontaneous radiation of emitters [16,17] in different frequency ranges.

Negative refraction may be achieved in these metamaterials thanks to the hyperbolic dispersion of these structures [1,2], which may also enable the conversion of a portion of the evanescent spectrum into propagating modes. As a result, subwavelength imaging can also be achieved in these structures, even at optical frequencies. It should be stressed that these media support a moderately broadband negative refraction phenomenon without a negative index of refraction [18], but instead based on the anomalous frequency and spatial dispersion of the metamaterial eigenmodes. This leads to more broadband performance compared to
conventional negative index materials, since many of the bandwidth limitations on the
effective parameter dispersion are relaxed. This is also combined with lower sensitivity to
material losses [19].

In this paper, we theoretically and numerically analyze a metamaterial composed of
alternating layers of dielectric and metal. This geometry has been shown to support
hyperbolic and flat dispersion, which leads to subwavelength resolution imaging [10]. In the
following, we apply a closed-form analytical technique to analyze, demonstrate and assess the
bandwidth performance of the device’s negative refraction operation based on the
transmission-line method [20], analogous to the transfer matrix technique [21]. This approach
is based on the fact that the negative refraction operation can be directly connected to the
condition for which the slope of the phase of the transmission coefficient is negative with
respect to the angle of incidence [11]. This allows us to efficiently analyze a broad range of
the design parameter space to explore and possibly optimize the negative refraction operation.
We show that several bands of high transmission can exist in different frequency ranges, for
which negative refraction can occur for a broad range of incident angles. In addition, our
theory allows to easily introduce into the picture other interesting anomalous effects arising
when optically active materials [22–25] and Kerr third-order nonlinear media [26] are
included in the alternating layers of the hyperbolic metamaterial. Loss compensation with
realistic gain materials is presented, opening new routes towards the design of broadband low
loss hyperbolic metamaterials. Finally, all-optical switching between positive refraction,
epsilon-near-zero behavior [27,28] and negative refraction with moderate input intensities is
also reported. We envision that the proposed metamaterial structures may lead to the design
of novel nonlinear electromagnetic devices, with exciting applications, such as all-optical
switches and tunable subwavelength imaging systems.

2. Theory of hyperbolic metamaterials

We start by studying the wave propagation in an inhomogeneous anisotropic slab composed
of alternating layers of dielectric (\(\varepsilon_d = 2\)) and plasmonic (silver, \(\varepsilon_{Ag}\)) materials with
thicknesses \(d_1\) and \(d_2\) respectively, as shown in Fig. 1(a). In the subwavelength limit
\((d_1 \ll \lambda, d_2 \ll \lambda)\), an effective medium description can be applied and the anisotropic effective
permittivity tensor of the structure is given by \(\varepsilon_{eff} = \varepsilon_t (\hat{x}\hat{x} + \hat{y}\hat{y}) + \varepsilon_z \hat{z}\hat{z}\), where [10,29]
\[
\varepsilon_t = \frac{\varepsilon_d d_1 + \varepsilon_{Ag} d_2}{d_1 + d_2}, \quad \varepsilon_z = \left(\frac{\varepsilon_d^{-1} d_1 + \varepsilon_{Ag}^{-1} d_2}{d_1 + d_2}\right)^{-1}.
\]

The dispersion relation of this anisotropic material for transverse-magnetic (TM)
propagation with magnetic field polarized along \(\hat{y}\) is then governed by the second-order
dispersion relation \(k_y^2/\varepsilon_t + k_z^2/\varepsilon_z = k_0^2\), where \(k_0\) is the free space wavenumber and \(k_x, k_z\)
are the transverse and longitudinal components of the wave vector, respectively. The ratio
between the power densities carried in the transverse and normal directions is then found
equal to
\[
\frac{S_x}{S_z} = \frac{\text{Re}(k_y/\varepsilon_t)}{\text{Re}(k_z/\varepsilon_z)}.
\]
Assuming for the moment real-valued permittivities, i.e., neglecting losses, Eq. (2) implies that, when the product $\varepsilon_x \varepsilon_z$ is negative (either $\varepsilon_x > 0, \varepsilon_z < 0$ or $\varepsilon_x < 0, \varepsilon_z > 0$), the wave is required to refract negatively at an interface with a conventional isotropic material with normal along $\hat{z}$, since $k_z$ and $k_S$ necessarily have opposite signs. This is consistent with the fact that the dispersion relation of the metamaterial becomes hyperbolic [6]. Inspecting Eq. (1), it is evident that for this to happen the condition $\varepsilon_x / \varepsilon_{\text{eff}} \geq -d_z / d_d$ is required. The “canalization” regime [10] is a special situation at the boundary between the two cases, and it is realized in the limit of $\varepsilon_x \rightarrow \infty$. In this case, a flat dispersion curve is obtained, opening the possibility for efficient subwavelength imaging. Ideally, an infinite number of spatial harmonics can be converted into propagating modes in this situation. To work in this regime, it is necessary for the parameters of the layered metamaterial to satisfy the relation $\varepsilon_x / \varepsilon_{\text{eff}} = -d_z / d_d$ [10]. As a result, the canalization regime can ideally be achieved only for a single frequency. In the present work, we focus most of our attention on the negative refraction condition for propagating waves in the layered metamaterial structure, which can be accessible over a continuous bandwidth, and of which the canalization regime is one of the boundaries.

Therefore, in the following, unless differently specified, we consider TM propagating waves with transverse wavenumber $k_z \leq k_0$ impinging on a metamaterial slab of length $L$ as indicated in Fig. 1(a). Later in the discussion, we will also consider higher spatial
harmonics that exhibit evanescent nature in free-space. For arbitrary spatial distribution, the magnetic field of the incoming beam traveling in air can be decomposed into a superposition of plane waves impinging upon the metamaterial slab at different oblique incident angles $\theta_i = \sin^{-1}(k_y/k_0)$ as: $H_y^i(x) = \int \tilde{H}_y(\mathbf{k}) e^{-j\mathbf{k}\cdot\mathbf{x}} d\mathbf{k}$. The interaction of each plane wave with the layered metamaterial may be efficiently, yet rigorously modeled using a transmission-line formalism [20], which allows calculating the overall ABCD matrix of the layered metamaterial through the multiplication of the ABCD matrices associated with each dielectric and plasmonic layer, as a function of the transverse momentum $k_y$. All these expressions are known in closed analytical form. Next, the total transmittance through the structure

$$T(k_y) = |T(k_y)| e^{j\phi(k_y)}$$

may be analytically calculated, and the magnetic field at the output of the device is simply

$$H^s_y(x) = \int \tilde{H}_y(\mathbf{k}) H_y(k_y, e^{j\phi(k_y)} x) d\mathbf{k}.$$  \hspace{1cm} (3)

Assuming that the impinging beam is a weakly modulated plane wave propagating along the incident angle $\theta_i$, i.e., its spectrum is confined around the wavenumber $k_y = k_0 \sin \theta_i$, we can consider a first-order Taylor expansion of the transmission spectrum around $k_y$, obtaining for the output beam the expression [11] $H^s_y(x) = T(k_y) e^{j\phi(k_y)} H_y(x - \Delta)$, where $\Delta = d\phi/dk_y$ is the spatial shift of the beam after the slab, and $\phi$ is the transmission phase. Essentially, in absence of sharp variations in the transmission coefficient around the wave number of interest, the transmitted beam is a copy of the impinging beam spatially translated by an amount proportional to the derivative of the phase of the transmission coefficient with respect to $k_y$. The effective angle of refraction of the transmitted wave can then be easily calculated from the spatial shift $\Delta$ as $\theta = \sin^{-1}(\Delta/L)$, where $L$ is the total length of the layered structure. If the spatial shift $\Delta$ is negative, i.e., the slope of the phase of the transmission coefficient as a function of the angle of incidence is negative, then negative refraction is expected. Note that this formula is in general more accurate for thicker structures and its precision deteriorates as the illuminating beam approaches grazing incident angles [12]. By having $T(k_y)$ in closed analytical form allows calculating in an efficient, yet rigorous way the refraction properties of the metamaterial by simply looking at the derivative of the phase of the transmission coefficient with respect to the transverse wave number. This allows us to quickly analyze the effect of several design parameters of the layered metamaterial on its index of refraction, and potentially optimize its design to achieve broader bandwidths of negative refraction. In general, the proposed analytical method represents a powerful tool to analyze negative refraction phenomena, obtained from complex metamaterial structures.

3 Results and discussion

3.1 Linear operation

The plasmonic layers of the structure in Fig. 1(a) are assumed to be made of Ag, for which the permittivity dispersion is based on experimental data retrieved in [30]. In this section we neglect absorption ($\text{Im}[\varepsilon_{Ag}] = 0$) in order to demonstrate more clearly the bandwidth performance in terms of negative refraction. The effect of realistic losses will be discussed in the next section. The electric field distribution of the layered metamaterial structure is shown in Fig. 1(b) for dimensions $d_1 = 500nm$, $d_2 = 250nm$ and 20 alternating layers, at the wavelength $\lambda = 323nm$ for which $\varepsilon_{Ag} = -0.5$. The fields are calculated with full-wave simulations based on the finite-integration method [31]. A TM polarized Gaussian beam
impinges on the structure with angle of incidence $\theta_i = 45^\circ$ and strong negative refraction ($\theta_i < 0$) is clearly achieved at this frequency. The weak reflection from the slab is caused by some impedance mismatch, which was not optimized in our design. Notice that, despite power flow undergoing a clear negative refraction inside the slab, the phase fronts refract positively, as expected from the previous considerations.

Next, we calculate the transmittance and angle of transmission of the proposed structure, based on the transmission-line method [20] and the previous discussion. For now, we focus only on the propagating spectrum of impinging waves. The transmittance and transmission angle ($\theta_t$) of the multi-layered slab with the aforementioned dimensions are shown in Figs. 1(c) and 1(d), respectively. The regions of special interest for our study are the ones in which the angle of transmission takes negative values, i.e., negative refraction, and at the same time a significant portion of the impinging energy is transmitted through the slab. The gray solid lines in the amplitude plot [Fig. 1(c)] enclose the region in which $\Delta < 0$ and negative refraction is dominant, in order to highlight our region of interest. With our analytical tools, and in particular with the derived condition to localize negative refraction, it is possible to efficiently and rapidly analyze arbitrary configurations and design parameters and localize the intersection between negative refraction and high transmittance, which is particularly interesting in order to improve the frequency and angular bandwidth of operation. Several bands of high transmission can be achieved, in which negative refraction occurs over a broad range of incident angles.

Note that negative refraction and transmission can strongly depend on the angle of incident radiation, as it can be clearly seen in Figs. 1(e) and 1(f), in which we calculate and plot the transmission and negative refraction based on the effective anisotropic permittivity tensor given by the approximate model of Eq. (1), where the slab has the same thickness as the layered metamaterial of Fig. 1(a). It is obvious that the homogenized model, based on the effective medium approximation, cannot take into account the inherent nonlocal effects and spatial dispersion of the layered metamaterial, as originally noticed in [32]. On the contrary, these interesting effects are fully taken into account when considering the layered structure in our transmission-line based calculations shown in Figs. 1(e) and 1(f). Using the proposed transmission-line method we can efficiently and rapidly evaluate the negative refraction angle, taking into full account the nonlocal effects of the metamaterial.
It is possible to apply the proposed theoretical analysis also to the evanescent portion of the impinging spectrum, in order to analyze the canalization of the evanescent spectrum into propagating waves and the potential for imaging with subwavelength resolution supported by the proposed structure. With this goal in mind, we extend the plot of transmittance of Fig. 1(c) to evanescent incident waves with wavenumbers $k_x > k_0$ in Fig. 2. It is seen that evanescent waves get fully transmitted over a broad range of frequencies and wave numbers, up to the cut-off around $k_x = 3k_0$, fundamentally determined by the granularity of the layered slab. The proposed analytical method is proven to be a useful tool not only for the prediction of negative refraction operation, but also to analyze the subwavelength resolution potentials of the proposed layered metamaterial hyperlens. It may be used to study and optimize different designs within the desired parameter space range, leading to a better understanding of the mechanisms of negative refraction and canalization of evanescent waves.

![Graphs](image)

Fig. 3. (a),(c) Transmittance and (b),(d) angle of transmission [in degrees] for the structure shown in Fig. 1(a) with parameters: (a),(b) $\varepsilon_d = 2.5$, $d_1 = 50nm$, $d_2 = 25nm$ and (c),(d) $\varepsilon_d = 2$, $d_1 = 50nm$, $d_2 = 35nm$ as a function of the incident angle ($k_x/k_0$) and the wavelength of operation ($\lambda$). In panels (a) and (c), the solid gray lines enclose the region where the angle of transmission is negative.

Our analysis and exploration of the available parameter space, made possible by the efficiency of the proposed analytical method, shows that the bandwidth of negative refraction of the presented metamaterial (region enclosed by the solid gray lines plotted at the transmittance figures above) can be broadened in two different ways: first, dielectric layers with higher permittivity should be used, if possible, relaxing one of the boundaries to obtain negative refraction, i.e., $\varepsilon_d / \varepsilon_{\infty} \geq -d_1 / d_2$. The computed transmittance and angle of transmission for $\varepsilon_d = 2.5$, with all other parameters fixed, are shown in Figs. 3(a) and 3(b), respectively. It is evident that the region of interest for which negative refraction occurs is...
larger compared to Figs. 1(c) and 1(d), as expected. As a side effect, the transmission bands in Fig. 3(a) are lower compared to the previous case of Fig. 1(c), because of a larger impedance mismatch. Alternatively, the thicknesses of the plasmonic layers may be increased \( d_2 = 35 \text{nm} \), which leads to an even broader area of negative refraction, as it can be seen in Figs. 3(c) and 3(d). The transmittance, however, is even more affected in this scenario compared to both previous cases shown in Figs. 1(c) and 1(d), due to the occurrence of larger resonance bands and a stronger dependence on the incident angle. A general trade-off exists between bandwidth and transmittance performance of the negative refracted waves.

3.2 Effect of losses and their compensation with gain materials

The results presented so far have been limited to lossless plasmonic layers, i.e., absorption has been neglected. We may expect significant changes in the response of the metamaterial structure when realistic losses are included. In the following, we include realistic Ag losses in the plasmonic layers and observe the effect on the negative refraction functionality. Now, the dielectric layers have permittivity \( \varepsilon_d = 2.5 \) and the structure has dimensions \( d_1 = 15\text{nm} \), \( d_2 = 12\text{nm} \) and 15 alternating layers. The metamaterial is composed of fewer and thinner layers to make sure that the impinging radiation is less dissipated. Moreover, we chose a different ratio of dimensions and permittivity values to achieve improved negative refraction bandwidth performance, applying the method described in the previous section. Realistic absorption is considered by introducing a non-zero imaginary part in the permittivity dispersion of silver \((\text{Im}[\varepsilon_{Ag}] \neq 0)\), in agreement with experimental data \[30\]. The transmittance and angle of transmission for this case are presented in Figs. 4(a) and 4(b),
respectively. Very poor transmission is obtained in Fig. 4(a) compared to the lossless case of Fig. 1(c). However, it is interesting that a relatively wide region of negative refraction is still present in Fig. 4(b), which is slightly affected by the introduction of losses, following a similar trend with the lossless response presented in Fig. 1(d). Note that the imaginary part of silver in the frequency region of interest is almost constant and equal to \( \text{Im} [\epsilon_{Ag}] = -0.6 \) [30].

This is consistent with recent experiments on structures realizing this effect based on semiconductor layers, which indeed supported negative refraction despite the very low transmission levels due to Ohmic losses [5]. We can conclude that negative refraction still exists for lossy structures and our method can be applied to efficiently analyze them and improve their performance, but the transmission levels are in general strongly affected, making the design of a realistic hyperlens with sufficient thickness quite impractical.

In order to alleviate this severe limitation, it has been proposed to use different semiconductor materials, such as aluminum-doped zinc oxide (AZO), which exhibit plasmonic response at IR frequencies combined with lower losses [33]. As an alternative approach, gain media can be introduced instead of ordinary dielectrics in the layers of the metamaterial structure. This is expected to compensate the inherent losses of the plasmonic material and the transmission will be restored to moderately higher values [24,25]. Here, we assume a gain material with inverse Lorentzian dispersion in the dielectric layers of the structure of Fig. 1(a) with dimensions \( d_1 = 15nm \), \( d_2 = 12nm \) and 15 alternated layers. The permittivity of the dielectric layers is now given by 
\[
\epsilon_d = \epsilon_\infty + \frac{\epsilon_p}{f^2 - f_0^2 - j\gamma f},
\]
which takes into full account the dispersive nature of the gain medium. The parameters are chosen to provide, around the frequency range of interest, a real part of permittivity close to \( \text{Re} [\epsilon_d] = 2.5 \) and a small positive imaginary part with peak value \( \text{Im} [\epsilon_d] = 0.06 \), consistent with dye-doped dielectric polymers [24,25]. The real and imaginary part of the gain material permittivity is plotted in the inset of Fig. 4(c) and the parameters of the Lorentz model are: 
\( \epsilon_\infty = 2.5 \), \( f_p = 100\text{THz} \), \( f_0 = 900\text{THz} \) and \( \gamma = 100\text{THz} \). The real part of permittivity of the gain medium is relatively flat and the imaginary part takes small positive values in a narrow frequency range around our design wavelengths. The results for transmittance and angle of transmission for a gain-assisted layered metamaterial slab are shown in Figs. 4(c) and 4(d), respectively. In this case, the bandwidth of negative refraction [Fig. 4(d)] remains similar to the lossy case [Fig. 4(b)] and slightly lower compared to the lossless case [Fig. 1(d)]. It is interesting that the transmittance performance [Fig. 4(c)] in part of the region of negative refraction is almost doubled compared to the lossy case [Fig. 4(a)]. Obviously this small gain cannot fully compensate the losses in silver, but our optimized design ensures strongly enhanced fields in the gain material, maximizing its effect. Higher transmission through the gain-assisted hyperlens is obtained for normal incidence. Hence, our analysis demonstrates that realistic gain media can indeed partly compensate the losses of plasmonic layers within a moderately wide range of frequencies and incident angles, improving the metamaterial negative refraction performance.

### 3.3 Nonlinear operation

To further extend the reach of these concepts and highlight the efficiency and generality of the proposed analytical method, in this section we consider the introduction of third-order \( \chi^{(3)} \) nonlinear effects in the dielectric layers of the lossless structure shown in Fig. 1(a). The dimensions of the structure are \( d_1 = 50nm \), \( d_2 = 25nm \), and 20 alternating layers are considered, similar with the lossless metamaterial presented before. In our previous works, we have shown that Kerr nonlinearities can induce bistability and tunability in the response of plasmonic structures [28,34–36]. Earlier works [37] have also considered multilayered stacks.
of nonlinear materials for different purposes. Here we introduce a nonlinear Kerr material in the dielectric layers with relative permittivity \( \varepsilon_r = \varepsilon_L + \chi^{(3)} |E|^2 \), where \( \varepsilon_L = 0.25 \) and \( \chi^{(3)} = 4.4 \times 10^{-18} \text{ m}^2/\text{V}^2 \) [26]. The permittivity essentially depends on the local fields at the thin dielectric layers \( d_i = 50 \text{nm} \) induced by the incident radiation intensity. The local fields inside each layer are almost uniform, due to the subwavelength thickness and, therefore, with very good approximation we can consider their average value to compute the nonlinear permittivity of each layer as a function of input intensity, as commonly done in similar configurations [28,37]. For simplicity, we neglect in this analysis the metal losses, but similar nonlinear response, yet with smaller excursions, is expected also when losses are considered.

![Figure 5](image)

Fig. 5. (a),(c) Transmittance and (b),(d) angle of transmission [in degrees] as a function of the input intensity, for the structure shown in Fig. 1(a) with third-order Kerr nonlinear material introduced in the dielectric layers. The results are computed for two different wavelengths: (a),(b) \( \lambda = 315 \text{ nm} \) and (c),(d) \( \lambda = 311 \text{ nm} \).

The transmittance and the angle of transmission for this nonlinear layered metamaterial structure as a function of the input intensity are shown in Figs. 5(a) and 5(b), respectively. Here we assume to operate at a single wavelength equal to \( \lambda = 315 \text{ nm} \), for which the plasmonic layers have permittivity \( \varepsilon_{Ag} = -0.2 \), and at an incidence angle \( \theta_i = 45^\circ \). The transmission drastically varies with the input intensity and the angle of the refracted beam may take positive, zero and negative values. In particular, there are three points of maximum transmission at \( I_{in} = 0 \text{GW/cm}^2 \), \( I_{in} = 2.7 \text{GW/cm}^2 \) and \( I_{in} = 6 \text{GW/cm}^2 \), where different behavior is obtained: positive refraction, epsilon-near-zero behavior, with an angle of refraction along the normal, like in a hyperlens, and negative refraction, respectively. Such high input intensities may be achieved with ultrafast pulsed laser sources.

We also computed the transmittance and angle of transmission for the same layered metamaterial at \( \lambda = 311 \text{ nm} \), for which now the plasmonic layers have permittivity \( \varepsilon_{Ag} = -0.1 \) in Figs. 5(c) and 5(d). High transmittance combined with large negative refraction

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is obtained in this case at two different input intensities: $I_{in} = 1.5 GW/cm^2$ and $I_{in} = 4.2 GW/cm^2$, respectively. Hence, tunable operation can be obtained by increasing the input intensity of the laser source that illuminates the nonlinear layered metamaterial and tuning its wavelength of operation. These results open exciting possibilities in using layered metamaterials for steerable and controllable flat lenses.

4. Conclusions

We have theoretically analyzed the negative refraction response and transmission of evanescent waves for a composite metamaterial structure composed of alternating dielectric and plasmonic layers. Our analysis is based on the transmission-line method, which has been proven to be an efficient tool for the characterization of hyperbolic metamaterial structures, taking into account nonlocal and spatial dispersion effects. Bandwidth, transmission performance and limitations of the presented hyperlens have been fully assessed. Our analysis may be applied to develop metamaterial designs with improved functionality in terms of bandwidth and transmission levels within the available parameter space. Realistic losses have been found to be detrimental to the performance of the device, consistent with recent experiments, and, in order to overcome this limitation, a reasonable amount of gain can be introduced, which has been shown to be able to improve the transmission where negative refraction is obtained. Finally, Kerr nonlinear materials have been considered in the dielectric layers and interesting tunable functionalities have been presented for different wavelengths of operation. This allows a properly designed metamaterial structure to support positive refraction, epsilon-near-zero behavior and negative refraction for different input intensities at the same frequency of operation. The proposed analysis may help better understanding and improving the negative refraction functionality of hyperbolic media. Our findings may lead to novel designs of active and nonlinear electromagnetic devices with exciting applications, such as gain-assisted low-loss hyperlenses, all-optical planar tunable spatial filters and switches and tunable subwavelength imaging systems.

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