A lossless metamaterial with tunable permittivity and its application as a compact phase shifter

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In this Letter, we propose a new type of lossless metamaterial whose effective permittivity is tunable from negative to positive values. Its optical response is studied analytically and numerically. We further demonstrate that this tunable metamaterial can significantly modulate the phase of an incident pulse with negligible reflection loss, functioning as an efficient phase shifter. © 2010 Optical Society of America

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Metamaterials are man-made composites engineered on a subwavelength scale, designed
to produce an optimized combination of electromagnetic properties that may not be readily
available in nature [1–3]. They have applications ranging from perfect imaging lenses [4] to
electromagnetic invisibility [5–7]. One rapidly emerging branch of this field concerns tunable
hybrid metamaterials in which plasmonic composites are integrated with natural materials
whose electrical, optical, thermal or mechanical properties can be externally controlled [8–20].
The resultant media have tunable effective material parameters, and their electromagnetic
response can be adjusted in real time. This dynamic tuning ability makes metamaterial
devices more flexible and versatile. For instance, frequency-agile terahertz metamaterials
have been fabricated recently by incorporating silicon in critical regions of metallic split-
ring resonators. Through photoexcitation of free carriers in the semiconductor, the resonant
frequency of the hybrid metamaterial can be tuned considerably under specific excitations
[15]. It was further demonstrated experimentally that this frequency-agile metamaterial can
modulate the phase or amplitude of the incident electromagnetic pulse, and therefore can
function as a phase modulator [21]. Unfortunately, owing to the intrinsic ohmic loss as
well as the resonant nature of plasmonic nanostructures, these tunable metamaterials suffer
significant electromagnetic absorption and thus degraded performance. To date this has been
a serious drawback preventing the implementation of tunable metamaterials into practical
devices.

One approach to alleviate or even eliminate the metal absorption loss is to introduce gain
media into the metamaterial composites [22, 23]. This method has been applied to achieve
spasers [24, 25] and lossless negative permittivity metamaterials [26–28]. In this Letter, we
will integrate the hybrid metamaterials with gain media to realize lossless metamaterials with
tunable permittivities. The metamaterial considered here consists of coated nanospheres with
a gain core dispersed in a homogeneous host material whose permittivity can be controlled
externally. Because the characteristic size of the nanoparticle is far smaller than the free-
space wavelength, we study its optical response using the electrostatic (long-wavelength)
approximation. We demonstrate that the hybrid metamaterial is always lossless at the wave-
length where the absorption cross section of the constitutive particle vanishes. Furthermore,
modifying the permittivity of the host medium will tune the real part of the effective permitt-
ity of the composite. Under certain conditions, the resultant bulk permittivity is tunable
from negative, through zero, to positive values. The analytical predictions are then validated
by comparing them with those of rigorous full-wave simulations. Finally, we propose that
a thin slab of the tunable metamaterial can linearly manipulate the phase of an excitation
pulse with negligible reflection, functioning as a compact phase shifter [21, 29].

We start by considering a coated sphere with inner radius $r_1$ and outer radius $r_2$. When
its size is much smaller than the free-space wavelength, an effective permittivity can be
employed to describe its classical optical response [30]

$$\epsilon_c = \frac{\epsilon_2 (1 + 2\rho) + 2\epsilon_2 (1 - \rho)}{\epsilon_1 (1 - \rho) + \epsilon_2 (2 + \rho)},$$

where $\rho = r_3^3/r_2^3$ is the volume fraction, $\epsilon_1$ and $\epsilon_2$ are the permittivity of the core and shell, respectively. Moreover, a composite consisting of these particles has an effective bulk permittivity governed by the Clausius-Mossotti relation [30]

$$\epsilon_b = \frac{\epsilon_m \epsilon_c (1 + 2f) + 2\epsilon_m (1 - f)}{\epsilon_c (1 - f) + \epsilon_m (2 + f)},$$

where $f$ is the filling fraction of the sphere, and $\epsilon_m$ is the permittivity of the host medium which is assumed to be positive and real-valued. This equation immediately suggests that the imaginary part of $\epsilon_c$ must vanish to result in a lossless $\epsilon_b$ since $\epsilon_m$ is real valued [28]. Furthermore, requiring $\epsilon_b \leq 0$ leads to the condition

$$\epsilon_m \frac{2 + f}{f - 1} < \text{Re} \epsilon_c \leq \frac{2\epsilon_m - 2f}{1 + 2f}.$$  \hspace{1cm} (3)

We graphically plot this inequality in Fig. 1, and assume $\epsilon_m$ has a minimum of 1.96 and a maximum of 4.0. The overlapped triangular region indicates the fully negative zone in which a set of point $(f, \text{Re} \epsilon_c)$ always results in a negative value of $\epsilon_b$. The vertex of the triangle therefore corresponds to a critical filling fraction $f_c$, and a smaller filling fraction may lead to a positive value of $\epsilon_b$. In addition, the partial derivative of $\epsilon_b$ with respect to $\epsilon_m$ can be derived from Eq. (2)

$$\frac{d\epsilon_b}{d\epsilon_m} = (1 - f) \frac{(\epsilon_c + 2\epsilon_m)^2 + 2f(\epsilon_c - \epsilon_m)^2}{[\epsilon_c (1 - f) + \epsilon_m (2 + f)]^2},$$

which is always positive for any $f < 1$, implying that $\epsilon_b$ is a monotonically increasing function of $\epsilon_m$. This is not surprising because the optical properties of the composite medium directly relate to the volume average of the guest and host media in the electrostatic approximation.

The observations above suggest that a design procedure consisting of two nearly independent steps can be employed. In the initial step, the first-order surface mode of a core-shell nanosphere is nearly excited at the targeted frequency such that the resulting $\epsilon_c$ has a vanishing imaginary part with a considerable negative real part [28]. An externally controllable host medium is then introduced at the second step to tune the effective bulk permittivity of the composite. To demonstrate this easy-to-use design procedure, we consider the following realistic example. The nanoparticles are silver coated spherical semiconductor quantum dots (with optical gain) which are almost identical to those studied in Ref. [28] except for the geometrical parameters: the radius of the metallic shell and the semiconductor core is 1.38 nm and 8.62 nm, respectively. This coated sphere is found to possess a lossless negative $\epsilon_c$.
at a wavelength of 834 nm. The host material in this case is chosen to be a planar aligned nematic liquid crystal, which possesses a large electro-optics response and has been employed to achieve reconfigurable metamaterials with a negative-zero-positive index of refraction in the optical regime [10, 12–14]. It should be emphasized that this hybrid composite may be fabricated by a sputter doping approach [17]. For linearly polarized light incident as an extraordinary wave onto the liquid crystal host, its permittivity depends on the director axis orientation angle $\phi$ with respect to the incident wave vector [10]

$$\epsilon_m = \frac{\epsilon_{\parallel}\epsilon_{\perp}}{\epsilon_{\parallel}\cos^2\phi + \epsilon_{\perp}\sin^2\phi},$$

(5)

where $\epsilon_{\parallel} \approx 4.0$ ($\epsilon_{\perp} \approx 1.96$) is the permittivity for light polarized parallel (perpendicular) to the director axis. Through modulating $\phi$ either electrically or optically, we can change $\epsilon_m$ from $\epsilon_{\perp}$ to $\epsilon_{\parallel}$ [10].

Using Eq. (2), the bulk permittivities $\epsilon_b$, corresponding to two different filling fractions, are computed and the results are plotted in Fig. 2(a) as solid curves. The effective permittivities $\epsilon_b$ have vanishing imaginary parts while the real parts can be monotonically tuned from negative, through zero, to positive values with the increment of $\epsilon_m$ of the liquid crystal. In addition, the zero $\epsilon_b$ condition is given by

$$\epsilon_m = -\frac{\epsilon_c(1 + 2f)}{2(1 - f)}.$$  

(6)

Hence, a smaller filling fraction therefore requires a smaller $\epsilon_m$. To validate the electrostatic results, we consider the nanoparticles in a simple cubic lattice with lattice spacing of 34.7 nm.

Fig. 1. A graphical display of Eq. (3) with $\epsilon_m$ in the region [1.96, 4.0]. $\epsilon_b$ is negative in each of the shaded regions for a specific $\epsilon_m$, and $\epsilon_b$ is negative in the overlapped triangle, regardless of $\epsilon_m$. 

![Graphical display of Eq. (3)](image-url)
Fig. 2. (a) The effective permittivity $\epsilon_b$ of the bulk composite for two filling fractions $f$. Solid curves show analytical results, while the circles represent full-wave simulations. The empty circles correspond to an isotropic model of the liquid crystal, while the filled circles correspond to an anisotropic model. (b) The spectra of a thin metamaterial slab, with a thickness of 34.7 nm, under normal incidence.

(corresponding to $f = 0.1$). A full-wave finite-element method is then used to simulate a single layer of lattice under normal incidence [31]. The calculated spectra are shown in Fig. 2(b) and the absorption is always zero, consistent with a lossless $\epsilon_b$. The transmission/reflection method is finally applied to extract the effective permittivity [32]. The result is plotted in Fig. 2(a) with empty circles, and is found to be in good agreement with its analytical counterpart. A rigorous anisotropic treatment of the liquid crystal elements, the same as that carried out in Ref. [12], is also utilized. We avoid the complicated cross-polarization coupling by carefully aligning the incident wave vector with the liquid crystal director axis [12]. The corresponding numerical result is plotted in Fig. 2(a) with filled circles, and is nearly identical to its isotropic counterpart. Notice that the value of $\epsilon_m$ of the anisotropic model is within $[2.25, 3.61]$, narrower than the isotropic model.

Our tunable permittivity metamaterials with compensated losses may have a variety of applications, such as $\epsilon$-near-zero materials [33]. Of these, the phase shifter may be the most interesting [21,29]. To illustrate its principles, we consider a thin slab, with thickness $d$ much smaller than the free-space wavelength $\lambda$, embedded in a vacuum. Under normal incidence its transmittance can be approximated as

$$ T \approx 1 - \frac{d^2 \pi^2}{\lambda^2} (\epsilon - 1)^2, \quad (7) $$

with $\epsilon$ being the real-valued, negative or positive, permittivity of the slab. Clearly the re-
flectance is negligible for a modest $\epsilon$. Moreover, the phase of the transmission is given by

$$\theta \approx \frac{d\pi}{\lambda} (\epsilon + 1),$$

implying that $\theta$ is a linear function of the permittivity: It is negative when $\epsilon < -1$ and positive when $\epsilon > -1$. Consequently, the transmitted light can be delayed or advanced. The single layer slab studied above can serve as an example. As shown in Fig. 2(b), $\epsilon_b = -1$ indeed results in a zero $\theta$, and the accumulated phase difference $\Delta\epsilon_b d\pi/\lambda$, obtained by increasing $\epsilon_m$ from 2.0 to 4.0, is around $0.25\pi$. Notice that the thickness of the slab is only 34.7 nm, indicating that it operates as an efficient phase shifter. In addition, to achieve a larger accumulated phase difference, we need to increase the slab thickness which, however, decreases the transmittance. The linear dependence of $\theta$ and quadratic dependence of transmittance $T$ on $d$ suggest an approach to alleviate the reflection loss considerably: Replace a single layer slab with multilayer slabs while keeping the total thickness unchanged. It should be mentioned that a similar method has been employed to improve the imaging resolution of near-sighted superlenses [34,35].

In summary, we have described an innovative hybrid metamaterial, formed by integrating gain medium with an externally controllable host material. The composite exhibits a tunable effective permittivity with compensated losses. It is further suggested that a thin slab of this tunable material can function as an efficient phase shifter whose transmittance is nearly 100%.

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References

1. R. Marqués, F. Martín, and M. Sorolla, *Metamaterials with Negative Parameters* (Wiley, 2008).
2. L. Solymar and E. Shamonina, *Waves in Metamaterials* (Oxford University, 2009).
3. *Physics of Negative Refraction and Negative Index Materials*, edited by C. M. Krowne and Y. Zhang (Springer, 2007).
4. J. B. Pendry, “Negative Refraction Makes a Perfect Lens,” Phys. Rev. Lett. 85, 3966 (2000).
5. J. B. Pendry, D. Schurig, and D. R. Smith, “Controlling Electromagnetic Fields,” Science 312, 1780 (2006).
6. U. Leonhardt, “Optical Conformal Mapping,” Science 312, 1777 (2006).
7. D. Schurig, J. J. Mock, B. J. Justice, S. A. Cummer, J. B. Pendry, A. F. Starr, and D. R. Smith, “Metamaterial Electromagnetic Cloak at Microwave Frequencies,” Science 314, 977 (2006).
8. W. J. Padilla, A. J. Taylor, C. Highstrete, M. Lee, and R. D. Averitt, “Dynamical Electric and Magnetic Metamaterial Response at Terahertz Frequencies,” Phys. Rev. Lett. 96, 107401 (2006).
9. H.-T. Chen, W. J. Padilla, J. M. O. Zide, A. C. Gossard, A. J. Taylor, and R. D. Averitt, “Active Terahertz Metamaterial Devices,” Nature 444, 597 (2006).
10. I.-C. Khoo, D. H. Werner, X. Liang, A. Diaz, and B. Weiner, “Nanosphere Dispersed Liquid Crystals for Tunable Negative-zero-positive Index of Refraction in the Optical and Terahertz Regimes,” Opt. Lett. 31, 2592 (2006).
11. D. H. Werner, D.-H. Kwon, I.-C. Khoo, A. V. Kildishev, and V. M. Shalaev, “Liquid Crystal Clad Near-Infrared Metamaterials with Tunable Negative-Zero-Positive Refractive Indices,” Opt. Express 15, 3342 (2007).
12. X. Wang, D.-H. Kwon, D. H. Werner, I.-C. Khoo, A. V. Kildishev, and V. M. Shalaev, “Tunable Optical Negative-index Metamaterials Employing Anisotropic Liquid Crystals,” Appl. Phys. Lett. 91, 143122 (2007).
13. D.-H. Kwon, X. Wang, Z. Bayraktar, B. Weiner, and D. H. Werner, “Near-infrared Metamaterial Films with Reconfigurable Transmissive/Reflective Properties,” Opt. Lett. 33, 545 (2008).
14. J. A. Bossard, X. Liang, L. Li, D. H. Werner, B. Weiner, P. F. Cristman, A. Diaz, and I.-C. Khoo, “Tunable Frequency Selective Surfaces and Negative-zero-positive Index Metamaterials Based on Liquid Crystals,” IEEE Trans. Antennas Propag. 56, 1308 (2008).
15. H.-T. Chen, J. F. O’Hara, A. K. Azad, A. J. Taylor, R. D. Averitt, D. B. Shreken-
hamer, and W. J. Padilla, “Experimental Demonstration of Frequency-agile Terahertz Metamaterials,” Nat. Photonics 2, 295 (2008).

16. S. Xiao, U. K. Chettiar, A. V. Kildishev, V. Drachev, I. -C. Khoo, and V. M. Shalaev, “Tunable Magnetic Response of Metamaterials,” Appl. Phys. Lett. 95, 033115 (2009).

17. H. Yoshida, K. Kawamoto, H. Kubo, T. Tsuda, A. Fujii, S. Kuwabata, and M. Ozaki, “Nanoparticle-Dispersed Liquid Crystals Fabricated by Sputter Doping,” Adv. Mater. 22, 622 (2009).

18. H. Nˇ emec, P. Kuˇ zel, F. Kadlec, C. Kadlec, R. Yahiaoui, and P. Mounaix, “Tunable Terahertz Metamaterials with Negative Permeability,” Phys. Rev. B 79, 241108(R) (2009).

19. T. Driscoll, H. -T. Kim, B. -G. Chae, B. -J. Kim, Y. -W. Lee, N. M. Jokerst, S. Palit, D. R. Smith, M. D. Ventra and D. N. Basov, “Memory Metamaterials,” Science 325, 1518 (2009).

20. Q. Liu, Y. Cui, D. Gardner, X. Li, S. He, and I. I. Smalyukh, “Self-Alignment of Plasmonic Gold Nanorods in Reconfigurable Anisotropic Fluids for Tunable Bulk Metamaterial Applications,” Nano Lett. 10, 1347 (2010).

21. H. -T. Chen, W. J. Padilla, M. J. Cich, A. K. Azad, R. D. Averitt, and A. J. Taylor, “A Metamaterial Solid-state Terahertz Phase Modulator,” Nat. Photonics 3, 148 (2009).

22. D. J. Bergman and M. I. Stockman, “Surface Plasmon Amplification by Stimulated Emission of Radiation: Quantum Generation of Coherent Surface Plasmons in Nanosystems,” Phys. Rev. Lett. 90, 027402 (2003).

23. R. F. Oulton, V. J. Sorger, T. Zentgraf, R. M. Ma, C. Gladden, L. Dai, G. Bartal, and X. Zhang, “Plasmon Lasers at Deep Subwavelength Scale,” Nature 461, 629 (2009).

24. N. I. Zheludev, S. L. Prosvirnin, N. Papasimakis, and V. A. Fedotov, “Lasing Spaser,” Nat. Photonics 2, 351 (2008).

25. M. A. Noginov, G. Zhu, A. M. Belgrave, R. Bakker, V. M. Shalaev, E. E. Narimanov, S. Stout, E. Herz, T. Suteewong, and U. Wiesner, “Demonstration of a Spaser-based Nanolaser,” Nature 460, 1110 (2009).

26. Y. Fu, L. Thylén and H. Ágren, “A Lossless Negative Dielectric Constant from Quantum Dot Exciton Polaritons,” Nano Lett. 8, 1551 (2008).

27. A. Bratkovsky, E. Ponizovskaya, S. Y Wang, P. Holmström, L. Thylén, Y. Fu, and H. Ágren, “A Metal-wire/Quantum-dot Composite Metamaterial with Negative $\epsilon$ and Compensated Optical Loss,” Appl. Phys. Lett. 93, 193106 (2008).

28. Y. Zeng, Q. Wu and D. H. Werner, “Electrostatic Theory for Designing Lossless Negative Permittivity Metamaterials,” Opt. Lett. 35, 1431 (2010).

29. P. He, P. V. Parimi, Y. He, V. G. Harris, and C. Vittoria, “Tunable Negative Refractive Index Metamaterial Phase Shifter,” Electron. Lett. 43, 1440 (2007).

30. C. F. Bohren and D. R. Huffman, Absorption and Scattering of Light by Small Particles
(John Wiley & Sons, 1998).

31. COMSOL, www.comsol.com.

32. D. R. Smith, S. Schultz, P. Markoš, and C. M. Soukoulis, “Determination of Effective Permittivity and Permeability of Metamaterials from Reflection and Transmission Coefficients,” Phys. Rev. B 65, 195104 (2002).

33. M. Silveirinha and N. Engheta, “Tunneling of Electromagnetic Energy through Subwavelength Channels and Bends using $\varepsilon$-Near-Zero Materials,” Phys. Rev. Lett. 97, 157403 (2006).

34. E. Shamonina, V. A. Kalinin, K. H. Ringhofer, and L. Solymar, “Imaging, Compression and Poynting Vector Streamlines for Negative Permittivity Materials,” Electron. Lett. 37, 1243 (2001).

35. V. A. Podolskiy and E. E. Narimanov, “Near-sighted Superlens,” Opt. Lett. 30, 75 (2005).