Invited Paper

Nonlocal modification and quantum optical generalization of effective-medium theory for metamaterials

Martijn Wubs,[a,b] Wei Yan,[a,b] Ehsan Amooghorban,[c] and N. Asger Mortensen[a,b]

[a]DTU Fotonik, Department of Photonics Engineering, Technical University of Denmark, DK-2800 Kongens Lyngby, Denmark;

[b]Center for Nanostructured Graphene, Technical University of Denmark, DK-2800 Kgs. Lyngby, Denmark;

[c]Department of Physics, Faculty of Basic Sciences, Shahrekord University, P.O. Box 115, Shahrekord 88186-34141, Iran

ABSTRACT

A well-known challenge for fabricating metamaterials is to make unit cells significantly smaller than the operating wavelength of light, so one can be sure that effective-medium theories apply. But do they apply? Here we show that nonlocal response in the metal constituents of the metamaterial leads to modified effective parameters for strongly subwavelength unit cells. For infinite hyperbolic metamaterials, nonlocal response gives a very large finite upper bound to the optical density of states that otherwise would diverge. Moreover, for finite hyperbolic metamaterials we show that nonlocal response affects their operation as superlenses, and interestingly that sometimes nonlocal theory predicts the better imaging. Finally, we discuss how to describe metamaterials effectively in quantum optics. Media with loss or gain have associated quantum noise, and the question is whether the effective index is enough to describe this quantum noise effectively. We show that this is true for passive metamaterials, but not for metamaterials where loss is compensated by linear gain. For such loss-compensated metamaterials we present a quantum optical effective medium theory with an effective noise photon distribution as an additional parameter. Interestingly, we find that at the operating frequency, metamaterials with the same effective index but with different amounts of loss compensation can be told apart in quantum optics.

Keywords: Nanoplasmonics, hyperbolic medium, spontaneous emission, quantum optics, metamaterials, effective medium, balanced homodyne detection

1. INTRODUCTION

Metamaterials are artificial structures with unit cells much smaller than the operating frequency, which allow an effective-medium description of a type often not occurring in nature. Sometimes metamaterials are more specifically defined as structures that exhibit negative refraction. There are several ways to extract the effective-medium parameters. As long as the unit cells are much smaller than the operating frequency, the values for the effective parameters essentially do not depend on the parameter retrieval method. More importantly, for these metamaterials, the common opinion is that at the optical frequency, the only information that one obtains about the metamaterial is its overall shape and (possibly tensorial) effective index. Moreover, once the unit cell is small enough, varying the size of the unit cell does not change the effective parameters.

In practice it is extremely challenging to make unit cells much smaller than visible wavelengths. When unit cells are not much smaller than the operating frequency, then the values of effective parameters do depend on the parameter retrieval method chosen. This is not surprising and for these nanostructures it does not really make much sense to enforce an effective-medium description.

Further author information: (Send correspondence to M.W.)
M.W.: E-mail: mwubs@fotonik.dtu.dk, Telephone: +45 - 45 25 32 05
W.Y.: E-mail: wyan@fotonik.dtu.dk
E.A.: E-mail: ehsan.amooghorban@sci.sku.ac.ir
N.A.M.: E-mail: asger@mailaps.org

Metamaterials: Fundamentals and Applications VI, edited by Allan D. Boardman, Nader Engheta, Mikhail A. Noginov, Nikolay I. Zheludev, Proc. of SPIE Vol. 8806, 88061F · © 2013 SPIE · CCC code: 0277-786X/13/$18 · doi: 10.1117/12.2024465

However, returning to true metamaterials, one can ask what is the status of the statement that in a metamaterial only the effective index and the overall geometry play a role in optical experiments. Was this proven, or is it an experimental finding? In this paper we address two fundamental limits of metamaterials where the usual effective-medium theories no longer apply. First we consider the small-scale limit in Sec. 2, where the description of the constituents of the unit cells in terms of bulk refractive indices breaks down. Here our key references are Refs. 1, 2. Then in Sec. 3 we study the quantum optical limit, where an accurate description of the quantum states of light that propagate through the metamaterial in some cases requires an effective-medium theory that differs from the usual one for classical light. Here our key reference is Ref. 3. Our conclusions and outlook can be found in Sec. 4.

2. SMALL-SCALE LIMIT

Since we address fundamental limits of metamaterials, we consider simple geometries that allow for analytical treatment, so that our findings will not be obscured by numerical details. One-dimensional structures are obvious candidates for which powerful transfer matrix methods exist, while point-scattering models would allow also two-and three-dimensional inhomogeneities.[4,5] Here we will consider metamaterials of the former, layered type. A special class of layered metamaterials that attracts quite some attention nowadays are so-called hyperbolic metamaterials, where the effective dielectric function is positive-valued in one direction and negative in the other. This can occur in binary multilayer media for which one type of layer is a metal with negative-valued dielectric function ε_m, while the other dielectric layer has a positive ε_d.

2.1 Local and nonlocal dispersion relations of hyperbolic metamaterials

The effective-medium description requires that unit cells are much smaller than an optical wavelength. Here we ask what happens if we make the unit cells smaller and smaller, while keeping the filling fractions of the metal and the dielectric the same. At some point the description of the layers in terms of their dielectric functions ceases to apply, because new physical phenomena start to play a role. We anticipate that the first new phenomenon that kicks in will be nonlocal response in the metal layer. We will compare predictions for metamaterials with local and with nonlocal response. With local response it is meant that the polarization field $\mathbf{P}(\mathbf{r},\omega)$ can be written as $\varepsilon_0\varepsilon(\mathbf{r},\omega)\mathbf{E}(\mathbf{r},\omega)$, whereas for nonlocal response the latter expression becomes $\varepsilon_0\int \mathrm{d}\mathbf{r}'\varepsilon(\mathbf{r},\mathbf{r}',\omega)\mathbf{E}(\mathbf{r}',\omega)$. In our description of the metamaterials their nonlocal response arises because the free-electron response of their metallic constituents can no longer be described by the usual Drude model. We employ the hydrodynamic Drude model instead, where inhomogeneities in free-electron densities give rise to a pressure.[6,7] The corresponding pressure waves, with typical wavelengths of one nanometer or less, make the response of the metamaterials nonlocal.

For local response the effective dielectric function parallel to the layers is given by $\varepsilon_\parallel^{\mathrm{loc}} = f_\mathrm{d}\varepsilon_\mathrm{d} + f_\mathrm{m}\varepsilon_\mathrm{m}$, and perpendicular to the layers by $1/\varepsilon_\perp^{\mathrm{loc}} = f_\mathrm{d}/\varepsilon_\mathrm{d} + f_\mathrm{m}/\varepsilon_\mathrm{m}$, where $f_{m,d}$ are the space-filling fractions of the respective layers. The effective dispersion relation is $k_\perp^2/\varepsilon_\parallel^{\mathrm{loc}} + k_\parallel^2/\varepsilon_\perp^{\mathrm{loc}} = k_0^2$. Thus with local response, constant-frequency contours in the $(k_\parallel, k_\perp)$ plane can become hyperbolas for metal-dielectric multilayers, and hence the name hyperbolic metamaterials. With nonlocal response on the other hand, the dispersion relation of these hyperbolic media becomes more complicated, actually no longer a hyperbola for large wave vectors, as we reported in Refs. 1, 2. In the realistic parameter regime where the unit cell is much smaller than a wavelength while the metallic layers are many wavelengths of the pressure waves thick, the nonlocal dispersion relation can be approximated as

$$\frac{k_\perp^2}{\varepsilon_\parallel^{\mathrm{loc}}} + \frac{k_\parallel^2}{\varepsilon_\perp^{\mathrm{nloc}}} = k_0^2, \qquad \text{with} \quad \frac{1}{\varepsilon_\perp^{\mathrm{nloc}}} = \frac{1}{\varepsilon_\perp^{\mathrm{loc}}} + \frac{1}{\varepsilon_\perp^{\mathrm{hdm}}} \qquad \text{and} \quad \varepsilon_\perp^{\mathrm{hdm}} = \frac{k_m^{\mathrm{L}} d}{2i}\frac{\varepsilon_m^{\mathrm{T}}}{\varepsilon_m^{\mathrm{T}} - 1}, \tag{1}$$

where $d = d_m + d_d$ is the total thickness of the unit cell, and k_m^{L} is the wave vector of the longitudinal waves that equals $[\omega^2 + i\gamma\omega - \omega_p^2]^{1/2}/\beta$, in terms of the Drude damping γ, the plasma frequency ω_p, and the nonlocal parameter $\beta = \sqrt{3/5}v_\mathrm{F}$ that is proportional to the Fermi velocity.

The main message from Eq. (1) is that nonlocal response can modify effective parameters of metamaterials. The modification is stronger for smaller unit cells, as one can verify for oneself by considering the dependence of

$\varepsilon_\perp^{\rm nloc}$ on the unit cell thickness d. More generally, when making the unit cells smaller, one first enters the desired subwavelength regime where the effective-medium properties of the metamaterial no longer depend on the size of the unit cell, only on its geometry.[8] But when continuing to shrink the unit cell, in our case corresponding to unit cells with metal layers of less than 10 nm thick, then the size of the unit cell starts to play a role again because of nonlocal response. The associated longitudinal pressure waves have wavelengths of the order of a single nanometer in metals. Unit cells will not be in the subwavelength regime of the pressure waves, and these waves probe the size of the metal constituents of the unit cell.

We also see from Eq. (1) that nonlocal response modifies the effective dielectric response normal to the layers, not parallel to the layers. It is also intuitive that stronger nonlocal effects are to be expected in the direction in which the hydrodynamic pressure waves are confined by the metal boundaries. Notice also that in the limit $\beta \to 0$ nonlocal response becomes negligible and the dispersion relation (1) tends to the usual local-response hyperbolic dispersion relation, with effective dielectric response $\varepsilon_\perp^{\rm loc}$ in the normal direction.

2.2 Nonlocal blueshift of the optimal operating frequency

A hyperbolic metamaterial of finite thickness can be used as a so-called hyperbolic superlens. When placing the source in the near field at one end of the lens, the image in the near field at the other end can be almost perfect, at least for the operating frequencies that we discuss below. For perfect lensing, plane waves with different $k_\parallel$, even those with $k_\parallel > \omega/c$ should act together, i.e. in phase, to make the image. Such a lens is better if the phase accumulated upon propagation in the normal direction depends less on $k_\parallel$. When is this the case? From the dispersion relation (1) it follows that $k_\perp$ depends least on $k_\parallel$ when $\varepsilon_\perp^{\rm nloc} \to \infty$. For local response the condition is analogous but different, namely $\varepsilon_\perp^{\rm loc} \to \infty$. From these two different conditions it follows that the optimal operating frequencies for perfect lensing with hyperbolic metamaterials are different: for local response, it is at $\omega_{\rm res}^{\rm loc} = \omega_p[f_d/(f_d + f_m\varepsilon_d)]^{1/2}$, whereas the hydrodynamical Drude model with nonlocal response predicts that the optimal operating frequency should be chosen at the higher frequency $\omega_{\rm res}^{\rm nloc} \simeq \omega_{\rm res}^{\rm loc}[1 + \varepsilon_d/(k_m^{\rm L} d_d)]$, see Ref. 2 for more details.

We choose a metamaterial with 36 unit cells, each unit cell consisting of a dielectric layer with $d_d = 6\,$nm and $\varepsilon_d = 10$, and a metal layer with $d_m = 3\,$nm. The metal is described as a free-electron gas with the Drude parameters of gold.[2] With these parameters, it follows that the optimal operating frequency for the superlens can lie considerably higher than expected based on local-response theory: we find $\omega_{\rm res}^{\rm loc} = 0.41\omega_p$, whereas $\omega_{\rm res}^{\rm nloc} = 0.465\omega_p$. This is a manifestation in metamaterials of the nonlocal blueshift, the same effect that is believed to be largely responsible for the recently observed 0.5 eV blueshift of resonance frequencies in few-nanometer small plasmonic spheres.[9,10] The nonlocal blueshift of the operating frequency from $0.41\omega_p$ to $0.465\omega_p$ would be clearly observable if the assumed ultrathin metal-dielectric multilayers can be fabricated. There are interesting developments in this direction.[11] For less extremely thin slabs, the nonlocal blueshift would be smaller but could still be clearly observable in planar hyperbolic metamaterials.

2.3 Transmission through superlenses for local and nonlocal response

Since imaging has a lot to do with transmission, we will now first compare the transmission of plane waves through the hyperbolic metamaterial for local and for nonlocal response, and we will do this at both frequencies that we just identified as optimal according to the two respective theories.

We consider transmission both of propagating plane waves ($k_\parallel < k_0$) and of evanescent plane waves ($k_\parallel > k_0$), since it is only the combination of both that makes the lens "super". The result can be found in Fig. 1, at $\omega = \omega_{\rm res}^{\rm loc}$ in panel (a) and $\omega = \omega_{\rm res}^{\rm nloc}$ in panel (b). Notice that these graphs were calculated based on the full nonlocal dispersion relation that we did not present in its full glory here (but see Refs. 1, 2). What we can appreciate, however, is that in panel (a) the transmission graph as a function of $k_\parallel$ for local response is more smooth than for nonlocal response. This is expected based on the divergence of $\varepsilon_\perp^{\rm loc}$ of the local-response version of the approximate dispersion relation Eq. (1) near $\omega = \omega_{\rm res}^{\rm loc}$. By contrast, in panel (b) it is the nonlocal transmission curve that is smoothest. This is also as expected, since at $\omega = \omega_{\rm res}^{\rm nloc}$ it is $\varepsilon_\perp^{\rm nloc}$ (rather than $\varepsilon_\perp^{\rm loc}$) that diverges in the approximate dispersion relation Eq. (1).

The transmission is bounded below unity for propagating waves ($k_\parallel < k_0$), but shows higher resonances for those evanescent waves ($k_\parallel > k_0$) that correspond to a guided mode. Similar transmissions higher than unity

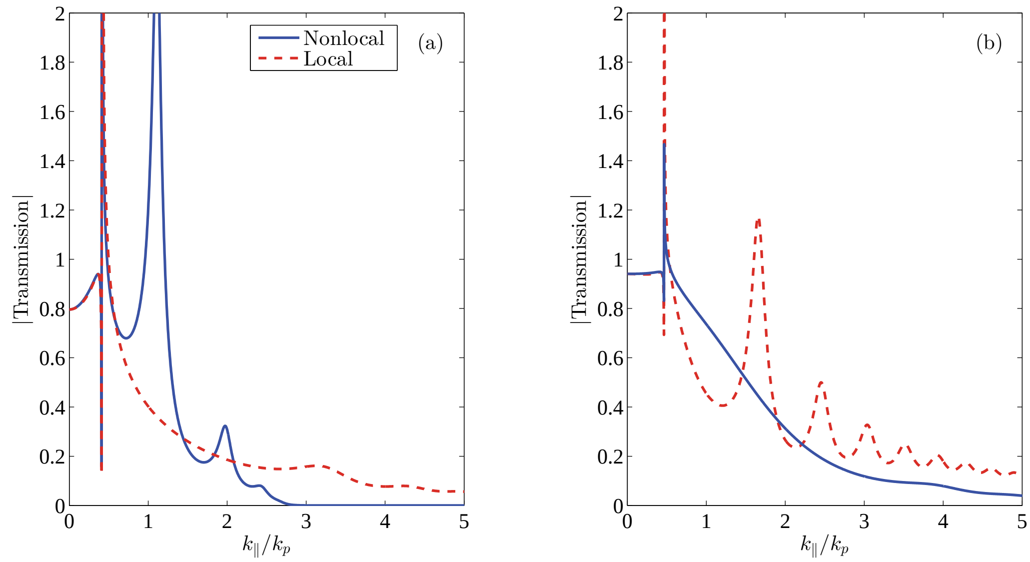

Figure 1. Transmission through the hyperbolic metamaterial as described in the main text, in panel (a) for $\omega = \omega_{\rm res}^{\rm loc} = 0.41\omega_p$; in panel (b) for $\omega = \omega_{\rm res}^{\rm nloc} = 0.465\omega_p$. At both frequencies, there are considerable differences between the predictions based on local and on nonlocal response.

can be found in Ref. 12. In Fig. 1 only the finite Drude damping prevents these resonances from being infinitely narrow and infinitely high.

The final point that we would like to make about Fig. 1 is that nonlocal response does not "just" shift the optimal resonance frequency to the blue: the local-response transmission curve at $\omega = \omega_{\rm res}^{\rm loc}$ in panel (a) is clearly different from the nonlocal transmission curve at $\omega = \omega_{\rm res}^{\rm nloc}$ in panel (b). This will have the further consequence, as we will see, that both theories predict different superlens images at their respective optimal operating frequencies.

2.4 Near-field images of local versus nonlocal superlenses

Now that we know how well the various plane-wave components can be transmitted through the metamaterial, we next study how in combination they can produce a nearly perfect near-field image. As the source we consider a line source proportional to $\delta(y)$ parallel to and positioned at a distance of only 1 nm away from the metamaterial lens. A large portion of the waves emanating from the source propagates all the way through the 36 unit cells, to form an image on the back plane of the lens.

Figure 2(a-d) displays such images for four different operating frequencies in our range of interest, namely from $\omega_{\rm res}^{\rm loc}$ to $\omega_{\rm res}^{\rm nloc}$. For every frequency, we compare the image as predicted by local-response as compared to hydrodynamic nonlocal response theory. Now the perfect image of an infinitely thin line source would again be an infinitely thin line source. The superlens does not quite achieve that, but at the right operating frequency the quality of the image comes close. For example, in panel (a) at $\omega_{\rm res}^{\rm loc}$ we see a subwavelength local-response image with a single dominant intensity peak with a FWHM width of less than half a plasma wavelength. In panel (d) nonlocal-response theory predicts a similar image, but at $\omega_{\rm res}^{\rm nloc}$ instead. Thus the superlens still performs as a superlens when taking nonlocal response into account. (We found an analogous result for refractive-index sensors.[13]) Panels (b) and (c) illustrate that the local-response image broadens while the nonlocal-response image narrows as we increase the operating frequency from $\omega_{\rm res}^{\rm loc}$ to $\omega_{\rm res}^{\rm nloc}$.

If we compare more closely the optimal local image in panel (a) to the optimal nonlocal image in panel (d), then we see that the nonlocal image has smaller side peaks and hence is more localized and therefore a better image of the line source than the local one. This is another important illustration of the fact that nonlocal

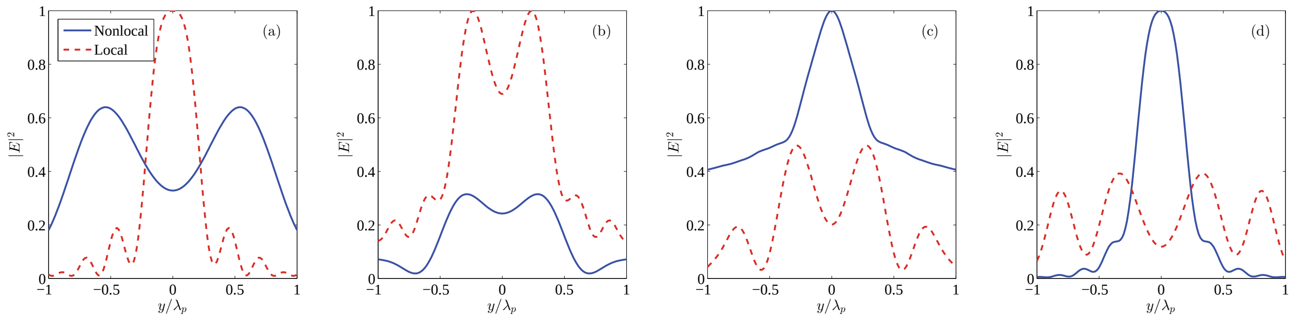

Figure 2. Field intensities at the back end of the hyperbolic superlens, calculated both with local and with nonlocal response, for different operating frequencies in the interval between $\omega_{\rm res}^{\rm loc}$ and $\omega_{\rm res}^{\rm nloc}$. Panel (a): $\omega = \omega_{\rm res}^{\rm loc} = 0.41\omega_p$, the ideal operating frequency for local response; panel (b): $\omega = 0.43\omega_p$; panel (c): $\omega = 0.45\omega_p$; panel (d): $\omega = \omega_{\rm res}^{\rm nloc} = 0.465\omega_p$, the ideal operating frequency for nonlocal response.

response does not "just" shift the optimal operating frequency, but has interesting other physical effects too. That nonlocal response predicts the sharper image in Fig. 2 is also interesting because it is counterintuitive: nonlocal response is otherwise better known for smearing out sharp features, for example field enhancements close to almost touching plasmonic dimers,[14] near corrugated surfaces,[15] or at the surface of nanowires with realistic surface smoothness.[16]

2.5 Spontaneous emission inside hyperbolic metamaterials

After transmission through hyperbolic metamaterials, let us also briefly discuss spontaneous emission of quantum emitters embedded inside them. Hyperbolic metamaterials have been proposed as simple structures by which broadband enhancement of spontaneous-emission rates could be realized,[17] with possible applications in the field of quantum plasmonics.[18,19] Actually in the limit of vanishing unit-cell size, the spontaneous-emission rate in local-response theory even diverges for hyperbolic metamaterials, since on a hyperbola corresponding to a fixed emission frequency there are infinitely many states $(k_\parallel, k_\perp)$, with no bounds on $k_\parallel$ or $k_\perp$, that contribute to the spontaneous-emission rate. There is no such broadband supersingularity for the more common elliptically shaped dispersion relations, where there is also a continuum of states but the wave vectors on iso-frequency contours are bounded.

Spontaneous emission in hyperbolic media has been measured recently, in layered[20] and in wire[21] metamaterials. Considerable enhancement of the rates was found. Infinitely high spontaneous-emission rates were not observed, though. There will always be something that keeps rates finite. The current main limiting factor is the size of the unit cell, other factors being the finite size of the quantum emitter and damping.[17,22]

It is an interesting open question how strongly spontaneous-emission rates can be enhanced in realistic hyperbolic metamaterials. To obtain high emission rates it is advantageous to fabricate multilayer metamaterials with very thin yet very smooth surfaces. In this quest for high emission rates, we expect that unit cells will be made smaller and spontaneous-emission rates will go up, and that at some point in the near future nonlocal effects will start to play a role in metamaterials, certainly nonlocal blueshifts but perhaps also effects on spontaneous-emission rates.

What influence will nonlocal response have on spontaneous-emission rates? In Ref. 1 we studied spontaneous-emission rates in multilayered hyperbolic metamaterials. Our main finding is that these rates will always be finite, even in the limit of infinitely small unit cells. This limit provides the upper bound for the enhancement. However, the upper bound for the enhancement of the local density of states (LDOS), which is proportional to $\omega^2/v_{\rm F}^3$, is large, namely of order 10^7 for most metals. So in practice there is ample room for enhancing spontaneous-emission rates before this fundamental limit set by nonlocal response is reached.

3. QUANTUM OPTICAL LIMIT

Let us now consider unit cells that are much smaller than an optical wavelength so that an effective-medium description is possible, but let us not make the unit cells so small that nonlocal effects play a role. Then the common assumption is that for external light sources at optical wavelengths the medium can be described fully in terms of its effective index and overall geometry. However, we have recently shown that this is not true in quantum optics: two metamaterials with identical shape and identical effective index can have different optical properties.[3] Identical quantum states of light entering these metamaterials will give rise to different quantum states of light after transmission or reflection, so these metamaterials that are effectively the same in classical optics can be told apart in quantum optics. We discuss some elements of quantum electrodynamics in dielectric media in Sec. 3.1, describe the experiment to tell apart two metamaterials with the same effective index in Sec. 3.2, and discuss our accurate effective-medium theory in quantum optics in Sec. 3.3.

3.1 Quantum noise in Maxwell's equations for lossy and amplifying media

In quantum electrodynamics, the electromagnetic fields become field operators. Without gain or loss, the quantum field operators satisfy the same wave equation as the corresponding classical fields. By contrast, the wave equation for the field operators in a lossy or amplifying medium differs from that in classical electrodynamics. For example, in a one-dimensional situation, the wave equation for the vector potential can be written as[23]

$$\left[\frac{\partial^2}{\partial x^2} + \frac{\omega^2}{c^2}\varepsilon(x,\omega)\right]\hat{A}(x,\omega) = \hat{j}(x,\omega), \tag{2}$$

where the current operator $\hat{j}(x,\omega)$ is proportional to $\sqrt{|\varepsilon_{\mathrm{I}}(x,\omega)|}$, and $\varepsilon_{\mathrm{I}}(x,\omega)$ is the imaginary part of the dielectric function describing either loss or gain.[3] Clearly, in the limit of no loss or gain, $\hat{j}$ vanishes and Eq. (2) becomes the classical wave equation for the vector potential in a dispersive and spatially inhomogeneous medium. The operator $\hat{j}(x,\omega)$ describes the quantum noise that is associated with gain or loss.[3,23–27]

That loss or gain is associated with quantum noise can be derived from the so-called fluctuation-dissipation theorem: the quantum noise is the fluctuation that is associated with loss or gain. One can also understand the need for a driving term on the right-hand side of the wave equation from the following argument: in classical optics, a homogeneous lossy medium has monochromatic damped plane waves as solutions. Both the vector potential and the electric-field operator associated with this damped plane wave decay. In quantum optics this would not be compatible with the requirement that the equal-time commutator between the electric field $\hat{E}(x,t)$ and the vector potential $\hat{A}(x',t)$ is constant in time, proportional to $\delta(x-x')$, and with the same value in all of space. Only when solving Eq. (2) by taking the quantum-noise driving term into account will equal-time commutators of field operators stay constant in time for media with gain or loss.

3.2 Telling apart metamaterials with the same effective index

We would like to investigate how metamaterials could be used in quantum optics, and started with the basic question whether metamaterials can be described by their effective index $\varepsilon_{\mathrm{eff}}$ in quantum optics.[3] Now since metals are an important constituent of many metamaterials, loss should be taken into account in metamaterials. An important branch of plasmonics is active plasmonics where one actively modifies the material properties.[28] An important example of active plasmonics is active loss compensation. In loss-compensated metamaterials, the loss in one part of the unit cell is compensated for by the gain elsewhere.[29] Such effectively lossless metamaterials would have a better performance as near-field lenses.[30]

For quantum optics it is is important to keep in mind that there is no lossy metamaterial without noise.[31] The existence of quantum noise does not immediately imply that the effective index no longer accurately describes the medium in quantum optics. For example, an "effective quantum noise" could perhaps be well described by the effective index. But if loss is compensated by gain, is an effectively lossless metamaterial also effectively free of quantum noise?

To answer this, we studied three types of multilayer metamaterials: one with two types of lossy media, one with two types of gain media, and finally a loss-compensated metamaterial where a lossy metal layer alternates with a dielectric layers with embedded dye molecules. The latter exhibits linear gain. For detailed descriptions

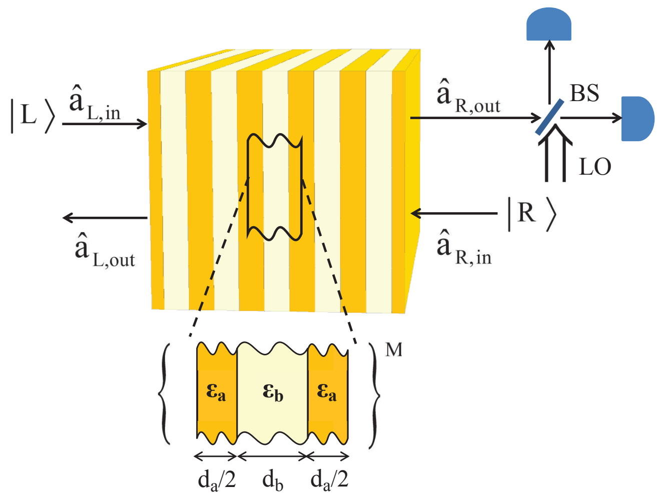

Figure 3. Sketch of the quantum optical experiment that can be used to detect at optical wavelengths different optical properties of two metamaterials with the same effective index but different amounts of loss compensation. Squeezed states of light enter the layered metamaterial from both sides. The quantum state of light that exits the metamaterial on the right enters a balanced homodyne detector, where its noise properties (variances, uncertainties) are measured.

how to create amplification in metamaterials we refer to Refs. 32, 33. For each of the three types of layered metamaterials, we shine squeezed light onto both of its sides, and analyze the output state of light using balanced homodyne detection, as sketched in Fig. 3. This is a standard experiment in quantum optics to measure the noise properties of the light (its variance, uncertainty). First we compared the variance of the electric field that leaves the metamaterial using an exact multilayer theory with the variance based on an effective-index theory. In this quantum optical effective-index theory, the effective quantum noise of a metamaterial can be described entirely in terms of its effective dielectric function. What this theory boils down to is to replace in the wave equation Eq. (2) the spatially varying dielectric function $\varepsilon(x,\omega)$ of the metamaterial by its effectively homogenized value $\varepsilon_{\rm eff}(\omega)$, and to replace the quantum noise by an effective quantum noise $\hat{j}_{\rm eff}(x,\omega)$ that is proportional to $\sqrt{|\varepsilon_{\rm eff,I}(\omega)|}$. We found that for the gain-gain and for the loss-loss metamaterials this is an accurate effective description of the metamaterial in quantum optics.

However, for loss-compensated metamaterials we find significant differences between the variance of the field according to the exact multilayer theory and according to the effective-index theory, as illustrated in Fig. 4. The blue solid line is the exact multilayer theory for the loss-compensated metamaterial, while the red dotted curve describes the variance of a homogeneous medium with the same effective index. Thus with balanced homodyne detection we can tell apart two metamaterials in quantum optics even if their effective indices are the same, if they differ by the amount of loss compensation.

3.3 Quantum optical effective-medium theory

Loss compensation is a successful strategy in active plasmonics and it also works in quantum optics: a lossy dielectric response and an amplifying response can cancel each other so as to produce an (almost) lossless effective response. Nevertheless the quantum optical effective-index theory breaks down in case of loss compensation, as illustrated in Fig. 4. The reason is that by taking an effective quantum noise $\hat{j}_{\rm eff}(x,\omega)$ proportional to $\sqrt{|\varepsilon_{\rm eff,I}(\omega)|}$, this theory assumes that quantum noise associated with loss can be compensated for by quantum noise associated with gain, just like loss is compensated by gain.

The exact multilayer theory does not exhibit such a quantum noise compensation: the noise sources add up rather than compensate each other. From the exact multilayer theory we distilled a quantum optical effective-medium theory that features an additional effective parameter: besides the usual effective index, an effective noise photon distribution is defined that grows when more loss is compensated by more gain. More details are

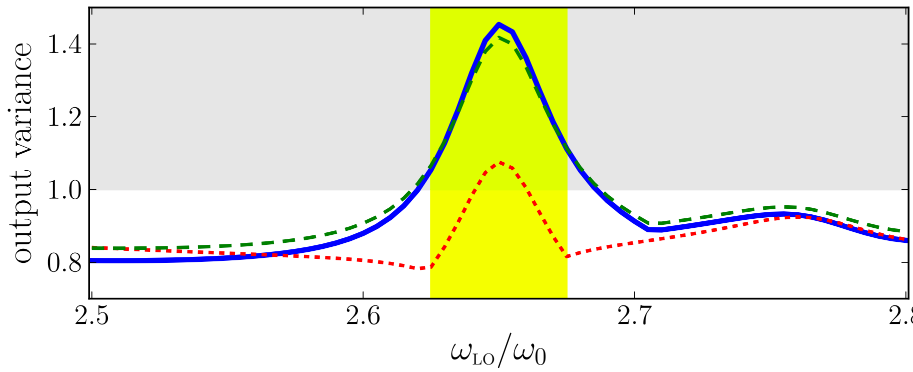

Figure 4. Output variance of a metamaterial consisting of 6 nm thin films of silver alternating with 60 nm thin dielectric layers doped with dye molecules that are pumped and produce linear gain (details in Ref. 3). Blue solid line: exact multilayer theory for propagation of quantum light. Red dotted curve: quantum optical effective-index theory; green dashed curve: quantum optical effective-medium theory of Ref. 3. The yellow-green frequency interval corresponds to net gain, outside of this interval the medium is effectively lossy. A variance below unity corresponds to squeezed light, otherwise the light is not squeezed.

given in our Ref. 3. This quantum optical efective-medium theory is shown in Fig. 4 as the green dashed curve, and notice that the agreement with the exact theory is very good.

4. CONCLUSIONS AND OUTLOOK

In this paper we have reviewed two fundamental limits for metamaterials. In the small-scale limit we found that nonlocal response starts to play a role when making characteristic lengths of the metal constituents of the unit cell of order smaller than 10 nm.[1] For quantum emitters embedded into hyperbolic metamaterials, spontaneous-emission rates can differ drastically from what would be expected on the basis of an effective-medium theory. In local-response theories, the rate even diverges in the limit of infinitely small unit cells. We find that effective-medium parameters are to be computed differently when entering the nonlocal-response regime, and that spontaneous-emission rates enhancements can be very large but stay finite even in the limit of infinitely small unit cells. We expect that our theoretical results for metamaterials in the small-scale limit will soon become relevant for a next-generation class of layered hyperbolic metamaterials, with layers of well-defined few-nanometer thickness.

If one makes a hyperbolic lens from a hyperbolic metamaterial of finite thickness, then objects in the near field of the lens are imaged nearly perfectly on the other side of the lens, at least for the optimal resonance frequency of the metamaterial. We find that nonlocal response blueshifts this optimal frequency, which is an important result when designing and optimizing hyperbolic superlenses.[2] Interestingly, sometimes nonlocal response predicts the better imaging when comparing local and nonlocal response at their respective optimal frequencies.

In the quantum optical limit, it is not enough to re-evaluate the values of the effective parameters. Rather, an additional effective parameter is needed that quantifies the amount of loss compensation that goes on inside the metamaterial. This information beyond the effective index enters our quantum optical effective-medium theory.[3]

ACKNOWLEDGMENTS

This work was financially supported by an H. C. Ørsted Fellowship (W.Y.), and by the Otto Mønsted Foundation (M.W.). The Center for Nanostructured Graphene is sponsored by the Danish National Research Foundation, Project DNRF58.

REFERENCES

[1] Yan, W., Wubs, M., and Mortensen, N. A., "Hyperbolic metamaterials: nonlocal response regularizes broadband supersingularity," *Phys. Rev. B* **86**, 205429 (2012).

[2] Yan, W., Mortensen, N. A., and Wubs, M., "Hyperbolic metamaterial lens with hydrodynamical nonlocal response," *Opt. Express* **21**, 15026 (2013).

[3] Amooghorban, E., Mortensen, N. A., and Wubs, M., "Quantum optical effective-medium theory for loss-compensated metamaterials," *Phys. Rev. Lett.* **110**, 153602 (2013).

[4] Sersic, I., Tuambilangana, C., Kampfrath, T., and Koenderink, A. F., "Magnetoelectric point scattering theory for metamaterial scatterers," *Phys. Rev. B* **83**, 245102 (2011).

[5] Poddubny, A. N., Belov, P. A., Ginzburg, P., Zayats, A. V., and Kivshar, Y. S., "Microscopic model of Purcell enhancement in hyperbolic metamaterials," *Phys. Rev. B* **86**, 035148 (2012).

[6] Boardman, A. D., [*Electromagnetic Surface Modes*], John Wiley and Sons, Chichester (1982).

[7] Raza, S., Toscano, G., Jauho, A.-P., Wubs, M., and Mortensen, N. A., "Unusual resonances in nanoplasmonic structures due to nonlocal response," *Phys. Rev. B* **84**, 121412(R) (2011).

[8] Yao, J., Yang, X., Yin, X., Bartal, G., and Zhang, X., "Three-dimensional nanometer-scale optical cavities of indefinite medium," *PNAS* **108**, 11327 (2011).

[9] Scholl, J. A., Koh, A. L., and Dionne, J. A., "Quantum plasmon resonances in individual metallic nanoparticles," *Nature* **483**, 421 (2012).

[10] Raza, S., Stenger, N., Kadkhodazadeh, S., Fischer, S. V., Kostesha, N., Jauho, A.-P., Burrows, A., Wubs, M., and Mortensen, N. A., "Blueshift of the surface plasmon resonance in silver nanoparticles studied with EELS," *Nanophotonics* **2**, 131 (2013).

[11] Formica, N., Gosh, D. S., Carrilero, A., Chen, T. L., Simpson, R. E., and Pruneri, V., "Ultrastable and atomically smooth ultrathin silver films grown on a copper seed layer," *ACS Appl. Mater. Interfaces* **5**, 3048 (2013).

[12] Christensen, J. and García de Abajo, F. J., "Slow plasmonic slab waveguide as a superlens for visible light," *Phys. Rev. B* **82**, 161103 (2010).

[13] Raza, S., Toscano, G., Jauho, A.-P., Mortensen, N. A., and Wubs, M., "Refractive-index sensing with ultrathin plasmonic nanotubes," *Plasmonics* **8**, 193 (2013).

[14] Toscano, G., Raza, S., Jauho, A.-P., Mortensen, N. A., and Wubs, M., "Modified field enhancement and extinction by plasmonic nanowire dimers due to nonlocal response," *Opt. Express* **20**, 4176 (2012).

[15] Toscano, G., Raza, S., Xiao, S., Wubs, M., Jauho, A.-P., Bozhevolnyi, S. I., and Mortensen, N. A., "Surface-enhanced Raman spectroscopy: nonlocal limitations," *Opt. Lett.* **37**, 2538 (2013).

[16] Wiener, A., Fernández-Domínguez, A. I., Horsfield, A. P., Pendry, J. B., and Maier, S. A., "Nonlocal effects in the nanofocusing performance of plasmonic tips," *Nano Lett.* **12**, 3308 (2012).

[17] Jacob, Z., Smolyaninov, I. I., and Narimanov, E. E., "Broadband Purcell effect: Radiative decay engineering with metamaterials," *Appl. Phys. Lett.* **100**, 181105 (2012).

[18] Cortes, C. L., Newman, W., Molesky, S., and Jacob, Z., "Quantum nanophotonics using hyperbolic metamaterials," *J. Opt.* **14**, 063001 (2012).

[19] Tame, M. S., McEnery, K. R., Özdemir, Ş. K., Lee, J., Maier, S. A., and Kim, M. S., "Quantum plasmonics," *Nature Phys.* **9**, 329 (2013).

[20] Tumkur, T., Zhu, G., Black, P., Barnakov, Y. A., Bonner, C. E., and Noginov, M. A., "Control of spontaneous emission in a volume of functionalized hyperbolic metamaterial," *Appl. Phys. Lett.* **99**, 151115 (2010).

[21] Noginov, M. A., Li, H., Barnakov, Y. A., Dryden, D., Nataraj, G., Zhu, G., Bonner, C. E., Mayy, M., Jacob, Z., and Narimanov, E. E., "Controlling spontaneous emission with metamaterials," *Opt. Lett.* **35**, 1863 (2010).

[22] Poddubny, A. N., Belov, P. A., and Kivshar, Y. S., "Spontaneous radiation of a finite-size dipole emitter in hyperbolic media," *Phys. Rev. A* **84**, 023807 (2011).

[23] Gruner, T. and Welsch, D.-G., "Quantum-optical input-output relations for dispersive and lossy multilayer dielectric plates," *Phys. Rev. A* **54**, 1661 (1996).

[24] Matloob, R., Loudon, R., Artoni, M., Barnett, S. M., and Jeffers, J., "Electromagnetic field quantization in amplifying dielectrics," *Phys. Rev. A* **55**, 1623 (1997).

[25] Wubs, M. and Suttorp, L. G., "Transient QED effects in absorbing dielectrics," *Phys. Rev. A* **63**, 043809 (2001).

[26] Suttorp, L. G. and Wubs, M., "Field quantization in inhomogeneous absorptive dielectrics," *Phys. Rev. A* **70**, 013816 (2004).

[27] Amooghorban, E., Wubs, M., Mortensen, N. A., and Kheirandish, F., "Casimir forces in multilayer magnetodielectrics with both gain and loss," *Phys. Rev. A* **84**, 013806 (2011).

[28] Boardman, A. D., Grimalsky, V. V., Kivshar, Y. S., Koshevaya, S. V., Lapine, M., Litchinitser, N. M., Malnev, V. N., Noginov, M., Rapoport, Y. G., and Shalaev, V. M., "Active and tunable metamaterials," *Laser Photonics Rev.* **5**, 287 (2011).

[29] Xiao, S., Drachev, V. P., Kildishev, A. V., Ni, X., Chettiar, U. K., Yuan, H.-K., and Shalaev, V. M., "Loss-free and active optical negative-index metamaterials," *Nature* **466**, 735 (2010).

[30] Ramakrishna, S. A. and Pendry, J. B., "Removal of absorption and increase in resolution in a near-field lens via optical gain," *Phys. Rev. B* **67**, 201101 (2003).

[31] Syms, R. R. A., Sydoruk, O., and Solymar, L., "Lossy metamaterials: no effective medium properties without noise," *Phys. Rev. B* **84**, 235150 (2011).

[32] Berini, P. and De Leon, I., "Surface plasmon polariton amplifiers and lasers," *Nature Phot.* **6**, 16 (2012).

[33] Hess, O. and Tsakmakidis, K. L., "Metamaterials with quantum gain," *Science* **339**, 654 (2013).