Nanosphere dispersed liquid crystals for tunable negative–zero–positive index of refraction in the optical and terahertz regimes

I. C. Khoo, D. H. Werner, X. Liang, and A. Diaz

Department of Electrical Engineering, Pennsylvania State University, University Park, Pennsylvania 16802

B. Weiner

Department of Physics, Pennsylvania State University, University Park, Pennsylvania 16802

Received May 5, 2006; revised June 12, 2006; accepted June 21, 2006;
posted June 23, 2006 (Doc. ID 70663); published August 9, 2006

An analysis of aligned nematic liquid crystal cells containing core-shell nanospheres shows that it is possible to devise a new type of metamaterial whose index of refraction is tunable from negative, through zero, to positive values. The design parameters for the constituents can be scaled for application in the optical as well as very long wavelength (e.g., terahertz and microwave) regions. © 2006 Optical Society of America

OCIS codes: 160.3710, 160.4670, 110.2990, 230.3720, 260.5740.

Current research and development in electro-optical and nonlinear optical materials for photonic applications are largely centered on nanostructured materials that exhibit unique physical and optical properties.[1–16] In particular, research in negative index materials[6–13] (NIMs) and zero index materials[14–16] (ZIMs) has been very active over the past few years. In most of these cases, the resulting structure's index and other optical properties tend to be fixed. On the other hand, it is highly desirable that these properties and functions could be reconfigured or tuned. In that regard, liquid crystals stand out as the preferred choice as they possess very broadband transparency and birefringence[17] and easy susceptibilities to applied fields (i.e., large electro-optics and nonlinear optical responses). They are also compatible with almost all widely used optoelectronic materials, and their fluid nature allows easy incorporation into various geometries and nanometer-scale pore sizes.[1–6] Recent studies[5,6] of liquid crystal infiltrated three-dimensional inverse opal structures and periodic nanostructured frequency selective surfaces,[6] for example, have demonstrated their exceptionally large spectral transmission tunability as well as the capability to exhibit negative index of refraction behavior.

Instead of such nanoscale periodic structures, which require complex fabrication techniques, others have reported the possibility of realizing negative index materials by dispersing metallic spheres in the bulk of some host materials. Wangberg *et al.*,[8] for example, have studied the possibility of left-handed propagation in a strongly anisotropic liquid crystal waveguide containing metallic nanospheres. Such a configuration, however, would encounter large scattering loss and light coupling difficulties at the entrance and exit sides of the waveguide.

In this Letter, we describe a configuration involving "free" propagation of polarized light through aligned nematic liquid crystal cells in which core-shell (coated) nanospheres are dispersed [see Fig. 1(a)]. While the treatment can be applied to various liquid crystal alignments (planar, homeotropic, cholesteric spiral) and other phases, we limit our attention here to planar aligned nematic liquid crystals (NLCs). The structure of the coated dielectric (nonmagnetic) spheres is as shown in Fig. 1(b).

The electromagnetic material properties of nanosphere dispersed liquid crystals may be calculated by employing the Maxwell Garnet mixing rule[18,19] for a medium with three distinct regions: the host liquid crystal (region 3), the shell (region 2), and the core (region 1). All three materials are nonmagnetic with relative permeability equal to 1. It is the combination of the permittivities at the appropriate resonances, in conjunction with the field-induced permittivity change in the liquid crystal host that give rise to the effective refractive index of nanosphere dispersed liquid crystals (NDLCs) that can vary from negative to zero to positive values.

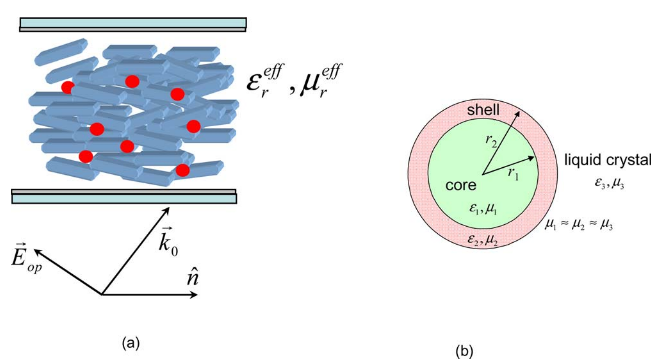

Fig. 1. (Color online) Homogeneous or planar aligned nematic liquid crystal containing a core-shell (coated) nanospheres sample. An applied ac electric field or a magnetic field can switch the director axis of the liquid crystal from parallel to perpendicular to the cell windows. (b) Cross section of the core $[\mu_1, \epsilon_1]$-shell $[\mu_2, \epsilon_2]$ nanosphere with the liquid crystal host $[\mu_3, \epsilon_3]$.

0146-9592/06/172592-3/$15.00

Recent studies have shown that polaritonic materials such as $LiTaO_3$ (Refs. 12 and 20) would be a good candidate material for the core. Its permittivity is described by

$$\epsilon_1 = \epsilon(\infty)\left(1 + \frac{\omega_L^2 - \omega_T^2}{\omega_T^2 - \omega^2 - i\omega\gamma_1}\right), \tag{1}$$

where $\epsilon(\infty)$ is the high-frequency limit of the permittivity, ω is the incident frequency, ω_T and ω_L are the transverse and longitudinal optical phonon frequencies, respectively, and γ_1 is the damping coefficient. The shell can also be a polaritonic material or a Drude material with a permittivity of the form

$$\epsilon_2 = 1 - \frac{\omega_p^2}{\omega^2 + i\omega\gamma_2}, \tag{2}$$

where ω_p is the plasma frequency and γ_2 is the damping term.

For linearly polarized light incident as an extraordinary wave onto the NLC host, the permittivity is given by[17]

$$\epsilon_3 = \frac{\epsilon_\parallel \epsilon_\perp}{\epsilon_\parallel \cos^2\theta + \epsilon_\perp \sin^2\theta}, \tag{3}$$

where $\epsilon_\parallel$ and $\epsilon_\perp$ are the respective permittivities for light polarized parallel and perpendicular to the director axis $\hat{n}$. Note that ϵ_3 is dependent on the director axis orientation angle θ with respect to the optical wave vector k_0, and does not carry any resonant dependence, unlike ϵ_1 and ϵ_2. In the optical region, losses are negligible for typical nematic cell thickness.

The Maxwell Garnet formula[18,19] is applied as the basic mixing rule to determine effective medium parameters for such NDLCs: in particular, the effective permittivity $\epsilon_r^{\mathrm{eff}}$ and permeability μ_r^{eff} are given, respectively, by

$$\epsilon_r^{\mathrm{eff}} = \epsilon_3\left(\frac{k_3^3 + j4\pi N a_1}{k_3^3 - j2\pi N a_1}\right), \tag{4a}$$

$$\mu_r^{\mathrm{eff}} = \frac{k_3^3 + j4\pi N b_1}{k_3^3 - j2\pi N b_1}, \tag{4b}$$

where

$$k_3 = \sqrt{\epsilon_3}k_0 = \left(\frac{\epsilon_\parallel \epsilon_\perp}{\epsilon_\parallel \cos^2\theta + \epsilon_\perp \sin^2\theta}\right)^{1/2} k_0. \tag{4c}$$

Note that for a fixed incident angle, the director axis orientation θ with respect to the optical wave vector can be modulated either electrically (by an ac bias field) or optically (through the optical intensity dependent director axis reorientation effect).[17] In both cases, the maximum reorientation angle is $\pi/2$, corresponding to an extraordinary refractive index change from $n_\perp \sim 1.4(\theta=0)$ to $n_\parallel \sim 2(\theta=\pi/2)$ [c.f. Fig. 1(a), i.e., permittivity change from ~2 to 4].

In Eqs. (4a) and (4b), a_1 and b_1 are the Mie scattering coefficients of the coated dielectric sphere,[19] N is the volume density of the spheres ($N=3f/4\pi r_2^3$), and f is the filling fraction of the composite. Only the a_1 and b_1 coefficients, corresponding, respectively, to the strength of the electric and magnetic dipole responses, are taken into account in the calculation of the effective permittivity and permeability. Detailed calculations and limitations of the current approach will be presented in a longer article elsewhere. Here we summarize the results pertinent to the optical infrared and terahertz regions.

The results for the real and imaginary parts of $\epsilon_r^{\mathrm{eff}}$, μ_r^{eff}, and the refractive index of NDLC for incident frequency in the optical infrared regime (~100 THz; wavelength $\lambda \sim 3.0\ \mu$m) are depicted in Figs. 2(a)–2(c). The parameters that produce the NIM–ZIM–PIM (positive index material) behavior for this frequency region are $r_2=0.143\ \mu$m, $r_1=0.13\ \mu$m, $f=0.12$, $\epsilon(\infty)=17$, $\omega_T/2\pi=240$ THz, $\omega_L/2\pi=570$ THz, $\gamma_1/2\pi=2.5$ THz, $\omega_p/2\pi=134$ THz, and $\gamma_2=\omega_p/60$. From Figs. 2(a) and 2(b), one can see that there is a frequency range where both $\epsilon_r^{\mathrm{eff}}$ and μ_r^{eff} are negative, a condition sufficient to yield a negative refractive index. Figure 2(c) depicts the real and imaginary parts

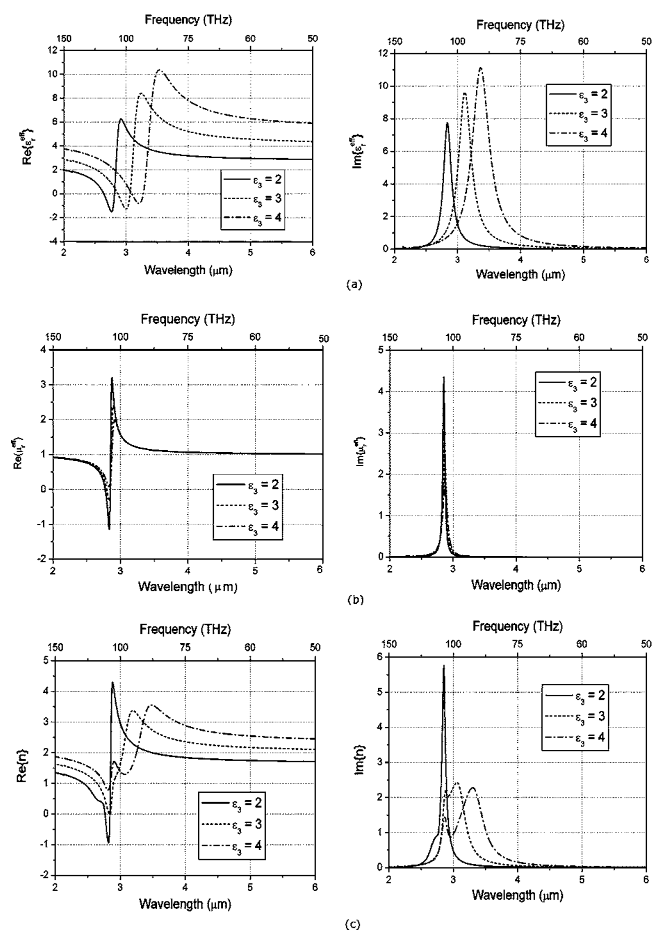

Fig. 2. Calculated real and imaginary part of the complex effective (a) permittivity, (b) permeability, and (c) refractive index of the NDLC as a function of liquid crystal host permittivity in the optical frequency interval between 50 and 150 THz (wavelength between 6 and 2 μm).

of the complex refractive index of NDLC for three representative permittivity values of the liquid crystal. It clearly shows that at the incident light frequency of 108 THz, the corresponding index of refraction n would change from +1 to 0 to −1. In other words, the NDLC metamaterial can be tuned to be a positive, zero, or a negative index material by changing the optical dielectric constant ϵ_3 of the nematic liquid crystal, which can be accomplished by an applied ac electric field, a magnetic field, or by another polarized optical field, or simply by changing the temperature of the sample.

The design of the material is scalable within a large dynamic range of operating frequencies. Figure 3, for example, shows the corresponding refractive index around the 3.8 THz resonance, calculated using the following parameters: $r_2=4.15\ \mu\text{m}$, $r_1=4\ \mu\text{m}$, $f=0.13$, $\epsilon(\infty)=13.4$, $\omega_T/2\pi=8$ THz, $\omega_L/2\pi=19$ THz, $\gamma_1/2\pi=0.05$ THz, $\omega_p/2\pi=4.2$ THz, and $\gamma_2=\omega_p/60$. Again, it clearly demonstrates that one can obtain a bulk material of negative, zero, and positive refractive index by varying the permittivity of the liquid crystal for incident frequencies around 3.8 THz.

In conclusion, we have described an innovative metamaterial, formed by randomly dispersing coated core-shell nanospheres in a nematic liquid crystal. The material will exhibit effective refractive indices ranging from negative, through zero to positive values as the host liquid crystal's permittivity is varied. By scaling the size of the spheres and their dielectric material properties, we can vary the operating frequency over a very large dynamic range, for example,

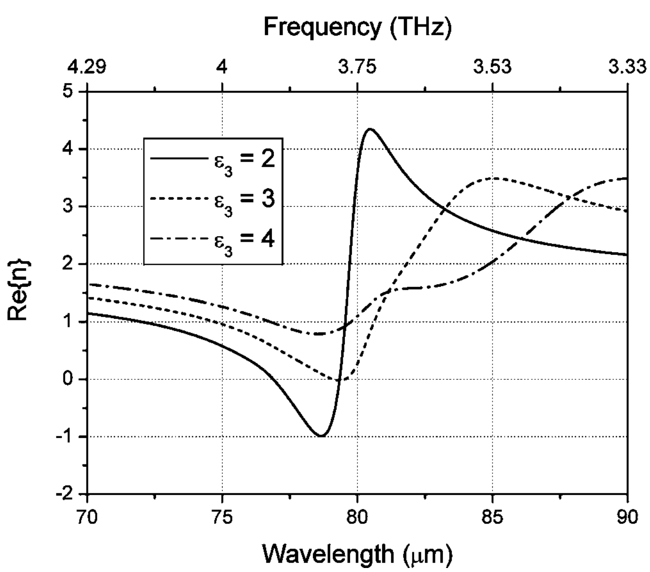

Fig. 3. Calculated real and imaginary part of the complex refractive index of the NDLC as a function of liquid crystal host permittivity in the terahertz region between 3.33 and 4.29 THz (wavelength between 90 and 70 μm).

from visible to the microwave region. Such reconfigurable materials will be very useful for designing tunable optical "flat" lenses or novel antenna coatings for microwave applications, in view of the broadband birefringence of nematic liquid crystal in the optical–microwave region.[17,21]

This work was supported by the Army Research Office, the Air Force Office of Scientific Research, and the National Science Foundation—Material Research Science and Engineering Center at Penn State. I. C. Khoo's e-mail address is ick1@psu.edu.

References

1. C. Schuller, F. Klopf, J. P. Reithmaier, M. Kamp, and A. Forchel, Appl. Phys. Lett. **82**, 2767 (2003).
2. G. V. Prakash, M. Kaczmarek, A. Dyadyusha, J. J. Baumberg, and G. D'Alessandro, Opt. Express **13**, 2201 (2005).
3. P. Mach, P. Wiltzius, M. Megens, D. A. Weitz, K.-H. Lin, T. C. Lubensky, and A. G. Yodh, Phys. Rev. E **65**, 031720 (2002).
4. T. T. Larsen, A. Bjarklev, D. S. Hermann, and J. Broeng, Opt. Express **11**, 2589 (2003).
5. E. Graugnard, J. S. King, S. Jain, C. J. Summers, Y. Zhang-Williams, and I. C. Khoo, Phys. Rev. B **72**, 233105 (2005).
6. I. C. Khoo, Y. Williams, A. Diaz, K. Chen, J. Bossard, D. Werner, E. Graugnard, and C. J. Summers, Mol. Cryst. Liq. Cryst. **453**, 309 (2006). See also Ref. 7.
7. J. A. Bossard, D. H. Werner, T. S. Mayer, J. A. Smith, Y. U. Tang, R. Drupp, and L. Li, IEEE Trans. Antennas Propag. **54**, 1265 (2006).
8. R. Wangberg, J. Elser, E. Narimanov, and V. A. Podolskiy, J. Opt. Soc. Am. B **23**, 498 (2006).
9. V. M. Shalaev, W. S. Cai, U. K. Chettiar, H. K. Yuan, A. K. Sarychev, V. P. Drachev, and A. V. Kildishev, Opt. Lett. **30**, 3356 (2005).
10. E. Schonbrun, M. Tinker, W. Park, and J.-B. Lee, IEEE Photonics Technol. Lett. **17**, 1196 (2005).
11. D. R. Smith, J. B. Pendry, and M. C. K. Wiltshire, Science **305**, 788 (2004).
12. M. S. Wheeler, J. S. Aitchison, and M. Mojahedi, Phys. Rev. B **73**, 045105 (2006).
13. G. V. Eleftheriades and K. G. Balmain, *Negative-Refraction Metamaterials* (Wiley, 2005).
14. R. W. Ziolkowski, Phys. Rev. E **70**, 046608 (2004).
15. V. A. Fedotov, A. V. Rogacheva, N. I. Zheludev, P. L. Mladyonov, and S. L. Prosvirnin, Appl. Phys. Lett. **88**, 091119 (2006).
16. M. A. Gingrich and D. H. Werner, Electron. Lett. **41**, 1266 (2005).
17. I. C. Khoo, *Liquid Crystals: Physical Properties and Nonlinear Optical Phenomena* (Wiley, 1995).
18. J. C. Maxwell Garnet, Philos. Trans. R. Soc. Ser. A **203**, 385 (1904).
19. C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles* (Wiley, 2004).
20. M. Schall, H. Helm, and S. R. Keiding, Int. J. Infrared Millim. Waves **20**, 595 (1999). See also Ref. 12.
21. F. Yang and J. Roy Sambles, Liq. Cryst. **30**, 599 (2003).