## Self-consistent calculation of metamaterials with gain

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We present a computational scheme allowing for a self-consistent treatment of a dispersive metallic photonic metamaterial coupled to a gain material incorporated into the nanostructure. The gain is described by a generic four-level system. A critical pumping rate exists for compensating the loss of the metamaterial. Nonlinearities arise due to gain depletion beyond a certain critical strength of a test field. Transmission, reflection, and absorption data as well as the retrieved effective parameters are presented for a lattice of resonant square cylinders embedded in layers of gain material and split ring resonators with gain material embedded into the gaps.

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The field of metamaterials<sup>1,2</sup> is driven by fascinating and far-reaching theoretical visions such as, e.g., perfect lenses,<sup>3</sup> invisibility cloaking,<sup>4,5</sup> and enhanced optical nonlinearities.<sup>6</sup> This emerging field has seen spectacular experimental progress in recent years. 1,2 Yet, losses are orders of magnitude too large for the envisioned applications. Achieving such reduction by further design optimization appears to be out of reach. Thus, incorporation of active media (gain) might come as a cure. The dream would be to simply inject an electrical current into the active medium, leading to gain and hence to compensation of the losses. However, experiments on such intricate active nanostructures do need guidance by theory via self-consistent calculations (using the semi-classical theory of lasing) for realistic gain materials that can be incorporated into or close to dispersive media to reduce the losses at THz or optical frequencies. The need for self-consistent calculations stems from the fact that increasing the gain in the metamaterial, the metamaterial properties change, in turn changes the coupling to the gain medium until a steady-state is reached. A specific geometry to overcome the severe loss problem of optical metamaterials and to enable bulk metamaterials with negative magnetic and electric response and controllable dispersion at optical frequencies is to interleave active optically pumped gain material layers with the passive metamaterial lattice.

For reference, the best fabricated negative-index material operating at around  $1.4\,\mu\mathrm{m}$  wavelength<sup>7</sup> has shown a figure of merit (FOM) =  $-\mathrm{Re}(n)/\mathrm{Im}(n) \approx 3$ , where n is the effective refractive index. This experimental result is equivalent to an absolute absorption coefficient of  $\alpha=3\times10^4\,\mathrm{cm}^{-1}$ , which is even larger than the absorption of typical direct-gap semiconductors such as, e.g., GaAs (where  $\alpha=10^4\,\mathrm{cm}^{-1}$ ). So it looks difficult to compensate the losses with this simple type of analysis, which assumes that the bulk gain coefficient is needed. However, the effective gain coefficient, derived from self-consistent microscopic calculations, is a more appropriate measure of the combined system of metamaterial and gain. Due to pronounced local-field enhancement effects in the spatial vicinity of the dispersive metamaterial, the effective gain coefficient can be substantially larger than its bulk counter-

part. While early models using simplified gain-mechanisms such as explicitly forcing negative imaginary parts of the local gain material's response function produce unrealistic strictly linear gain, our self-consistent approach presented below allows for determining the range of parameters for which one can realistically expect linear amplification and linear loss compensation in the metamaterial. To fully understand the coupled metamaterial-gain system, we have to deal with time-dependent wave equations in metamaterial systems by coupling Maxwell's equations with the rate equations of electron populations describing a multi-level gain system in semiclassical theory.<sup>8</sup>

In this paper, we apply a detailed computational model to the problem of metamaterials with gain. The generic fourlevel atomic system tracks fields and occupation numbers at each point in space, taking into account energy exchange between atoms and fields, electronic pumping and non-radiative decays.<sup>8</sup> An external mechanism pumps electrons from the ground state level  $N_0$  to the third level  $N_3$  at a certain pumping rate  $\Gamma_{\mathrm{pump}}$ , which is proportional to the optical pumping intensity in an experiment. After a short lifetime  $\tau_{32}$  electrons transfer non-radiatively into the metastable second level  $N_2$ . The second level  $(N_2)$  and the first level  $(N_1)$  are called the upper and lower lasing levels. Electrons can be transferred from the upper to the lower lasing level by spontaneous and stimulated emission. At last, electrons transfer quickly and non-radiatively from the first level  $(N_1)$  to the ground state level  $(N_0)$ . The lifetimes and energies of the upper and lower lasing levels are  $\tau_{21}$ ,  $E_2$  and  $\tau_{10}$ ,  $E_1$ , respectively. The center frequency of the radiation is  $\omega_a=(E_2-E_1)/\hbar$  which is chosen to equal  $2\pi \times 10^{14}$  Hz. The parameters  $\tau_{32}$ ,  $\tau_{21}$ , and  $\tau_{10}$  are chosen  $5 \times 10^{-14}$ ,  $5 \times 10^{-12}$ , and  $5 \times 10^{-14}$  s, respectively. The total electron density,  $N_0(t=0) = N_0(t) +$  $N_1(t) + N_2(t) + N_3(t) = 5.0 \times 10^{23} / \text{m}^3$ , and the pump rate  $\Gamma_{\text{pump}}$  are controlled variables according to the experiment. The time-dependent Maxwell equations are given by  $\nabla \times \mathbf{E} = -\partial \mathbf{B}/\partial t$  and  $\nabla \times \mathbf{H} = \varepsilon \varepsilon_o \partial \mathbf{E}/\partial t + \partial \mathbf{P}/\partial t$ , where  $\mathbf{B} = \mu \mu_o \mathbf{H}$  and  $\mathbf{P}$  is the dispersive electric polarization density from which the amplification and gain can be obtained. Following the single electron case, we can show<sup>8</sup> that the po-

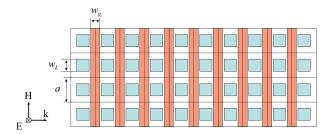


FIG. 1: (Color online) Square lattice of dielectric square cylinders (blue) that have a Lorentz behavior embedded in layers of gain material (red). The dielectric constant of the cylinders is given by  $\varepsilon=1+\omega_p^2/(\omega_p^2-2i\omega\gamma-\omega^2), \text{ where } f_p=\omega_p/2\pi=100\,\text{THz}$  and  $\gamma=2\pi f,$  and f takes different values in the cases we have examined. The dimensions are  $a=80\,\text{nm},\ w_L=40\,\text{nm}$  and  $w_g=30\,\text{nm}.$ 

larization density  $P(\mathbf{r},t)$  in the presence of an electric field obeys locally the following equation of motion

$$\frac{\partial^2 \mathbf{P}(t)}{\partial t^2} + \Gamma_a \frac{\partial \mathbf{P}(t)}{\partial t} + \omega_a^2 \mathbf{P}(t) = -\sigma_a \Delta N(t) \mathbf{E}(t) \quad (1)$$

where  $\Gamma_a$  is the linewidth of the atomic transition  $\omega_a$  and is equal to  $2\pi \times 5 \times 10^{12}\,\mathrm{Hz}$  or  $2\pi \times 20 \times 10^{12}\,\mathrm{Hz}$ . The factor  $\Delta N(\mathbf{r},t) = N_2(\mathbf{r},t) - N_1(\mathbf{r},t)$  is the population inversion that drives the polarization, and  $\sigma_a$  is the coupling strength of  $\mathbf{P}$  to the external electric field and its value is taken to be  $10^{-4}\,\mathrm{C}^2/\mathrm{kg}$ . It follows<sup>8</sup> from Eqn. 1 that the amplification line shape is Lorentzian and homogeneously broadened.<sup>9</sup> The occupation numbers at each spatial point vary according to

$$\frac{\partial N_3}{\partial t} = \Gamma_{\text{pump}} N_0 - \frac{N_3}{\tau_{32}} \tag{2a}$$

$$\frac{\partial N_2}{\partial t} = \frac{N_3}{\tau_{32}} + \frac{1}{\hbar \omega_a} \mathbf{E} \cdot \frac{\partial \mathbf{P}}{\partial t} - \frac{N_2}{\tau_{21}}$$
 (2b)

$$\frac{\partial N_1}{\partial t} = \frac{N_2}{\tau_{21}} - \frac{1}{\hbar \omega_a} \mathbf{E} \cdot \frac{\partial \mathbf{P}}{\partial t} - \frac{N_1}{\tau_{10}}$$
 (2c)

$$\frac{\partial N_0}{\partial t} = \frac{N_1}{\tau_{10}} - \Gamma_{\text{pump}} N_0 \tag{2d}$$

where  $\frac{1}{\hbar\omega_a}\mathbf{E}\cdot\frac{\partial\mathbf{P}}{\partial t}$  is the induced radiation rate or excitation rate depending on its sign.

In order to solve the behavior of the active materials in the electromagnetic fields numerically, the finite-difference timedomain (FDTD) technique is utilized,  $^{10}$  using an approach similar to the one outlined in Refs. 10–12. In the FDTD calculations, the discrete time and space steps are chosen to be  $\Delta t=8.33\times 10^{-18}\,\mathrm{s}$  and  $\Delta x=5.0\times 10^{-9}\,\mathrm{m}$  for simulations on the structure as shown in Fig. 1, and  $\Delta t=8.33\times 10^{-19}\,\mathrm{s}$  and  $\Delta x=1.0\times 10^{-9}\,\mathrm{m}$  for simulations on the structure as shown in Fig. 5. The initial condition is that all the electrons are in the ground state, so there is no field, no polarization and no spontaneous emission. Then the electrons are pumped from  $N_0$  to  $N_3$  (then relaxing to  $N_2$ ) with a constant pumping rate  $\Gamma_{\mathrm{pump}}$ . The system begins to evolve according to the system of equations above.

We have performed numerical simulations on onedimensional (1D) and two-dimensional (2D) systems with

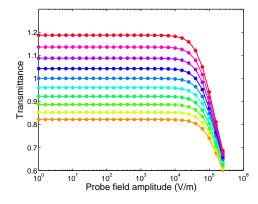


FIG. 2: (Color online) The transmittance vs. probe field amplitude for the loss-compensated metamaterial of Fig. 1 with gain bandwidth 5 THz, loss bandwidth 20 THz (i.e., f=10 THz), for different pumping rates  $\Gamma_{\rm pump}$ .  $\Gamma_{\rm pump}$  is increased from  $2.15\times10^9\,{\rm s}^{-1}$  (lowest) to  $3.05\times10^9\,{\rm s}^{-1}$  (highest) in steps of  $0.1\times10^9\,{\rm s}^{-1}$ . The metamaterial response is linear in a very wide range. When the loss-compensated transmission is exactly unity, the pumping rate  $\Gamma_{\rm pump}=2.65\times10^9\,{\rm s}^{-1}$ , which is called the critical pumping rate. For incident fields stronger than  $10^4\,{\rm V/m}$  this metamaterial becomes non-linear

gain. 13 Previous studies 14,15,16,17,18 have considered loss reduction by incorporating gain but where not self-consistent (see introduction). <sup>14,15,16,17</sup> As the first simple model system, we will discuss a 2D metamaterial system (shown in Fig. 1) which consists of layers of gain material and dielectric wires that have a resonant Lorentz type electric response to emulate the resonant elements in a realistic metamaterial. We will have to study whether we will be able to compensate the losses of the metamaterials associated with the Lorentz resonance in the wires by the amplification provided by the gain material layers without destroying the linear response of the metamaterial. First we generate a narrow band Gaussian pulse of a given amplitude and let it propagate through the metamaterial without gain, and we calculate the transmitted signal emerging from the metamaterial which has also Gaussian profile but the amplitude is much smaller than that of the incident pulse depending on how much loss occurs in the metamaterial. Then we introduce the gain and start increasing the pumping rate and find a critical pumping rate,  $\Gamma_{\text{pump}} = 2.65 \times 10^9 \, \text{s}^{-1}$ , for which the transmitted pulse is of the same amplitude as the incident pulse. In addition, for fixed pumping rate, we start increasing the amplitude of the incident Gaussian pulse and we would like to see how high we can go in the strength of the incident electric field and still have full compensation of the losses, i.e. the transmitted signal equals the incident signal, independent on the signal strength. In this region we have compensated loss and still linear response of the metamaterial; here, the shape of the transmitted Gaussian is only affected by the dispersion but not dependent on the signal strength.

We have calculated the transmission versus the strength of the electric field of the incident signal for several pumping rates close to the critical pumping rate. As shown in Fig. 2, we found that for a rather broad region of low intensity input signal we have a linear response all the way up to incident elec-

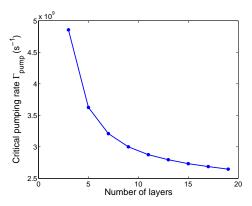


FIG. 3: (Color online) The critical pumping rates for different numbers of layers of the system in Fig. 1. Parameters for gain and dielectric materials are the same as Fig. 2.

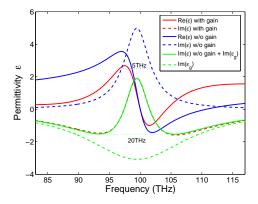


FIG. 4: (Color online) The retrieved results for the real and the imaginary parts of the effective permittivity,  $\varepsilon$ , with and without gain. In addition, we have plotted  ${\rm Im}(\varepsilon_g)$  versus frequency. Below compensation, t=0.75; Gain and Lorentz bandwidths are 20 and  $5~{\rm THz}$ , respectively.

tric field of  $10^3 \, \mathrm{V/m}$ . If we use only three layers (rods - gain material - rods), the critical pumping rate is  $4.85 \times 10^9 \,\mathrm{s}^{-1}$ , which is two times higher than the 19-layer case of Fig. 1. In Fig. 3, we present detailed results for the critical pumping rate versus the number of layers of the system shown in Fig. 1. Notice that as the number of layers increases, the critical  $\Gamma_{\mathrm{pump}}$  decreases. The linear regime for three layers exists up to  $10^4\,\mathrm{V/m},$  and for higher strength drops slower than that of Fig. 2. In all the following simulations, the strength of the incident signal is chosen to be 10 V/m, which is far away from  $10^3 \,\mathrm{V/m}$ , so we operate in the linear regime of the metamaterial. As an example, we have studied three layers, rods - gain material - rods, to see how much  $\Gamma_{\mathrm{pump}}$  we need to compensate the losses. As expected, we found that  $\Gamma_{\mathrm{pump}}$ is proportional to the imaginary part of the permittivity  $\varepsilon$  of the dielectric.

We first present results for three layers of the system shown in Fig. 1. First, the full width at half maximum (FWHM) for Lorentz dielectric and gain are chosen to be 5 and  $20\,\mathrm{THz}$ , respectively. With the introduction of gain the absorption at the resonance frequency of  $100\,\mathrm{THz}$  decreases, ultimately reach-

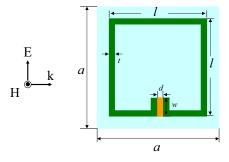


FIG. 5: (Color online) Geometry for a unit cell of the square SRR system with gain. The gain (shown in orange) is introduced in the gap region of the SRR. The dimensions are  $a=100\,\mathrm{nm},\,l=80\,\mathrm{nm},\,t=5\,\mathrm{nm},\,d=4\,\mathrm{nm}$  and  $w=15\,\mathrm{nm}.$ 

ing zero (not shown). So the gain compensates the losses. In Fig. 4, we plot the retrieved results for the real and imaginary parts of  $\varepsilon$  without gain and with gain slightly below compensation (see Ref. 19 for the retrieval method). Notice that we can have the  $Re(\varepsilon) \approx -1$  with  $Im(\varepsilon) \approx 0$  at  $102 \, THz$ , slightly off the resonance frequency. From Fig. 4, one can also see that  $Re(\varepsilon) \approx 2.5$  with  $Im(\varepsilon) \approx 0$  at 97 THz. So one can obtain a lossless metamaterial with positive or negative  $Re(\varepsilon)$ . Once we introduce gain, the imaginary part of  $\varepsilon$  of our total system with gain is equal to the sum of  $\mathrm{Im}(\varepsilon)$ without gain and the imaginary part of  $\varepsilon_q$ , the dielectric function of the gain material. This result is unexpected, because there is no coupling between the 2D Lorentz dielectric with the gain material. This is indeed true because of the continuous shape of the Lorentz dielectric cylinders and the gain material slabs have zero depolarization field. In contrast to finite length wires (hence a 3D problem) where the dipole interactions between Lorentz dielectric and gain material would be dominated by the quasi-static nearfield  $O(1/r^3)$ , here the interaction is order  $O(\omega \ln |kr|)$ , only via the propagating field, and much weaker. Therefore, for this 2D model, gain and loss are approximately independent. The behavior would obviously be different in a 3D situation, which, however, is computationally excessively demanding. Thus, we consider a 2D version of the split ring resonator (SRR) as a more realistic and also more relevant model. Here, the relevant polarization is across the finite SRR gap and, therefore, the coupling to the gain material is in fact dipole like.

In Fig. 5, we present the unit cell of our SRR system with gain material embedded in the SRR gap. The dimensions of the SRR are chosen such that a magnetic resonance frequency at  $100\,\mathrm{THz}$  results, which can overlap with the peak of the emission of the gain material. The FWHM of the gain material is  $20\,\mathrm{THz}$ , and  $\Gamma_{\mathrm{pump}}$  is  $1.4\times10^9\,\mathrm{s^{-1}}$ . Simulations are done for one layer of the square SRR. In Fig. 6a, we plot the retrieved results of the real and the imaginary parts of the magnetic permeability,  $\mu$ , with and without gain. With the introduction of gain, the weak and broad resonant effective  $\mu$  (FWHM =  $5.85\,\mathrm{THz}$ ) of the lossy SRR becomes strong and narrow (FWHM =  $1.66\,\mathrm{THz}$ ); the gain effectively undamps the LCR resonance of the SRR. Notice that here losses in the

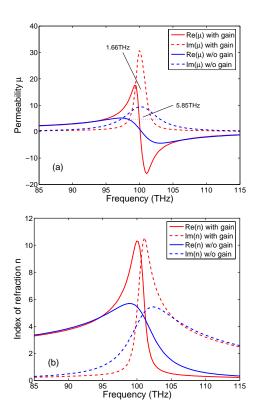


FIG. 6: (Color online) The retrieved results for the real and the imaginary parts of (a) the effective permeability  $\mu$  and (b) the corresponding effective index of refraction n, with and without gain for a pumping rate  $\Gamma_{\rm pump} = 1.4 \times 10^9 \, {\rm s}^{-1}$ . The gain bandwidth is 20 THz. Notice that the width of the resonance with gain is 1.66 THz.

magnetic effective response are compensated by electric gain in the SRR gap. So with the introduction of gain, we obtain a negative  $\mu$  with a very small imaginary part in an otherwise typical SRR response, which means that the losses have been compensated by the gain. In Fig. 6b, we plot the retrieved results for the effective index of refraction n, with and without gain. Note that for a lossless SRR n is purely real away from the resonance and imaginary in a small band above the resonance where  $\mu$  is negative. Comparing  $\mathrm{Re}(n)$  slightly below the resonance at 97 THz, we find an effective extinction coefficient  $\alpha = (\omega/c)\,\mathrm{Im}(n) \approx 3.50 \times 10^4\,\mathrm{cm}^{-1}$  without gain, and  $\alpha \approx 1.24 \times 10^4\,\mathrm{cm}^{-1}$  with gain, and hence an effective amplification of  $\alpha \approx -2.26 \times 10^4\,\mathrm{cm}^{-1}$ . This is much larger

than the expected amplification  $\alpha \approx -1.39 \times 10^3 \, \mathrm{cm}^{-1}$  for the gain material at the given pumping rate. <sup>20</sup> The difference can be explained by the field enhancement in the gap of the resonant SRR. The induced electric field in the gap is around 550 V/m, which is still in the linear regime, and the incident electric field is 10 V/m. Indeed, taking the observed field enhancement factor in the SRR gap of  $\approx 55$ , the energy per unit cell produced by the gain material inside the gap is  $\approx 18$  times larger than for the homogeneous gain medium which compares very well to the factor  $\approx 20$  between the simulated SRR effective medium and the homogeneous gain medium. If we further increase the pumping rate the magnetic resonance becomes even narrower (0.96 THz for  $\Gamma_{\text{pump}} = 1.8 \times 10^9 \, \text{s}^{-1}$ ). When the pumping rate reaches  $\Gamma_{\text{pump}} = 1.9 \times 10^9 \,\text{s}^{-1}$ ,  $Im(\mu)$  becomes negative and we have overcompensated at the resonance frequency. By increasing  $\Gamma_{\mathrm{pump}}$  even more  $(\approx 5 \times 10^9 \,\mathrm{s}^{-1})$  one starts seeing lasing (spasing)<sup>21,22</sup> in our system (not shown), which is not in the focus of this work. As long as we are in the linear regime, we do not need to have a self-consistent calculation, our results agree very well with the results obtained using the susceptibilities given in Ref. 9. However, the self-consistent calculation is necessary to determine the range of signals for which we can expect approximately linear response and it is needed if we have very strong fields and we are in the nonlinear regime, especially when we want to study lasing.

In conclusion, we have proposed and numerically solved a self-consistent model incorporating gain in 2D dispersive metamaterials. We show numerically that one can compensate the losses of the dispersive metamaterials. There is a relatively wide range of signal amplitudes for which the loss-compensated metamaterial still behaves linearly; at higher amplitudes the response is non-linear due to the gain. As an example, we have demonstrated that the losses of the magnetic susceptibility  $\mu$  of the SRR can be easily compensated by the gain material. The pumping rate needed to compensate the loss is much smaller than the bulk gain. This aspect is due to the strong local-field enhancement inside the SRR gap.

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<sup>&</sup>lt;sup>9</sup> The real and imaginary parts of the gain profile is given by  $P/\varepsilon_0 E = \chi' + i\chi''$ , where  $\chi' = -\chi''_0 \Delta x/(1+\Delta x^2)$  and  $\chi'' = \chi''_0/(1+\Delta x^2)$  with  $\Delta x = 2(\omega-\omega_a)/\Gamma_a$  and  $\chi''_0 = -\sigma_a \Delta N/(\varepsilon_0 \omega_a \Gamma_a)$  where  $\Delta N = N_2 - N_1$ .

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