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We describe a class of liquid crystalline photonic metamaterials that exhibit tunable negative-zero-positive refractive indices. As a result of the extreme sensitivity of the nematic liquid crystal constituent, these metamaterials also exhibit extraordinarily large optical nonlinearities associated with the optical field induced director axis reorientation and birefringence change. Incorporation of a gain medium such as laser dye reduces losses of the metamaterial.

Keywords: Liquid crystals, nano-spheres, supra optical nonlinearity, negative and zero index, tunable metamaterials

1. INTRODUCTION

Current material systems that exhibit negative index properties tend to be passive and highly lossy. Recently, we demonstrated two types of nano-dispersed liquid crystalline metamaterials [1, 2] whose effective refractive index can be tuned from negative through zero to positive values over a very wide spectral range. The tunability is provided by incorporating electro-optics active and nonlinear-optical nematic liquid crystals [3]. In this paper, we also explore the ultimate optical nonlinearity possible in such metamaterials due to the supra-nonlinear optical properties of the nematic liquid crystal constituents. Furthermore, we demonstrate that one could reduce the loss associated with the dielectric resonances by introducing laser dye in the system.

2. NANO SPHERES DISPERSING LIQUID CRYSTALLINE METAMATERIALS

Fig 1 shows a schematic of an aligned nematic liquid crystal doped with randomly dispersed core-shell nano-spheres. The nematic liquid crystal can be in pure or doped (e.g. by Methyl-Red) forms, while the nano-spheres can be in the form of simple single constituent metallic (gold, silver, semiconductor) spheres or core-shell structures. The electromagnetic analysis for such metamaterials is considerably simpler than other schemes involving metallic spheres dispersed in liquid crystal waveguides as we can employ the effective medium approach.

We have studied a variety of nano-spheres in various combinations and liquid crystal alignments. Here we summarize the principal results for the case of core-shell polaritonic and Drude-type nano-spheres discussed in the literature [1, 4]. All constituent materials are non-magnetic with relative permeability equal to 1. For a polaritonic core, its permittivity is given by:
\[ \varepsilon_1 = \varepsilon(\infty) \left( 1 + \frac{\omega_T^2 - \omega_L^2}{\omega_L^2 - \omega^2 - i\gamma_1} \right) \]  

(1)

where \( \varepsilon(\infty) \) is the high-frequency limit of the permittivity, \( \omega \) is the incident frequency, \( \omega_T \) is the transverse optical phonon frequency, \( \omega_L \) is the longitudinal optical phonon frequency, and \( \gamma_1 \) is the damping coefficient.

The shell can be a polaritonic or a Drude material with a permittivity of the form:

\[ \varepsilon_2 = 1 - \frac{\omega_p^2}{\omega^2 + i\gamma_2}, \]

(2)

where \( \omega_p \) is the plasma frequency and \( \gamma_2 \) is the damping term. The optical permittivity of the host nematic liquid crystal (NLC) for a linearly polarized light incident at an oblique angle \( \theta \) is given by [3]

\[ \varepsilon_3 = \frac{\varepsilon_e \varepsilon_o}{\varepsilon_e \cos^2 \theta + \varepsilon_o \sin^2 \theta} \]

(3)

where \( \varepsilon_e \) and \( \varepsilon_o \) are the respective permittivities for light polarized parallel and perpendicular to the director axis \( \hat{n} \), and \( \theta \) is the angle made by the director axis with the optical wave vector \( k_0 \). Note that \( \varepsilon_3 \) does not carry any resonant dependence except for some small variation over the optical- infrared wavelength regime of interest here.

For a fixed incident angle, the director axis orientation \( \theta \) 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 (3)]. In both cases, the maximum reorientation angle is \( \pi/2 \), corresponding to an extraordinary refractive index change from \( n_e \sim 1.4 \) \( [\theta = 0] \) to \( n_e \sim 2 \) \( [\theta = \pi/2] \) i.e. permittivity change from \( \varepsilon_e = 2 \) to \( \varepsilon_e = 4 \) (\( \varepsilon_e \) for extraordinary and \( \varepsilon_o \) for ordinary waves). Such changes in the host index give rise to large changes in the effective refractive index of the nano-dispersed liquid crystal (NDLC) that in many cases. We have previously shown that the effective refractive indices of these metamaterials could be unusual negative-zero regions in some cases.

The effective refractive index of the nano-dispersed NLC is calculated by using the Maxwell Garnet mixing rule [5]. We begin by calculating the effective permittivity and permeability \( \varepsilon_{eff} \) and \( \mu_{eff} \) for NDLC as follow.

\[ \varepsilon_{eff} = \varepsilon \left( \frac{k_i^3 + j4\pi Na_i}{k_i^3 - j2\pi Na_i} \right), \]

(4a)

\[ \mu_{eff} = \frac{k_i^3 + j4\pi Nb_i}{k_i^3 - j2\pi Nb_i}. \]

(4b)

where

\[ k_i = \sqrt{\varepsilon_1 k_0 \left( \frac{\varepsilon_e \varepsilon_o}{\varepsilon_e \cos^2 \theta + \varepsilon_o \sin^2 \theta} \right)^{1/2}} k_0 \]

(4c)

In equations (4a-4b), \( a_i \) and \( b_i \) are the MIE scattering coefficients of the coated dielectric sphere [5], \( N \) is the volume density of the spheres (\( N = 3f/4\pi r_i^3 \)) and \( f \) is the filling fraction of the composite. Note that the Maxwell Garnet mixing method is valid only for small filling fractions, i.e. \( f < 1 \).

Some exemplary results for the effective refractive indices are shown in Fig. 2 calculated with the set of parameters for the constituent core, shell and host medium [1, 4]: \( \varepsilon(\infty) = 17, \omega_d / 2\pi = 570 \text{ THz}, \omega_d / 2\pi = 240 \text{ THz}, \gamma / 2\pi = 2.5 \text{ THz}, \mu_i = \mu_s = 1, \gamma_L = \omega_d / 60, \omega_d / 2\pi = 134 \text{ THz}, r_1 = 0.13 \mu m, r_2 = 0.143 \mu m \) and a filling fraction \( f = 0.1 \). In general, as a result of incorporating nano-spheres with higher dielectric constants than the host nematic liquid crystals (NLC), the effective birefringence \( n_e - n_o \) of the metamaterial is larger than the birefringence of the host liquid crystal (\( n_e \)}}
\( n_\text{eff} \), which is assumed to be 0.59 in these calculations [corresponding to the NLC dielectric constants varying from 2 to 4]. In the low-loss frequency regime (around 50 – 70 THz) where the imaginary component of \( n_\text{eff} \) is small, the effective [real part] refractive index change \( \Delta n \) is \( \approx 0.75 \) [at 50 THz] or 0.9 [at 70 THz] as the NLC permittivity is varied from 2 to 4. At 80 THz, where the loss is still acceptably low, the effective index change is even larger \([-1.1]\).

It is interesting to note that for some frequency interval around 106 THz, the effective refractive index of the liquid crystal metamaterial can be negative and is tunable from negative, through zero, to positive values as the host NLC dielectric constant (birefringence) is varied. For other parameter sets and filling fractions, the operating frequencies for these negative-zero-positive tunable refractive indices could be anywhere from the visible, near-, mid and far-IR region through THz and into the microwave regime, mainly due to the extremely broadband and large birefringence of the nematic liquid crystal host.

**Fig. 2** Real [left] and Imaginary [right] parts of the complex effective refractive index of the core-shell nano-spheres dispersed nematic liquid crystals showing tunable refractive index from negative to positive values.

### 3. LOSS REDUCTION WITH GAIN (DYE-DOPED LIQUID CRYSTAL HOST)

In spite of these unusual and remarkable properties of the liquid crystalline metamaterials, there is the inevitable presence of high optical losses in the same frequency region. The inevitability is associated with the relationship [Kramers-Kronig] between the real and imaginary part of the susceptibility of the medium. Nevertheless, within the constraint of the Kramers-Kronig relationship, one could ascertain an operation parameter set in which the loss is minimal, while the real part of the index is in the desirable range. We have investigated the possibility of reducing the loss by introducing laser dyes in the host liquid crystal matrix to provide gain [6]. Although the actual molecular levels and transitions in dye molecules are much more complex, we illustrate the effects by a two-level inverted system characterized by a susceptibility of the form \( \chi_g = \chi^r_g + i\chi^\prime_g \). Accordingly, the dielectric constant of the host [liquid crystal] becomes \( \varepsilon_h = \varepsilon_j + \chi^r_g \). Following Yariv [7], the imaginary part of the susceptibility \( \chi_g \) is given by (with an appropriate change of sign because of different conventions used here):

\[
\chi^\prime_g = \frac{\Delta \nu}{8\pi^3 n_s \Delta \nu} \frac{1}{\nu_\text{res}^3 n_s \Delta \nu} \times \frac{1}{(\Delta \nu)^2} = A \times \frac{1}{\nu^3 n_s \Delta \nu} \times \frac{1}{1 + 4(\nu - \nu_\text{res})^2 / (\Delta \nu)^2}
\]

with \( A = \frac{(N_s - N_j)c^2}{8\pi^3 n_s \Delta \nu} \) and \( \chi^\prime_\text{res}(\nu = \nu_\text{res}) \times \nu^3 n_s \Delta \nu \) where \( \chi^\prime_\text{res}(\nu = \nu_\text{res}) \) is the imaginary part of the susceptibility at the resonant frequency, and \( \Delta \nu \) is the transition linewidth, and \( n_s \) is the refractive index of the surrounding medium. From \( \chi^\prime_g \), the real part of the susceptibility of the gain medium may be derived from the Kramers-Kronig relations (7):
\[ \chi' = \frac{(N_f - N_i) \lambda^2}{8 \pi n t \Delta \nu} - \frac{2(v - v_c)}{\Delta \nu} \]

Fig. 3 show the real and imaginary part of the effective refractive index of the nano- and dye-doped nematic liquid crystals for three representative values of the imaginary part of the susceptibility at the resonant frequency \( \chi_{\text{max}} \) [0; 0.13 and 0.26] and a similar set of core-shell annospheres material parameters. It is clear that one could maintain negative-zero [real] refractive index while reducing the loss (\( \text{Im}[n_{\text{eff}}] \)) considerably. It is interesting to note that the real part of the refractive index (\( \text{Re}[n_{\text{eff}}] \)) could also be decreased even further with the incorporation of the gain medium (for \( \chi_{\text{max}} = -0.13 \)).

4. Liquid crystal clad metallo-dielectric nano-structures for tunable negative-zero-positive refractive indices.

Another nematic liquid crystal based metamaterial [2] capable of tunable negative-zero-positive refractive indices we have studied is schematically depicted in fig. 4. It consists of two aligned nematic liquid crystal layers sandwiching a metallo-dielectric nanostructure. The latter comprises a magnetic resonator made of two strips of silver of thickness 30 nm separated by a thin layer of alumina of thickness 20 nm. Negative permittivity needed for negative-index behavior is provided by thin silver films bounding the periodic array of magnetic resonators. The space between neighboring magnetic resonators is filled with silica. The optical properties of the metamaterial are analyzed using a rigorous full-wave electromagnetic scattering analysis based on the finite-element boundary-integral method [8]. Electric field values in a single unit cell of an infinitely periodic structure are determined by imposing periodic boundary conditions in the computational domain. Once the complex reflection and transmission coefficients are determined from the numerical analysis, the effective complex index of refraction can be unambiguously determined from well-established inversion procedures [9, 10].

The plots of the complex refractive index \( n = n' + n'' \) in Fig. 4 for two different incident light wavelengths as a function of the liquid crystal dielectric constant \( n_{\text{LC}} \) show dramatic changes in the effective refractive index created by the inclusion of these ‘resonant’ structures. For example, the lower right solid curve for incident light wavelength \( \lambda = 1.45 \mu\text{m} \) shows that the effective index of the metamaterials changes by 1.3 [from -1 to 0.3] as the LC dielectric constant is tuned from 2.25 to 2.9 [LC index change of 0.2 from 1.5 to 1.7]. From the preceding discussion on the effect of gain, clearly if the liquid crystal is doped with a dye to provide gain, both the real and the imaginary parts of the refractive index could be further modified.

Fig. 3. Real [upper] and imaginary [lower] parts of the refractive index of nano-dispersed dye-doped nematic liquid crystals showing enhanced negative index behavior and a lowered loss [smaller imaginary refractive index]
5. SUPRA-OPTICAL NONLINEARITY OF NANO-DISPERSED NEMATIC LIQUID CRYSTALLINE METAMATERIALS

As remarked earlier, the refractive index [dielectric constant] of the host liquid crystal can be electrically or optically modified. Since electrical contacts are extremely cumbersome to incorporate in the nanostructure, optical tuning is a much more desirable means. We close this paper with a discussion of the possibility of achieving high supra-optical nonlinearity in these dye-doped liquid crystal metamaterials. Nonlinear light scatterings in liquid crystals have been extensively investigated over the past two decades [3, 11-36]. Perhaps the most nonlinear mechanism is the optically induced director axis reorientation, which is characterized by an optical index change coefficient $n_2$ [defined by $n_2=\Delta n/I$, where $\Delta n$ is the light induced index change and $I$ the optical intensity]. In dye-doped nematic liquid crystals, supra-optical nonlinearities with $n_2 >>1 \text{ cm}^2/\text{W}$ were first discovered [23] in methyl red dye molecules-, and subsequently also in C60- and carbon nanotube- doped NLC [26-28]. In this section, we re-examine the fundamental principles for such supra-nonlinearity and illustrate the possibility of realizing even larger optical nonlinearity with the higher-birefringence liquid crystal metamaterials discussed in the preceding sections.

Consider the basic light-LC interaction as depicted in Fig. 1. The energy density involved in reorienting the LC axis by an angle $\theta$ is

$$U = \frac{\text{erg}}{\text{cm}^2} \approx K \left( \frac{\partial \theta}{\partial x} \right)^2 L,$$

where $L$ is the interaction length and $K$ the LC elastic constant [3]. In a wave mixing type of interaction in which the impinging optical intensity is sinusoidal, the reorientation angle $\theta$ is of the form $\theta = \theta_0 \sin (qx)$ where $q=2\pi/\Lambda$ is the wave vector of the grating, and $\Lambda$ is the grating constant. In this case, we have $U_{LC} = K \pi^2 \theta^2 L/\Lambda^2$. On the other hand, the energy provided by the light beam is $E_{\text{light}} = I\tau (1-e^{-\alpha\tau}) \sim \alpha LI\tau$ where $\alpha$ is the loss coefficient (due to the transfer of energy from light to nematic reorientation per unit length) and $\tau$ is the response time for the process. Equating $U$ and $E_{\text{light}}$ by assuming complete conversion of transferred light energy to reorientation,
we get \( K \pi^2 \theta^2/\alpha \Lambda^2 \approx I \tau \). In the case when the liquid crystal is initially homeotropically aligned, a reorientation of angle \( \theta \) will give rise to a change in the index experienced by the incident extraordinary-wave \( \Delta n \sim (n_e-n_o) \theta^2/4 \pi^2 \alpha \Lambda^2 I \tau \). Writing \( \Delta n = n_2 I \) yields the nonlinear index coefficient \( n_2 \)

\[
n_2 \sim (n_e-n_o) \tau \alpha \Lambda^2/K \pi^2. \tag{7}
\]

Depending on various parameters such as the birefringence and viscosity, sample thickness, and other factors like laser intensity, presence of other applied fields or photosensitive dopants, as well as the actual process involved, the value of \( n_2 \) and the response time can vary considerably. In wave mixing studies, typical \( r \) is on the order of 10’s of millisecond (10^{-2} sec), for \( \Lambda \sim 20 \mu m \). Using \( K \sim 10^3 \text{erg/cm,} \ (n_e-n_o) \sim 0.2 \), \( \alpha \sim 100 \text{cm}^2/\text{s} \), we have \( n_2 \sim 1 \text{cm}^2/\text{W} \). Even larger \( n_2 \) values approaching 1000 cm^2/W can be expected and have indeed been observed in dyed doped nematic liquid crystals [23, 28]. From the expression for \( n_2 \), it is also clear that a larger effective birefringence provided by the liquid crystalline metamaterials discussed in the preceding sections would result in an enhancement of these supra-optical nonlinearities. Accordingly, the optical power density required to effect the desired refractive index change in these metamaterials could be as low as 100 nW (nanowatt) as reported before in dye-doped nematic liquid crystals [35].

6. CONCLUSION

In conclusion, we have described two new forms of liquid crystalline metamaterials that possess tunable and highly nonlinear optical properties. By field induced reorientation of the liquid crystal host, thereby changing its permittivity, the material will exhibit effective refractive indices ranging from negative, through zero to positive values. The large effective refractive index change also results in enhancing the optical nonlinearity. The proposed structures are highly scalable in that the physical dimensions of its constituents can be varied over a very wide range, resulting in metamaterials whose operating wavelengths can cover the optical, through near- and far-IR to the microwave regimes. In this rather wide spectral band, nematic liquid crystals possess large birefringence and extreme photosensitivity that would allow tuning with low applied field thresholds [3].

ACKNOWLEDGEMENT

This work was supported by the Army Research Office, the Air Force Office of Scientific Research and Penn State MRSEC under NSF grant DMR-00800190.

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