Overcoming Losses with Gain in a Negative Refractive Index Metamaterial

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On the basis of a full-vectorial three-dimensional Maxwell-Bloch approach we investigate the possibility of using gain to overcome losses in a negative refractive index fishnet metamaterial. We show that appropriate placing of optically pumped laser dyes (gain) into the metamaterial structure results in a frequency band where the nonbianisotropic metamaterial becomes amplifying. In that region both the real and the imaginary part of the effective refractive index become simultaneously negative and the figure of merit diverges at two distinct frequency points.

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Negative refractive index metamaterials offer the possibility of revolutionary applications, such as subwavelength focusing [1], invisibility cloaking [2], and "trapped rainbow" stopping of light [3]. The realization of these materials has recently advanced from the microwave to the optical regime [4,5]. However, at optical wavelengths metamaterials suffer from high dissipative losses due to the metallic nature of their constituent metamolecules. It is therefore not surprising that overcoming loss restrictions is currently one of the most important topics in metamaterials research [6].

It has been suggested that, owing to causality, simultaneous loss-compensation and negative refractive index might only be attainable in a very narrow bandwidth with high losses nearby [7]. In an ongoing discussion several authors have reasoned that causality-based criteria have to be applied carefully and do not in general lead to such a strict result [8,9]. This said, the theoretical possibility to compensate losses in optical metamaterials does not necessarily imply that the gain available from optically active media suffices to achieve this goal. Indeed, bulk gain coefficients are usually an order of magnitude smaller than the absorption coefficients of metals at optical frequencies.

A vital clue as to how the aforementioned limitation could be overcome came from [10]. There it was shown that the incorporation of gain in regions of high field intensity gives rise to an effective gain coefficient that can exceed its bulk counterpart by potentially orders of magnitude. Exploiting this gain-enhancement effect loss reduction and giant field enhancement have been reported for a double-fishnet metamaterial using frequency-domain models [11,12]. The particular model used in [11] accounts for spatial nonuniformity and saturation of the gain. However, the dynamic evolution of the gain system, nonlinearly pumped by a short intense pulse, cannot be described self-consistently with such a frequency-domain approach and relies on assumptions of cw excitation. A time-domain calculation of gain in two-dimensional electric or magnetic metamaterials has recently been reported in [13], but therein the effect of the pump field has again not been considered self-consistently.

In this Letter, we study the optical response of a negative refractive index (NRI) metamaterial with a gain medium embedded in the structure and we find that complete loss-compensation and even amplification is possible using realistic gain parameters. To this end we use a full-vectorial time-domain approach that manages to self-consistently couple the evolution of the occupation densities in the gain medium directly to Maxwell's equations in three dimensions [14–16]. Nonlinearity, saturation of the gain medium, and spatiotemporal variations of both absorption and emission are inherent to our model, avoiding the need for external, precalculated inputs.

The considered structure is an optical double-fishnet metamaterial [11,17,18] with a square periodicity of $p = 280$ nm, perforated with rectangular holes of sides $a_x = 120$ nm and $a_y = 80$ nm (see Fig. 1). The additional geometrical parameters are $h_m = 40$ nm, $h_d = 60$ nm, and $h_c = 60$ nm. This type of NRI metamaterial, which has been the topic of intense research (see, e.g., [11,17–19]),

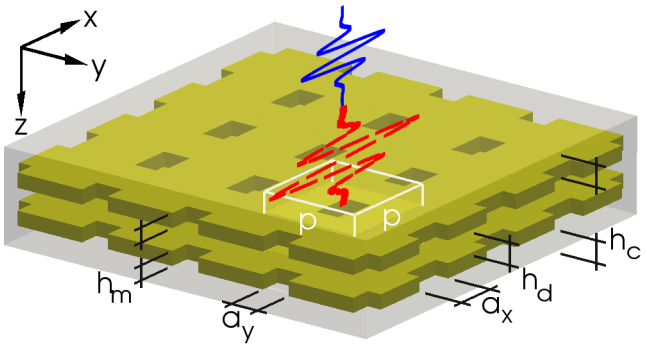

FIG. 1 (color online). Illustration of the double-fishnet structure with a square unit cell of side-length p highlighted. The two perforated silver films are embedded in a dielectric host material which holds the dye molecules (translucent). Dimensions are given in the text. Pump (red dashed line) and probe (blue solid line) pulses illustrate the pump-probe configuration with the electric field polarized along the x direction.

exhibits low absorption compared to other metamaterials in the optical wavelength range. Its relatively low absorption makes it the most promising structure for complete loss-compensation [20].

We consider two configurations, passive and active. In the passive configuration two silver fishnet films are embedded inside a dielectric host that has a real refractive index of $n_h = 1.62$ (see Fig. 1). The permittivity of silver follows a Drude model corrected by two Lorentzian resonances to match experimental data at visible wavelengths [21]. In the active configuration we insert Rhodamine 800 dye molecules into the dielectric host and excite them optically in numerical pump-probe experiments. The chosen geometric parameters ensure a good overlap of the metamaterial's resonant response with the emission spectrum of the dye for an electric field polarization along the long side a_x of the rectangular holes (see Fig. 1).

In order to self-consistently calculate the gain dynamics in this system the dye molecules are described using a semiclassical four-level model with two optical dipole transitions [14–16]. This model is implemented by introducing auxiliary differential equations for the position- and time-dependent polarization densities $\mathbf{P}_i$ and occupation densities N_j into the three-dimensional finite-difference time-domain (FDTD) algorithm. The time evolution of the polarization densities for the absorption ($i = a$) and emission ($i = e$) lines is then given by

$$\frac{\partial^2 \mathbf{P}_i}{\partial t^2} + 2\Gamma_i \frac{\partial \mathbf{P}_i}{\partial t} + \omega_{0,i}^2 \mathbf{P}_i = -2\omega_i \frac{e^2 d_i^2}{\hbar} \Delta N_i \cdot \mathbf{E}_{\mathrm{loc}}, \quad (1)$$

where $\omega_{0,i} = (\omega_i^2 + \Gamma_i^2)^{1/2}$ are the oscillator frequencies, $\hbar\omega_i$ the electronic transition energies, Γ_i the half-widths of the resonances, and ed_i the dipole strengths. The dye molecules embedded in the dielectric host experience, in the Lorentz approximation, the local electric field $\mathbf{E}_{\mathrm{loc}} = [(n_h^2 + 2)/3]\mathbf{E}$ and not the average electric field $\mathbf{E}$ [22]. Saturation is accounted for by the electric field dependence of the occupation inversions $\Delta N_a = N_3 - N_0$ and $\Delta N_e = N_2 - N_1$ for absorption and emission, respectively, which couple to Eq. (1). Their dynamics are governed by

$$\frac{\partial N_3}{\partial t} = \frac{1}{\hbar\omega_a}\left(\frac{\partial \mathbf{P}_a}{\partial t} + \Gamma_a \mathbf{P}_a\right) \cdot \mathbf{E}_{\mathrm{loc}} - \frac{N_3}{\tau_{32}}, \quad (2a)$$

$$\frac{\partial N_2}{\partial t} = \frac{N_3}{\tau_{32}} + \frac{1}{\hbar\omega_e}\left(\frac{\partial \mathbf{P}_e}{\partial t} + \Gamma_e \mathbf{P}_e\right) \cdot \mathbf{E}_{\mathrm{loc}} - \frac{N_2}{\tau_{21}}, \quad (2b)$$

$$\frac{\partial N_1}{\partial t} = \frac{N_2}{\tau_{21}} - \frac{1}{\hbar\omega_e}\left(\frac{\partial \mathbf{P}_e}{\partial t} + \Gamma_e \mathbf{P}_e\right) \cdot \mathbf{E}_{\mathrm{loc}} - \frac{N_1}{\tau_{10}}, \quad (2c)$$

$$\frac{\partial N_0}{\partial t} = \frac{N_1}{\tau_{10}} - \frac{1}{\hbar\omega_a}\left(\frac{\partial \mathbf{P}_a}{\partial t} + \Gamma_a \mathbf{P}_a\right) \cdot \mathbf{E}_{\mathrm{loc}}. \quad (2d)$$

Nonradiative decay of the occupation densities is quantified by the lifetimes τ_{jk}. The $\Gamma_i \mathbf{P}_i$ terms stem from the transformation from complex- to real-valued polarizations ([14], p. 174)

Absorption and emission cross-sections, taken from experimental data, are used to calculate the dipole length d_i via $\sigma_i = (\omega_{0,i} e^2 d_i^2/\hbar)/(\epsilon_0 c n_h \Gamma_i)$, with ϵ_0 being the vacuum permittivity and c the vacuum speed of light. The parameters for the four-level system are chosen as follows (cf. [23]): $\lambda_e = 2\pi c/\omega_e = 710$ nm, $\lambda_a = 680$ nm, $\Gamma_e = \Gamma_a = 1/(20\ \mathrm{fs})$, $d_e = 0.09$ nm, and $d_a = 0.1$ nm; $\tau_{32} = \tau_{10} = 100$ fs and $\tau_{21} = 500$ ps. These values correspond to cross sections $\sigma_e = 2.43 \times 10^{-16}$ cm^2 and $\sigma_a = 3.14 \times 10^{-16}$ cm^2. We set the density of the dye molecules as $N = \sum_{j=0}^{3} N_j = 6 \times 10^{18}$ cm^{-3} ≈ 10 mM leading to a bulk gain coefficient of approximately $g \approx N \cdot \sigma_e \approx 1460$ cm^{-1} at full inversion.

In order to study the active configuration we first pump the dye molecules with a short, intense pulse of duration 2 ps. After a short delay of 7 ps we probe the structure with a weak broadband pulse of duration 12 fs. Figures 2(a) and 2(c) show a snapshot of the spatial distribution of the occupation inversion generated by the pump pulse at two perpendicular planes inside the unit cell of the active metamaterial. The effective gain coefficient in the structure can be maximized with a good matching between the spatial distribution of the inversion and that of the plasmon-enhanced electric field amplitude at the emission wavelength λ_e [Figs. 2(b) and 2(d)]. Indeed, we see from

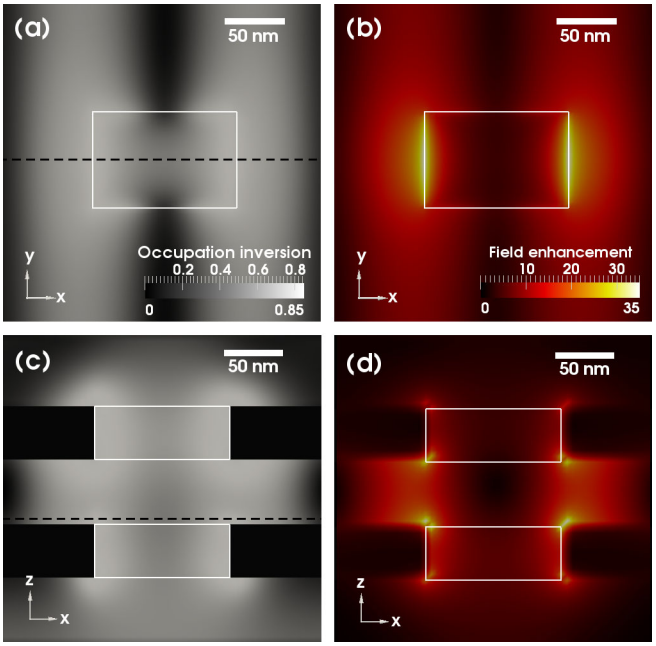

FIG. 2 (color online). (a) Snapshot of the occupation inversion ΔN_e in a plane 5 nm below the upper silver fishnet film just before probing and (b) the electric field enhancement at 710 nm in the same plane; both for a pump-field amplitude of 2.0 kV/cm. The silver film perforation is indicated by a white rectangle. (c) and (d) show the same physical quantities in a plane given by the propagation direction and the long side of the perforation. The dashed black lines in (a) and (c) highlight the positions of the intersection with the other plane, respectively.

Fig. 2 that such a matching is achieved when the pump and the probe have the same electric field polarization.

We remark that the considered planar structure has a low cavity Q-factor ($Q < 50$). In addition, the modal volume, which is directly associated with the feedback from metallic interfaces, is orders of magnitude larger than in the case of nanospheres used for "spasing" [24]. Therefore lasing is not expected in this structure. This is also supported by the fact that in our simulations we do not observe any appreciable depletion of the occupation inversion by the probe pulse. Further, on the short ps time scale under investigation, which is 3 orders of magnitude shorter than the free-space spontaneous emission lifetime of the dye, amplified spontaneous emission is not expected to play a significant role [25].

We use the standard retrieval method [26] to extract the effective refractive index $n = n' + in''$, first, of the passive configuration. Note that the metamaterial structure considered in this work is surrounded by air above and below the dielectric host; i.e., it is deliberately not placed on a thin substrate, in order to be symmetric and nonbianisotropic [27]. The spectral variation of n' (n'') in the passive structure is similar to that shown by the cyan solid (dashed) line in Fig. 3(a) (corresponding to the metamaterial that has dye molecules included but is not pumped). We find that in this passive case the figure of merit FOM $= -n'(\lambda)/n''(\lambda)$ has a maximum value of 2.7 at 713 nm.

Next, we study the active metamaterial configuration and, in particular, the effect that an increase of the pump intensity has on the obtained FOM. In the calculations we ensure that the gain system is probed within the linear regime where the standard retrieval method can be applied. Figure 3(a) shows the real and imaginary parts of the retrieved effective refractive indices for progressively increased peak pump-field amplitudes. We see that increasing the pump intensity, and therefore the gain available for the probe, leads to a decrease in the imaginary part of the refractive index in the region around the maximum emission cross section of the dye (710 nm) indicating reduced optical losses. The stronger resonance that arises from the gradually intensified pump also reduces the real part of the refractive index towards more negative values. Increasing the pump-pulse amplitude from 0.5 kV/cm to 1.5 kV/cm improves and blueshifts the maximum of the FOM from a value of 3 at 716 nm to a value of 12 at 712 nm (for the higher amplitude). An even higher amplitude of the pump pulse of 2 kV/cm results in a wavelength region (approx. 706–714 nm) where the losses in the metamaterial are completely compensated. In this region, the active metamaterial exhibits a negative absorption [cf. Fig. 4(b)] and both the real and imaginary part of the refractive index become simultaneously negative. Note from Fig. 3(b) that in this case the FOM diverges at the two wavelength points bounding the negative-absorption (amplification) region owing to n'' becoming exactly zero at these two wavelengths.

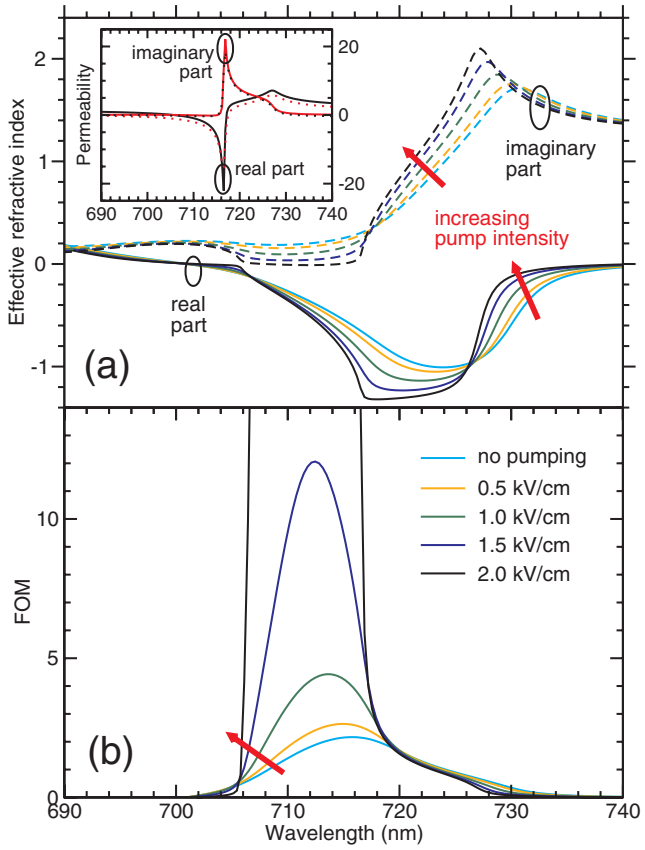

FIG. 3 (color online). (a) Real and imaginary part of the retrieved effective refractive indices of the double-fishnet structure for different pump amplitudes. The peak electric field amplitude of the pump increases in steps of 0.5 kV/cm from no pumping (cyan line, lightest) to a maximum of 2.0 kV/cm (black line, darkest). The inset shows the real and imaginary part of the effective permeability (black and red line, respectively) and the result of the Kramers-Kronig relation (black and red dotted lines) for the highest peak electric field amplitude of 2.0 kV/cm. (b) The figures-of-merit (FOM) for the same pumping amplitudes.

To further verify the causal nature of the obtained effective parameters we use the method of [28] to calculate, based on the Kramers-Kronig relations, the real (imaginary) part of the effective permeability from the imaginary (real) part of the numerically retrieved [26] effective permeability. An example of such a calculation for a pump amplitude of 2 kV/cm (corresponding to the negative-absorption regime) is shown in the inset of Fig. 3(a). The excellent agreement between the results obtained from the standard retrieval method and the complementary Kramers-Kronig approach further confirms that the extracted parameters do obey causality.

Finally, Fig. 4 presents a more detailed look at n'', the absorption coefficient, and the FOM for pump intensities close to and above complete loss compensation. We note that there is a critical amplitude of approximately 1.85 kV/cm for the pump pulse beyond which the present

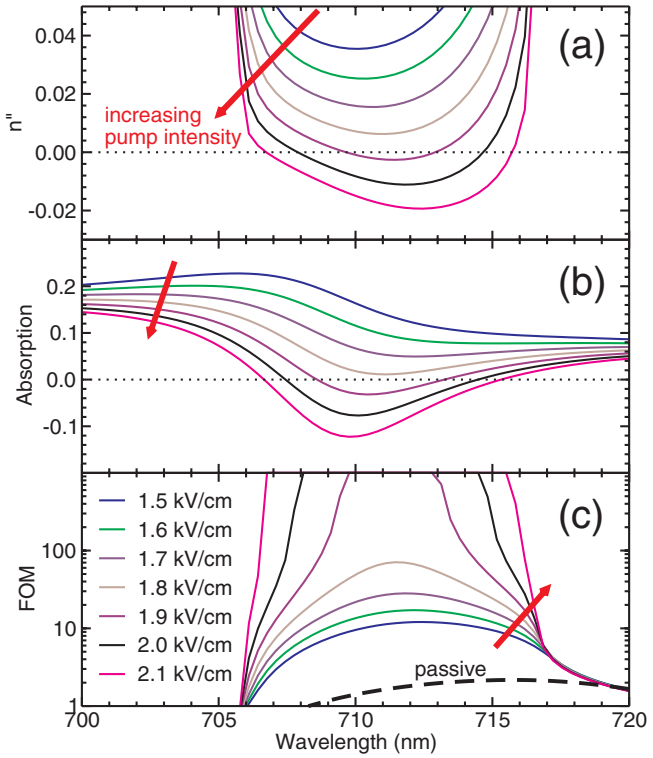

FIG. 4 (color online). Detailed view of (a) the imaginary part of the retrieved effective refractive indices n'', (b) the absorption, and (c) the figures of merit (FOM) for peak pump-field amplitudes close to and above compensation between 1.5 and 2.1 kV/cm in steps of 0.1 kV/cm.

metamaterial configuration becomes amplifying. A further increase of the pump field up to levels of 2.1 kV/cm leads to a spectral broadening of the region of negative absorption. At very large pump-field amplitudes above 2.2 kV/cm we observe discontinuities in the effective refractive index (data not shown here) and the Kramers-Kronig relation for the permeability is not obeyed any more. This occurs despite a smooth change in amplitude and phase of both the transmission and the reflection coefficients with no sign of gain depletion or discontinuities in other physical quantities. Investigation of this regime will be the subject of future work.

In conclusion, we have shown how the incorporation of a gain medium (Rhodamine 800 dye) into the structure of a double-fishnet nonbianisotropic metamaterial can fully compensate losses in the regime where the real part of the metamaterial's effective refractive index is negative. In this spectral range the imaginary part of the refractive index of the metamaterial becomes negative, too. We believe that this work could guide experimental efforts towards lossless and amplifying metamaterials, which offer access to exciting photonic applications.

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