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Reduced reflection from roughened hyperbolic metamaterial

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Abstract: We show that roughened surfaces of hyperbolic metamaterials scatter light preferentially inside the media, resulting in a very low reflectance. This phenomenon of fundamental importance, demonstrated experimentally in arrays of silver nanowires grown in alumina membranes, is consistent with a broad-band singularity in the density of photonic states. It paves the road to a variety of applications ranging from the stealth technology to high-efficiency solar cells and photodetectors.

OCIS codes: (160.3918) Metamaterials; (160.1190) Anisotropic optical materials.

References and links

1. V. G. Veselago, "The electrodynamics of substances with simultaneously negative values of permittivity and permeability," Sov. Phys. Usp. **10**, 509–514 (1968).
2. J. B. Pendry, "Negative refraction makes a perfect lens," Phys. Rev. Lett. **85**(18), 3966–3969 (2000).
3. J. B. Pendry, D. Schurig, and D. R. Smith, "Controlling electromagnetic fields," Science **312**(5781), 1780–1782 (2006).
4. W. Cai, U. K. Chettiar, A. V. Kildishev, and V. M. Shalaev, "Optical cloaking with metamaterials," Nat. Photonics **1**(4), 224–227 (2007).
5. T. Ergin, N. Stenger, P. Brenner, J. B. Pendry, and M. Wegener, "Three-dimensional invisibility cloak at optical wavelengths," Science **328**(5976), 337–339 (2010).
6. Z. Jacob, L. V. Alekseyev, and E. Narimanov, "Optical Hyperlens: Far-field imaging beyond the diffraction limit," Opt. Express **14**(18), 8247–8256 (2006).
7. A. Salandrino and N. Engheta, "Far-field subdiffraction optical microscopy using metamaterial crystals: Theory and simulations," Phys. Rev. B **74**(7), 075103 (2006).
8. Z. Liu, H. Lee, Y. Xiong, C. Sun, and X. Zhang, "Far-field optical hyperlens magnifying sub-diffraction-limited objects," Science **315**(5819), 1686 (2007).
9. I. I. Smolyaninov, Y. J. Hung, and C. C. Davis, "Magnifying superlens in the visible frequency range," Science **315**(5819), 1699–1701 (2007).
10. D. R. Smith and D. Schurig, "Electromagnetic wave propagation in media with indefinite permittivity and permeability tensors," Phys. Rev. Lett. **90**(7), 077405 (2003).
11. Z. Jacob, I. I. Smolyaninov, and E. E. Narimanov, "Broadband Purcell effect: Radiative decay engineering with metamaterials," Appl. Phys. Lett. **100**, 181105 (2012).
12. I. I. Smolyaninov and E. E. Narimanov, "Metric signature transitions in optical metamaterials," Phys. Rev. Lett. **105**(6), 067402 (2010).
13. W. Dickson, G. A. Wurtz, P. Evans, D. O'Connor, R. Atkinson, R. Pollard, and A. V. Zayats, "Dielectric-loaded plasmonic nanoantenna arrays: a metamaterial with tunable optical properties," Phys. Rev. B **76**(11), 115411 (2007).
14. J. Yao, Z. Liu, Y. Liu, Y. Wang, C. Sun, G. Bartal, A. M. Stacy, and X. Zhang, "Optical negative refraction in bulk metamaterials of nanowires," Science **321**(5891), 930 (2008).
15. M. A. Noginov, Yu. A. Barnakov, G. Zhu, T. Tumkur, H. Li, and E. E. Narimanov, "Bulk photonic metamaterial with hyperbolic dispersion," Appl. Phys. Lett. **94**(15), 151105 (2009).
16. A. J. Hoffman, L. Alekseyev, S. S. Howard, K. J. Franz, D. Wasserman, V. A. Podolskiy, E. E. Narimanov, D. L. Sivco, and C. Gmachl, "Negative refraction in semiconductor metamaterials," Nat. Mater. **6**(12), 946–950 (2007).
17. H. N. S. Krishnamoorthy, Z. Jacob, E. Narimanov, I. Kretzschmar, and V. M. Menon, "Metamaterial based broadband engineering of quantum dot spontaneous emission," arXiv:0912.2454v1 [physics.optics] (2009).
18. Z. Jacob, J. Y. Kim, G. V. Naik, A. Boltasseva, E. E. Narimanov, and V. M. Shalaev, "Engineering photonic density of states using metamaterials," Appl. Phys. B **100**(1), 215–218 (2010).
19. T. Tumkur, G. Zhu, P. Black, Yu. A. Barnakov, C. E. Bonner, and M. A. Noginov, "Control of spontaneous emission in a volume of functionalized hyperbolic metamaterial," Appl. Phys. Lett. **99**(15), 151115 (2011).

20. G. V. Naik, J. Liu, A. V. Kildishev, V. M. Shalaev, and A. Boltasseva, "Demonstration of Al:ZnO as a plasmonic component for near-infrared metamaterials," Proc. Natl. Acad. Sci. U.S.A. **109**(23), 8834–8838 (2012).
21. L. D. Landau and E. M. Lifshits, *Quantum Mechanics* (Butterworth Heinemann, 1999).
22. M. A. Noginov, H. Li, Y. A. Barnakov, D. Dryden, G. Nataraj, G. Zhu, C. E. Bonner, M. Mayy, Z. Jacob, and E. E. Narimanov, "Controlling spontaneous emission with metamaterials," Opt. Lett. **35**(11), 1863–1865 (2010).
23. J. Kim, V. P. Drachev, Z. Jacob, G. V. Naik, A. Boltasseva, E. E. Narimanov, and V. M. Shalaev, "Improving the radiative decay rate for dye molecules with hyperbolic metamaterials," Opt. Express **20**(7), 8100–8116 (2012).
24. H. N. S. Krishnamoorthy, Z. Jacob, E. Narimanov, I. Kretzschmar, and V. M. Menon, "Topological Transitions in Metamaterials," Science **336**(6078), 205–209 (2012).
25. E. E. Narimanov, M. A. Noginov, H. Li, and Yu. Barnakov, "Darker than Black: Radiation-absorbing Metamaterial," presented at the Quantum Electronics and Laser Science Conference (QELS), San Jose, CA, May 16, 2010, QELS Postdeadline Session I (QPDA).
26. E. M. Purcell, "Spontaneous emission probabilities at radiofrequencies," Phys. Rev. **69**, 681 (1946).
27. L. V. Alekseyev and E. E. Narimanov, "Radiative decay engineering in metamaterials" in *Tutorials in Metamaterials*, M. A. Noginov and V. A. Podolskiy, ed. (CRC Press, Taylor&Francis Group, 2011).
28. E. Kretschmann and E. Kröger, "Reflection and transmission of light by a rough surface, including results for surface-plasmon effects," J. Opt. Soc. Am. **65**(2), 150–154 (1975).
29. R. Orlowski and H. Raether, "The total reflection of light at smooth and rough silver films and surface plasmons," Surf. Sci. **54**(2), 303–308 (1976).
30. S.-K. Kim, H.-S. Ee, W. Choi, S.-H. Kwon, J.-H. Kang, Y.-H. Kim, H. Kwon, and H.-G. Park, "Surface-plasmon-induced light absorption on a rough silver surface," Appl. Phys. Lett. **98**(1), 011109 (2011).
31. M. A. Noginov, S. U. Egarievwe, H. J. Caulfield, N. Noginova, M. Curley, P. Venkateswarlu, A. Williams, and J. Paitz, "Diffusion and pseudo-phase-conjugation effects in coherent backscattering from $Nd_{0.5}La_{0.5}Al_3(BO_3)_4$ ceramic," Opt. Mater. **10**(1), 1–7 (1998).
32. H. K. Raut, V. A. Ganesh, A. S. Nair, and S. Ramakrishna, "Anti-reflective coatings: A critical, in-depth review," Energy Environ. Sci. **4**(10), 3779–3804 (2011).

1. Introduction: metamaterials with hyperbolic dispersion and high density of photonic states

Metamaterials – engineered composite materials with rationally designed subwavelength inclusions – have unique responses to electromagnetic fields, which are unavailable in conventional media. Their fascinating properties and applications include negative index of refraction [1,2], invisibility cloaking [3–5], and sub-diffraction imaging and focusing [2,6–9].

The research in *hyperbolic metamaterials* (also known as *indefinite media* [10]), originally stimulated by tantalizing possibilities offered by the absence of diffraction limit in a hyperlens [6–9], has uncovered a number of novel effects resulting from a broadband singular behavior of the density of photonic states in these materials [11,12].

Known hyperbolic metamaterials include arrays of metallic nanowires grown in dielectric membranes [13–15] as well as lamellar metal-dielectric or semiconductor structures [16–20]. These uniaxial materials are highly anisotropic, with the dielectric permittivity components of opposite signs in two perpendicular directions. An isofrequency dispersion curve for extraordinary wave in a uniaxial (meta)material is given by

$$\frac{k_y^2}{\varepsilon_{x,z}}+\frac{k_x^2+k_z^2}{\varepsilon_y}=\left(\frac{\omega}{c}\right)^2, \qquad (1)$$

where k_i are the components of the wave-vector ($i = x,y,z$; the optical axis is in the y direction), ε_i are the values of dielectric permittivity, ω is the angular frequency, and c is the speed of light.

The nature of the "super-singularity" in hyperbolic metamaterials can be understood from a visual representation of the density of states in terms of the phase space volume enclosed by two surfaces corresponding to different values of the light frequency. In dielectric media, electric permittivities are positive, $\varepsilon_i>0$, and the dispersion law describes an ellipsoid in the k-space (sphere if $\varepsilon_x = \varepsilon_y = \varepsilon_z$). The phase space volume enclosed between two such surfaces (Fig. 1(a), left) is then finite, corresponding to a finite density of photonic states. However,

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when one of the components of the dielectric permittivity tensor is negative, Eq. (1) describes a hyperboloid in the phase space. As a result, the phase space volume between two such hyperboloids (corresponding to different values of frequency) is nominally infinite (Fig. 1(a), right), leading to a nominally infinite density of photonic states. While there are many mechanisms leading to a singularity in the density of photonic states, this one is unique – as it results in a very large density of states for every frequency where different components of the dielectric permittivity have opposite signs. (Note that in actual hyperbolic metamaterials, the maximal length of the wave-vector is limited, among other reasons, by characteristic dimensions of the metamaterial's nanostructure. Although this makes the density of photonic states not infinite, it remains to be at least one order of magnitude larger than that in vacuum.)

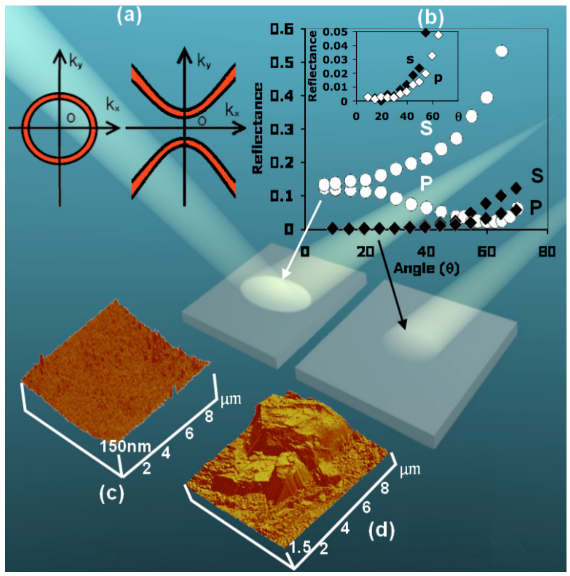

Fig. 1. Dramatic reduction of reflectance off corrugated hyperbolic metamaterial. Panel (a): the phase space "volume" enclosed by two different surfaces of constant frequency, in the cases when components of the dielectric permittivity tensor are all positive (left) and have opposite signs (right). Panel (b): angular reflectance profiles measured on untreated (circles) and roughened (diamonds) parts of the same membrane sample in s-polarization and p-polarization. Inset: reflectance profiles in the corrugated sample (same as in main panel (b), zoomed). Panels (c) and (d): topography profiles of the untreated (c) and corrugated (d) samples.

As the scattering rate $W_{k\to k'}$, in the spirit of the Fermi Golden Rule [21], is proportional to the density of states $\rho(\omega)$,

$$W_{k\to k'} \propto \delta(\omega_k - \omega_{k'})\,\rho(\omega), \tag{2}$$

the super-singularity in $\rho(\omega)$ is expected to enhance a variety of phenomena, which depend on the density of final photonic states. One of such processes is a spontaneous emission, which enhancement in vicinity of hyperbolic metamaterials has been theoretically predicted (by E. E. Narimanov and co-authors [11]) and experimentally proven by several research groups [17–19, 22–25].

2. Experimental observation of reduced reflectance

Hyperbolic metamaterials, which we used in our experiments, were based on arrays of 35 nm-thick silver nanowires grown (using an electroplating technique) in 1cm x 1cm x 51μm anodic alumina membranes [15]. Nanowires were oriented perpendicularly to the membrane's surface, and the nominal filling factor of silver was ~12%, Fig. 2(a).

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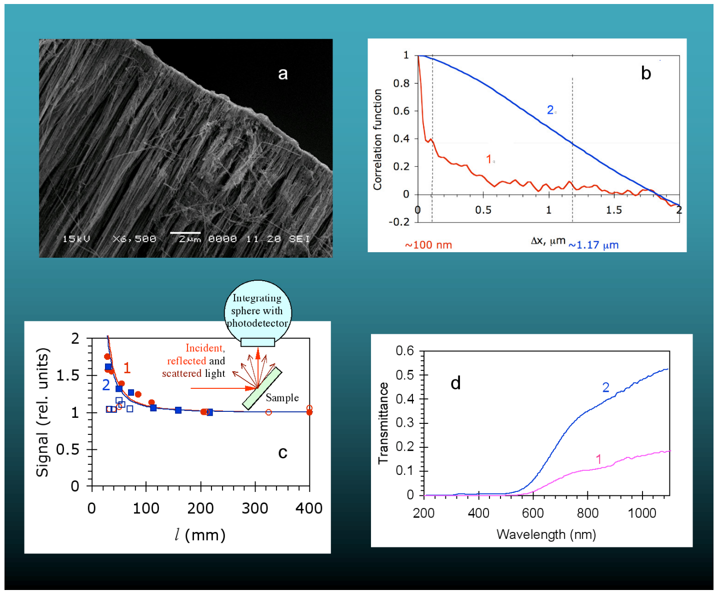

Fig. 2. Panel (a): Scanning Electron Microscope (SEM) image of silver nanowires grown in alumina membrane (cleaved and etched sidewall). Panel (b): Correlation functions characterizing surface roughness of the untreated (1) and corrugated (2) samples. Panel (c): Reflected and scattered light intensities measured as a function of the sample-to-detector distance *l* with a wide-aperture detector in the untreated (open characters) and corrugated (closed characters) samples; circles – s-polarization, squares – p-polarization. The measurements were done at the 41° reflectance angle. Red (1) and blue solid (2) lines are the fittings of the experimental data (in corrugated samples) with the Lambertian model; trace 1 – s polarization, trace 2 – p polarization. Inset: Schematic of the experimental setup. Panel (d): Transmittance spectra of the untreated (trace 1) and corrugated (trace 2) samples measured at near-normal incidence.

The fabricated uniaxial metamaterial reported here had hyperbolic dispersion (negative dielectric permittivity in the direction perpendicular to the membrane's surface, $\varepsilon_\perp$, and positive dielectric permittivity in the direction parallel to the membrane, $\varepsilon_{||}$) in the near-infrared spectral range. From the measurements of the angular dependence of reflectance in s and p polarizations, Fig. 1(b), we have deduced the following materials parameters at $\lambda = 873$ nm: $\varepsilon_\perp' = -0.15$, $\varepsilon_\perp'' = 1.07$, $\varepsilon_{||}' = 4.99$, and $\varepsilon_{||}'' = 0.02$. The measurements and the retrieval procedure are discussed in detail in Ref [15]. The roughness of the untreated sample, measured with the atomic force microscope (AFM), was equal to rms = 40 nm, Fig. 1(c). The lateral correlation length, at which the correlation function

$$C\left(r_m, r_n\right) = \frac{\sum_m \sum_n \left(z_m\left(r_m\right) - \bar{z}\right)\left(z_n\left(r_n\right) - \bar{z}\right)}{\sum_m \left(z_m\left(r_m\right) - \bar{z}\right)^2}$$

decreased from 1 to 1/e, was equal to ~100 nm, Fig. 2(b). (Here z_i is the height of the topology profile in the position r_i and $\bar{z}$ is the mean height).

The surface of the sample was then roughened by grinding it with 3 μm Al_2O_3 polishing powder, resulting in the roughness equal to rms = 600 nm [Fig. 1(d)] and the correlation length equal to 1.17 μm, Fig. 2b. The reflectance measurements were repeated in the grinded sample, resulting in much smaller intensities of reflected light, especially at small incidence angles, Fig. 1(b). Although the reduction of reflectance was observed in both polarizations, it was stronger in p polarization than in s polarization, inset of Fig. 1(b).

As corrugated samples produced not only specular reflection but also diffused scattering, we have evaluated the ratio of reflected and scattered light intensities by measuring optical signal with a large-aperture detector in the direction of the reflected beam at different distances from the sample, inset of Fig. 2(c). The dependence of the detected light intensity on the distance *l* between the sample and the integrating sphere with 1 inch diameter opening is shown in Fig. 2(c). The diffused scattering, characterized by an increase of the signal at small distances *l,* has been observed in the roughened sample and was practically absent in the untreated one. By fitting the experimental result to a simple model, it has been shown that the ratio of scattering and specular light intensities is much smaller than the difference between the measured reflectances in the grinded and smooth samples. Thus, according to the most conservative estimate (assuming Lambertian scattering distribution diagram), the ratio of diffusely scattered light intensity to specularly reflected light intensity in the roughened sample did not exceed 3.5. This value is much smaller than the ratio of reflectances in the untreated and the corrugated samples, ~30 at small angles, see Fig. 1(b) and inset of Fig. 1(b).

3. Discussion and summary

One should note that high densities of photonic states and their effects on physical processes, such as spontaneous emission, are not unique to hyperbolic metamaterials. Thus, besides the classical example of cavities [26], the control of emission with high densities of photonic states in vicinity of metallic surfaces is predicted in Ref [27]. The only difference between emission quenching in vicinity of metals and in vicinity of hyperbolic metamaterials is that in the former case molecules emit to predominantly propagating modes (*k'*>>*k"*), while in the latter case – to predominantly evanescent modes (*k"*>>*k'*). (Here *k'* and *k"* are real and imaginary components of the wavenumber.)

High density of photonic states near plasmonic metals causes not only enhancement of emission decay rates but also reduction of reflection off roughened metallic surfaces. This effect, known for decades [28–30], has the same physical origin: high density of final photonic states increases scattering rate and determines its directionality – with majority of photons scattering inside the metal and only very small fraction scattered (reflected) to the air.

However, scattering of light to lossy modes in metal was not the predominant reason for the strong reduction of reflection observed in our experiment. First of all, the reduced reflectance off roughened silver-filled alumina membrane was accompanied by an enhancement of the sample's transmission, Fig. 2(d). This effect is consistent with light scattering to propagating modes in a hyperbolic metamaterials and is not expected at scattering to lossy modes in metallic components. Second, no dramatic reduction of reflectance was observed in an almost similar (control) membrane sample, which had a little bit smaller silver filling factor and did not have hyperbolic dispersion at the measurement wavelength of 873 nm (real components of both $\varepsilon_{||}$ and $\varepsilon_{\perp}$ positive). The reduction of specular reflection in the roughened control sample (~3 fold) was primarily due to onset of diffused scattering and redistribution of energy between specularly reflected light and diffusely reflected light.

Thus, the experimentally observed reduction of reflection off a roughened silver-filled alumina membrane is consistent with enhanced scattering of light into propagating modes of a hyperbolic metamaterial rather than evanescent modes of constituent metallic components. The fact that the reduced reflection was observed not only for p polarized incident light (at predicted by the theory) but also for s polarized light suggests change of the state of polarization upon scattering, which is commonly observed in scattering media [31]. We also note that characteristic vertical (600 nm) and horizontal (1.17 μm) dimensions of the surface roughness do not provide for good index matching with air, which is the known reason for reduction of reflectance in many nano-textured structures [32].

We thus have demonstrated that the reflectance of a hyperbolic metamaterial is significantly reduced upon roughening of its surface, in an agreement with the theoretical prediction. With the original concept equally applicable to all parts of the electromagnetic spectrum, our result thus opens an entirely new route towards radiation-absorbing materials

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and surfaces, with wide range of applications spanning from solar light harvesting to radar stealth technology.

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