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Wave scattering by metal-dielectric multilayer structures with gain

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We study wave scattering by multi-layer metal-dielectric structures of a finite extent characterized by a hyperbolic-like dispersion. We find the regimes of wave propagation when substantial compensation of losses becomes possible with the use of a gain medium with realistic parameters. We discuss the Purcell effect in these structures and its possible implications on the performance of the loss compensation.

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Metal-dielectric layered metamaterials (MDLMs) can exhibit several unusual properties and potentially useful functionalities [1], including superlenses with subwavelength resolution [2–4], hyperlenses [5], electromagnetic cloaking [6], nanolithography [7], and others. Significant part of the unusual properties of MDLMs are caused by the hyperbolic dispersion, i.e. when effective permittivity tensor has different signs of the two principal components (parallel and perpendicular to the layers) [8]. Hyperbolic media is now an active research area of metamaterials: they allow to strongly modify the spontaneous emission rate of the emitters located in such media [9–12] and find applications in subwavelength imaging [5], heat transport [13], and acoustics [14]. The use of MDLMs at optical frequencies is limited by the effect of losses which originates from absorption by metal layers. Losses are especially severe at low frequencies, where hyperbolic regime is observed. One of the straightforward ways to mitigate the losses in such structures is to incorporate a gain medium, such as organic dye molecules, semiconductor quantum wells or quantum dots, in dielectric layers [15–17].

The use of gain was suggested theoretically and in some cases demonstrated experimentally for various structures, including single-periodic and double-periodic photonic crystals [18, 19], fishnet metamaterials with negative refractive index [20–22], structures supporting localized surface plasmon polaritons [23–26], multilayered metamaterials [27–30], and others. In Ref. [28], it was theoretically predicted, that gain in MDLM in the photonic bandgap elliptical-dispersion regime at high frequencies improves the super-resolution properties. In Ref. [29], a loss compensation was claimed to be achieved in the hyperbolic-dispersion regime for the longitudinal propagation, i.e. when the transverse component of the wavevector remains the same as in the surrounding medium. It was found that full loss compensation is possible in an infinite MDLM in hyperbolic-dispersion regime only for a narrow range of the propagation angles, mostly because of the amplified spontaneous emission [30]. Importantly, compensation of losses is difficult to achieve because of high values of the gain required. Indeed, the values of the imaginary part of the dielectric permittivity of the gain medium required to compensate losses in a realistic structure [30] are large compared to the gain that can be achieved in experiments. In this letter, we predict that noticeable signal amplification may be achieved in realistic, finite-size MDLMs with a small number (several tens) of layers, that can be realized experimentally [9, 31, 32]. In particular, we consider a configuration that allows to couple the incident radiation, amplify and transmit it in large wavenumber hyperbolic-dispersion regime [8]. We also investigate a possibility of improving the experimentally measured characteristics, such as reflectance and transmittance, in realistic finite-size MDLM structures.

Geometry of the considered system is shown in Fig. 1. The TM-polarized plane wave impinges at an angle θ on the MDLM structure which has N periods of alternating metal (subscript "1") and dielectric (subscript "2") layers, and one additional metal layer (making the system symmetric). In what follows, we consider the dielectric layers made of PMMA with permittivity $\varepsilon_2 = 2.25$, and metal as silver described by experimentally measured dielectric function ε_1 taken from the Ref. [33].

In order to decrease losses and increase the amplification caused by the presence of gain, one should

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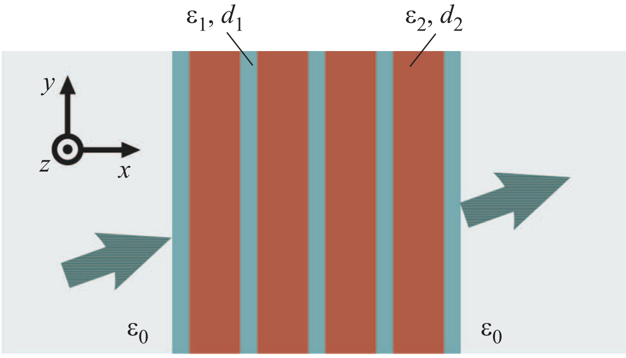

Fig. 1. (Color online) The geometry of a finite symmetric MDLM structure, consisting of $2N$ alternating metal and dielectric layers (marked by the subscripts "1" and "2", respectively) and one additional metal layer; ε_0 is permittivity of the cladding layers

choose thinner metal layers and thicker dielectric layers. On the other hand, there appears a certain range of the ratios of layers thicknesses for which hyperbolic-dispersion regime exists [34]. Here we choose the experimentally viable thicknesses of the layers $d_1 = 25$ nm and $d_2 = 75$ nm, $D = d_1 + d_2 = 100$ nm. To achieve efficient coupling of incident and transmitted light, one can either use a layer of random scatterers, diffraction grating or high-refractive index prisms [31, 35]. We choose the latter option, assuming that the coupling of light is performed using rutile prisms with high refractive index (with permittivity $\varepsilon_0 = 6.25$), and the prisms are placed on both sides of MDLM. As is shown in Fig. 2, hyperbolic-dispersion modes are

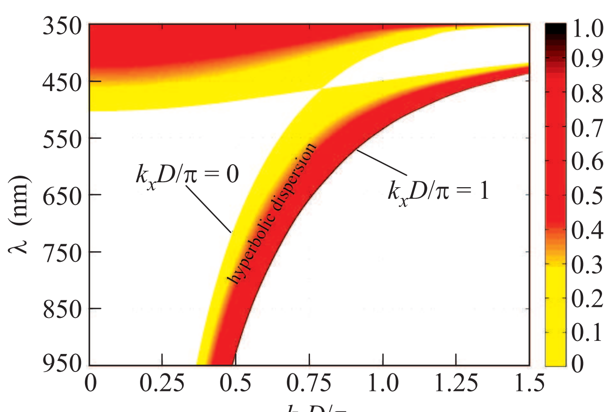

Fig. 2. (Color online) Dispersion diagram of an infinite MDLM; different values of the normalized transverse component of the Bloch wavevector $k_x D/\pi$ are shown by different colors

characterized by large longitudinal wavevector components. Choosing the refractive index of cladding layers as $n_0 = \sqrt{\varepsilon_0} = 2.5$ allows us to excite modes with maximum value of $(k_y D/\pi)_{\max} = 2(D/\lambda)n_0 = 5D/\lambda$, which cover the range of wavenumbers $\approx 0 \div 0.5$ for $\lambda = 1000$ nm, and $\approx 0 \div 1.0$ for $\lambda = 500$ nm, i.e. practically complete region characterized by the hyperbolic dispersion.

All calculations here are based on the transfer-matrix method, which is a common well-tested approach for the description of optical response of metal-dielectric layered structures [36–38]. We define a matrix that relates the complex amplitudes F and B of the H_z components of the waves propagating in the forward and backward directions, respectively, in two adjacent layers with numbers j and $j+1$:

$$\begin{pmatrix} F_j \\ B_j \end{pmatrix} = M_j \begin{pmatrix} F_{j+1} \\ B_{j+1} \end{pmatrix}. \qquad (1)$$

The matrix elements can be found from the boundary conditions for electromagnetic field, and they can be written as

$$M_{j-1} = \frac{1}{2}\begin{pmatrix} \left(1 + \frac{b_j}{b_{j-1}}\right) e^{-iq_j d_j} & \left(1 - \frac{b_j}{b_{j-1}}\right) e^{iq_j d_j} \\ \left(1 - \frac{b_j}{b_{j-1}}\right) e^{-iq_j d_j} & \left(1 + \frac{b_j}{b_{j-1}}\right) e^{iq_j d_j} \end{pmatrix}, \qquad (2)$$

where $b_j = q_j/\varepsilon_j$, d_j is the thickness of the j-th layer ($d_j = d_1$ for odd j, $d_j = d_2$ for even j, $d_{2N+2} = 0$), $q_j = k_0\sqrt{\varepsilon_j - (k_y/k_0)^2}$ is the normalized x-component of the wavevector in the layer j, ε_j is the permittivity of the j-th layer, $j = 1, \ldots, 2N+2$, $k_0 = n_0\omega/c$. Reflectance and transmittance can be calculated from the transfer matrix of the whole structure that relates the amplitudes of the incident, reflected and transmitted waves $M = M_0 \times M_1 \times \ldots \times M_{2N+1}$:

$$R = |r|^2 = \left|\frac{M_{2,1}}{M_{1,1}}\right|^2; \; T = |t|^2 = \left|\frac{1}{M_{1,1}}\right|^2. \qquad (3)$$

We note that such structures are often treated by using an effective medium approximation, since the period of the structure is much smaller than the wavelength. However, due to the existence of surface plasmon polaritons, the effective medium approach cannot provide an accurate description of the system even for large wavelengths [34, 36, 37], and one needs to use an exact description of an optical response. Truly hyperbolic-dispersion relation for the eigenmodes of such structures can be obtained only for relatively low frequencies, and it depends strongly on the ratio of the layer thicknesses [34].

Reflectance and transmittance spectra calculated by using Eq. (3) for the 21-layers structure with losses are shown in Fig. 3. Excited "hyperbolic modes" can be well

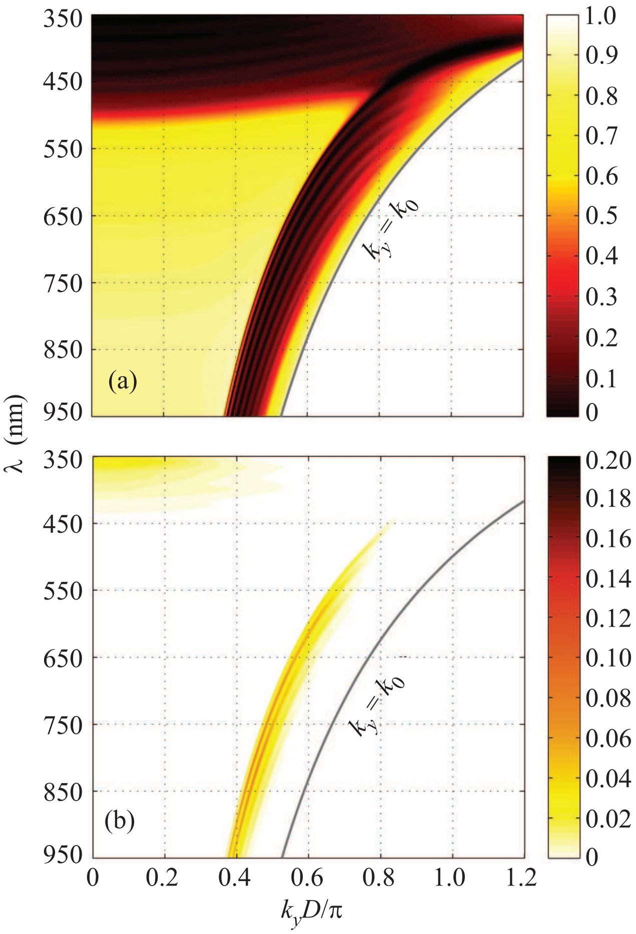

Fig. 3. Reflectance (a) and transmittance (b) calculated with Eq. (3) as functions of the wavelength λ and normalized longitudinal component of the wavevector k_yD/π. Areas below the curve $k_y = k_0$ correspond to the values of λ and k_y that cannot be excited in the considered configuration

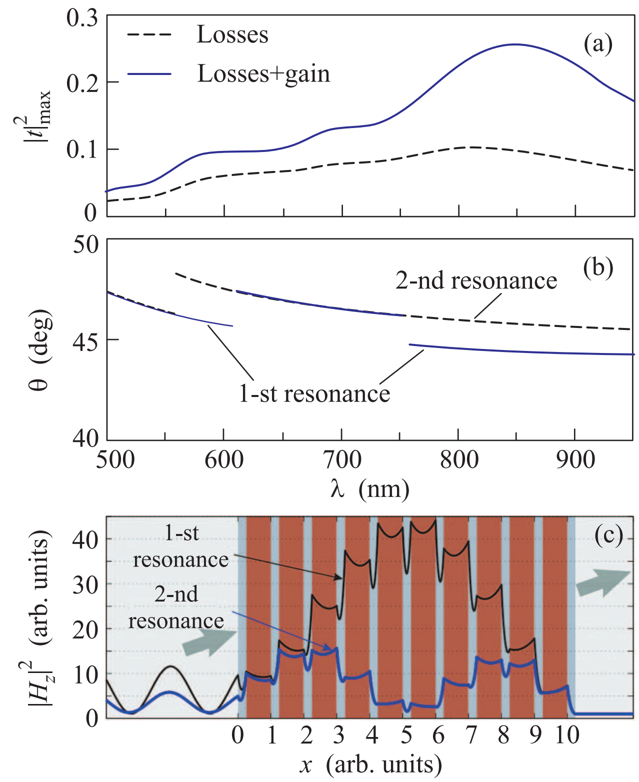

Fig. 4. (Color online) (a) – Maximum transmittance $|t|^2_{\max}$ as function of the operational wavelength in the case with losses and no gain (black dashed curve) and losses with gain (blue solid curve). (b) – Angles of incidence at which the maximum transmittance is observed. (c) – Field distributions on the first two resonances at $\lambda = 850$ nm

seen as the dips in reflectance (Fig. 3a) and peaks in transmittance (Fig. 3b) spectra. We notice that transmittance hardly reaches values of ≈ 0.2 because of the severe losses in metal. The field in the modes with larger k_y (i.e. those excited at larger angles of incidence) is concentrated more in metal layers, therefore absorption for larger angles of incidence is very high, and transmittance rapidly vanishes. As we are looking for experimentally observable effects, only the first few modes are of interest here.

To model the effect of gain in dielectric layers, we add a negative imaginary part to the permittivity of dielectric $\mathrm{Im}\,\varepsilon_2 = -0.005$ at the wavelength corresponding to the peak of the dye emission. There exists a number of dyes with different emission peak wavelengths, so for the frequency range of interest it is possible to choose one with required emission wavelength. Larger values of gain are very hard to obtain in experiments with MDLMs, and they cause undesirable effects of gain saturation and amplified spontaneous emission, while smaller values of gain do not allow to observe any noticeable improvement in both reflectance and transmittance spectra.

In Fig. 4a we show calculated maximum transmittance (at any angle of incidence) as a function of the wavelength for the case without (dashed black curve) and with gain (thin blue curve). In Fig. 4b we show angle of incidence at which maximum in transmittance is achieved. If one uses pumping across the layers, the angle of pump incidence θ_p should be close to the one of the first resonances, in order to produce maximal gain in the system. Since the emission and absorption peaks of the dye molecules are reasonably close to each other, θ_p should be approximately the same as in Fig. 4b (exact values of θ_p can be calculated from the Fig. 3b, by choosing the wavelength corresponding to absorption peak of dye molecules, rather than emission peak). Imaginary part of the silver permittivity $\mathrm{Im}\,\varepsilon_1$ which is responsible for losses increases for the large wavelengths, and the

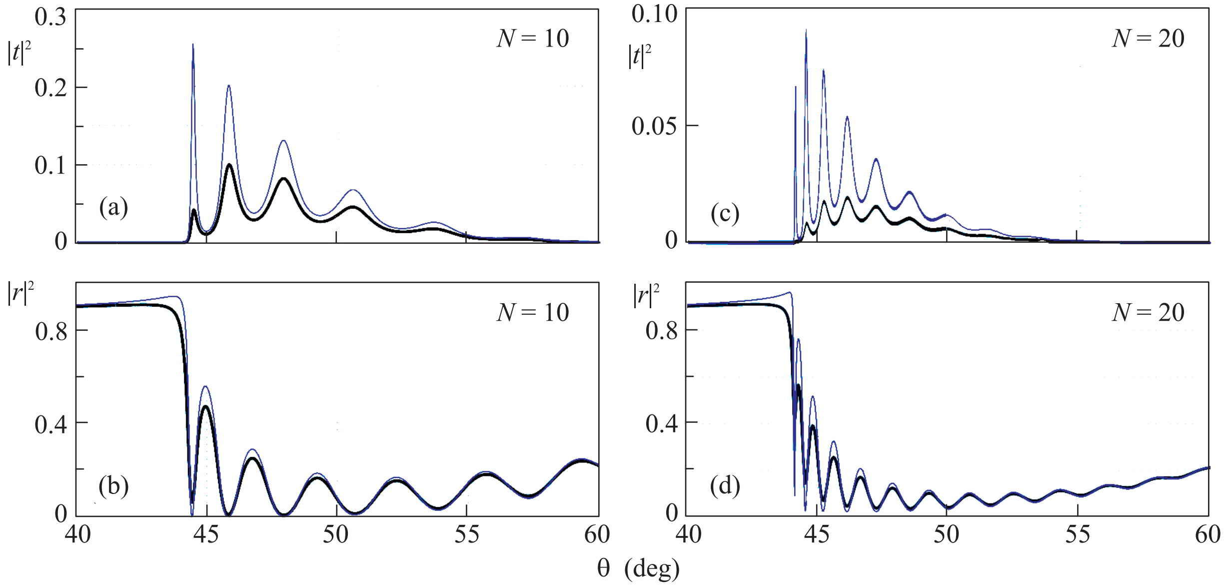

Fig. 5. (Color online) Transmittance (a, c) and reflectance (b, d) as functions of the angle of incidence for the wavelength of $\lambda = 850$ nm. (a, b) – Structure with 10 periods. (c, d) – Structure with 20 periods. Thin blue curves correspond to the case with gain $\mathrm{Im}\,\varepsilon_2 = -0.005$, thick black curves correspond to the case of no gain

real part $\mathrm{Re}\,\varepsilon_1$ becomes more negative, thus the field penetrates less into metal layers, which leads to lower overall losses. Such interplay leads to the existence of some optimal wavelength (850 nm for our structure) at which the maximum transmittance is achieved. Also, we observe that at any wavelength the maximum transmittance may be achieved either at the first or at the second resonance. Field distributions at these resonances represent the standing waves that consist of the set of coupled plasmons. They are shown in Fig. 4c in the case without gain (for $\lambda = 850$ nm). In the presence of small amount of gain they remain approximately the same.

Transmittance and reflectance as functions of the incident angle θ for the wavelength of $\lambda = 850$ nm are shown in Fig. 5. Reflected wave is formed in a first few layers of MDLM and therefore the effect of loss compensation by gain is rather difficult to observe in the reflectance spectra. On the contrary, a multiple increase in transmittance for the first several resonances is observed with rather modest amount of gain in the structure. However, this case is still far from complete compensation of losses since transmittance in the system with gain decreases with increase of the number of periods N. To achieve transmittance close to unity, higher gain is required, which might be difficult to obtain due to the effects of gain saturation and amplified spontaneous emission, as well as practical complications with pumping in experiments.

Since a strong increase of spontaneous emission rate may be considered as an inherent property of hyperbolic metamaterials, excited gain centers always radiate much faster in hyperbolic metamaterials, and therefore in the CW regime the required gain coefficient will increase proportionally to the Purcell factor [26]. In Fig. 6

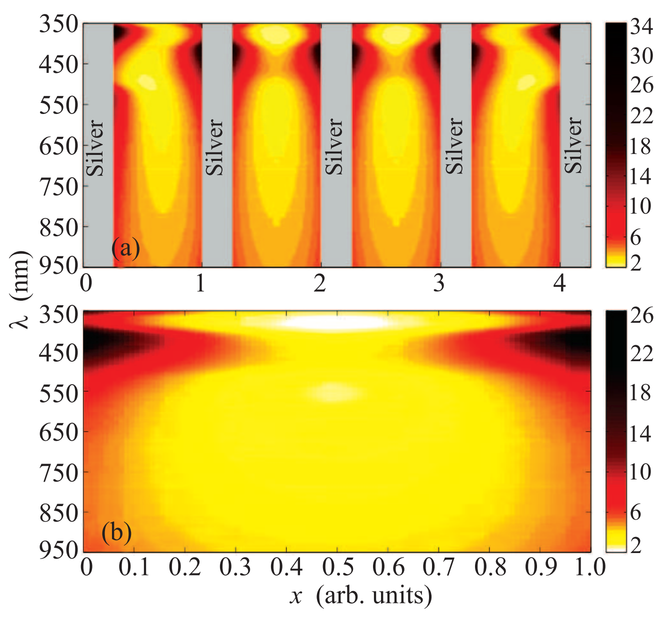

Fig. 6. Purcell factor for a finite-size MDLM structure with 9 layers (a) and one layer of an infinite MDLM structure (b) as function of wavelength λ and transverse coordinate z

we show the dependence of the Purcell factor, calculated with classical electrodynamics approach [39, 40] for randomly oriented dipole, on the wavelength λ and transverse coordinate z for the finite-size structure with 9 layers (Fig. 6a) and for one layer of an infinite structure (Fig. 6b). We observe that Purcell factor calculated for the infinite structure coincides with the one in finite structures even for small number of layers. The only noticeable difference occurs at the edges of the finite structure, where there is an additional channel of transferring energy from excited molecules, as the surface plasmon polaritons can be excited at the interface of the MDLM structure. For the structure with chosen parameters, the Purcell factor varies from about 2 in the center of dielectric layers to 10 and more at the edges, which makes the gain coefficient required for the full loss compensation even less attainable. Note, that the Purcell factor rapidly increases for thinner dielectric layers, and it can reach values of several hundreds and thousands [10]. However, in experiments one mostly uses pulsed optical pumping, and the effective gain coefficient will depend on the duration of the pump pulses. Since the lifetime of dye molecules in the excited state is several nanoseconds in vacuum [41], the use of pumping lasers with picosecond and shorter pulses will be required in MDLMs with gain.

In conclusion, we have studied the gain-induced loss compensation in metal-dielectric layered metamaterials with small number of layers and realistic parameters, operating in the regime of hyperbolic dispersion. We have shown that a substantial increase in transmittance can be achieved in a narrow range of the incidence angles. We have emphasized that the conditions for the full loss compensation in MDLM with the hyperbolic dispersion are hard to achieve due an amplified spontaneous emission and the Purcell effect in the continuous wave regime.

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