Optical chiral metamaterial based on the resonant behaviour of nanodiscs

Mahdi Kordi\textsuperscript{a, b} and Mir Mojtaba Mirsalehi\textsuperscript{b}

\textsuperscript{a}Communications and Computer Research Center, Ferdowsi University of Mashhad, Mashhad, Iran; \textsuperscript{b}Electrical Engineering Department, Ferdowsi University of Mashhad, Mashhad, Iran

ABSTRACT
Circular dichroism and optical activity have been achieved by chiral metamaterials in the optical spectrum, but for the case of negative index of refraction, remarkable achievements have not been obtained in this region so far. We employ nanoparticles to shift the resonant frequency of a chiral metamaterial based on twisted cross wires to optical domain. Our proposed structure provides giant optical activity, strong circular dichroism and also negative refractive index in the optical wavelengths. Optical activity in our structure has a rotary power similar to a gyrotropic crystal of quartz, but in a thickness which is four orders of magnitude smaller. The foundation of our method for realizing such an optical chiral metamaterial is based on creating a different coupling between longitudinal modes of localized surface plasmons for right and left circularly polarized incident waves.

ARTICLE HISTORY
Received 28 November 2015
Accepted 8 February 2016

KEYWORDS
Optical chiral metamaterial; resonant frequency of nanoparticles; optical activity; circular dichroism

1. Introduction
Metamaterials are artificial materials that their unusual properties, such as negative permeability and negative refractive index, depend on their geometries not their components. These unusual properties can offer special applications such as electromagnetic absorbers, electrically small antennas (1) and the cloak of invisibility (2). In 2003 (3) and later in 2004 (4), chiral structures were used as an alternative to create metamaterials. In general, chiral structures have no symmetry planes. In addition to negative index of refraction, chiral metamaterials also show optical activity and circular dichroism. Optical activity can be used to manipulate polarization, and circular dichroism can serve as circular wave polarizer. Several different designs have been proposed and investigated for chiral metamaterials having all three features (optical activity, circular dichroism and negative index of refraction) in the microwave (5–8) and THz regimes (9–11). Also, at optical frequencies, various designs are reported for chiral metamaterials, but only optical activity and circular dichroism have been achieved, not negative index of refraction (12–15). In (16) and (17), chiral metamaterials with negative refractive index in the near infrared and optical spectrums are reported, respectively.

For realizing chiral metamaterials, unlike conventional methods, there is no need for the permittivity and the permeability to be negative simultaneously. Using this feature, metamaterials can be realized easier. Chiral metamaterials show different behaviour against right circularly polarized (RCP) and left circularly polarized (LCP) waves. One of the parameters used to show this difference is the chirality coefficient. In natural materials, this parameter is very weak, but in chiral metamaterials, it is so large that makes one of the circularly polarized waves to have negative refractive index.

At high frequencies, metals do not behave as a perfect conductor. As a result, a type of propagating surface mode that is called surface plasmon polariton, is created. The dispersion diagram, i.e. frequency versus propagation constant diagram, of the surface plasmon polaritons has a negative slope that can be used to make metamaterials (18–20). Also, as another result of the finite conductivity, when a metallic nanoparticle is exposed to an electric field, it resonates and creates a strong field around itself that can be up to several hundred times of the applied field. Alu and coworkers have used this point to create a metamaterial in the visible range of the spectrum (21).

In this paper, we use nanoparticles and their surface plasmon frequencies to shift the resonance frequency of a chiral metamaterial to the optical region. In Section 2, the proposed structure is described in detail. The simulation results and the retrieved effective parameters are provided in Section 3. The last section of the paper is devoted to the conclusions.

2. Proposed structure
In (6), a chiral metamaterial based on twisted cross wires is reported in which, resonance occurs when the lengths
We present a design method to select the frequency band of negative refractive index in a wide range of optical spectrum. One can make major changes to this frequency band by changing the radii of the nanodiscs or minor changes by changing the thickness of the dielectric spacer or the size of the unit cell, but three points should be noticed in this regard.

As the first point, the twist angle of the nanodisc crosses between successive layers, $\Delta \varphi$, should be proportional to the thickness of the dielectric spacer. It might seem that a $22.5^\circ$ twist angle would produce the most chirality in the structure, at least in one with two layers of nanodisc crosses, but, the simulations show different results. If this twist angle is equal to the retardation phase of a circularly polarized wave going through the dielectric spacer, longitudinal modes of localized surface plasmons are excited efficiently. Since in our structure, the direction of rotation of nanodisc crosses between successive layers is left handed, this excitation could happen easier for the case of LCP illumination. This event could have occurred for RCP illumination, if the direction of rotation is right handed. Therefore, the twist angle should be

$$\Delta \varphi = \frac{2\pi nd}{\lambda},$$

where $d$ and $n$ are the thickness and the refractive index of dielectric spacer, respectively, and $\lambda$ is the wavelength that corresponds to the resonant frequency of nanodisc crosses. Let $T_+$ and $T_-$ be the transmission coefficients of the structure for the incident RCP and LCP waves, respectively. If Equation (1) is satisfied, the deepest notch is created in the magnitude diagram of $T_+$ or $T_-$ (in our case, $T_-$.). The maximum depth means the maximum difference between the two different circular polarization and consequently maximum circular dichorism, maximum optical activity and also maximum chirality. Therefore, $\Delta \varphi$ could be seen as a tuning parameter for the amount of circular dichorism. In order to reach negative index of refraction, one does not need maximum chirality, but the chirality must be large enough to overcome the first term on the right side of the following equation (24)

$$n_{\pm} = \sqrt{\mu \epsilon} \pm \kappa,$$

where $\mu$ and $\epsilon$ are the effective permeability and effective permittivity of the medium, respectively, $\kappa$ is the chirality coefficient of the structure and finally, (+) and (−) refer to RCP and LCP waves, respectively. However, larger chirality leads to more bandwidth for negative index of refraction. On the other hand, for the negative refractive indices, $n = -1$ is the best case and only under this condition, perfect reconstruction of the image is possible (25).
Figure 2. Schematic representation of the proposed chiral metamaterial with three layers of nanodisc crosses. (a) Top view. (b) Side view. The second layer is embedded in the middle of the dielectric. The parameters are: $a_x = a_y = 500 \text{ nm}$, $\varphi_1 = 27.5^\circ$, $\Delta \varphi_1 = \Delta \varphi_2 = 17.5^\circ$ and $t_2 = 60 \text{ nm}$. For better understanding the structure, an array of $3 \times 3$ unit cells is shown in (c). (The colour version of this figure is included in the online version of the journal.)

As the second point, our simulations on various designs show that the designed structure should have fourfold rotational symmetry, i.e. the rotation of the structure by $90^\circ$, results in similar structure. Under this constraint, the two linear components of the incident circularly polarized wave (e.g. $E_x$ and $E_y$, if the propagation direction of the incident wave is $z$-axis) see similar structures. In other words, it helps that the model of structure follows the model of the reciprocal chiral mediums, i.e. when the incident wave is RCP (LCP), the outgoing wave remains RCP (LCP) too and the reflected wave would be LCP (RCP).

As the third point, since the foundation of chirality is based on the difference between $T_+ \text{ and } T_-$, the magnitude of one of them which corresponds to the negative refractive index, should be small. As a result, the area of the nanoparticles on the surface of the unit cell, should not be small. Indeed, there is a trade-off between transmission coefficient and, as mentioned earlier, an easier way to reach the negative index materials. On the other hand, relatively low transmission coefficient means relatively high insertion loss.

We simulated the structure with two and three layers of nanodisc crosses. In the structure with two layers of nanodisc crosses, large circular dichorism and also optical activity were achieved but not large enough to create a noticeable negative refractive index. By increasing the number of layers to three, circular dichorism, optical activity and negative index of refraction were all obtained.

3. Simulation and retrieving the effective parameters

Both of our proposed structures, with two and three layers of nanodisc crosses are classified in the reciprocal chiral mediums and their electromagnetic behaviours are characterized by the following constitutive relations (26)

$$
\begin{align*}
D &= \varepsilon \varepsilon_0 E + i k / c_0 H, \\
B &= -i k / c_0 E + \mu \mu_0 H,
\end{align*}
$$

where $c_0$ is the speed of light in vacuum.

We used Lumerical FDTD Solutions to simulate our proposed structures. All simulations were done for normal incidence of circularly polarized waves propagating in the $+z$-direction. Our simulations show that the resonant wavelength of a single nanodisc cross placed above a 20-nm thick dielectric, is about 600 nm. So, from Equation (1), $\Delta \varphi = 17.85^\circ$, but as mentioned above different parameters can affect the resonant wavelength of nanoparticles. After optimization, we set $\Delta \varphi = 17^\circ$ and $\Delta \varphi_1 = \Delta \varphi_2 = 17.5^\circ$ for the structures with two and three layers of nanodisc crosses, respectively.

The transmission coefficients of RCP and LCP waves are shown in Figure 3(a) and (b) for the structures with two and three layers of nanodisc crosses, respectively. For both structures, the conversion between two polarizations is negligible (below $5 \times 10^{-5}$) in all of the frequency band of simulation. The polarization azimuth rotation, $\theta$,
and the ellipticity angle, $\eta$, for the linearly polarized light are calculated using the following relations (27)

\[
\theta = \frac{1}{2} \left[ \arg(T_+) - \arg(T_-) \right] \quad (4)
\]
\[
\eta = \frac{1}{2} \arcsin \left( \frac{|T_+| - |T_-|}{|T_+| + |T_-|} \right). \quad (5)
\]

The results are shown in Figures 4(a) and 5(a) for the structures with two and three layers of nanodisc crosses, respectively. As can be seen in Figure 3(a) and (b), there is a deep difference between the transmission coefficients of RCP and LCP waves that results in a strong circular dichorism for both structures. This strong circular dichorism can also be understood from the relatively large ellipticity angles shown in Figures 4(a) and 5(a).

As can be seen in these figures, the maximum values of polarization azimuth rotations are $19^\circ$ and $135^\circ$ for two and three layers of nanodisc crosses, respectively. The optical activity in our proposed structure is noticeable. The polarization azimuth rotation of gyrotropic crystal of quartz is approximately $20^\circ$ for a 1 mm thick plate at a wavelength of 589 nm (28), while our proposed structure (the case with three layers) provides this rotation only in 100-nm thickness at a wavelength of 622 nm. At this wavelength, $\eta = 0$ and $\theta = 22.5^\circ$ and also $|T_+| = |T_-| = -1.15$ dB, i.e. a low insertion loss. The $\eta = 0$ case means that a linear polarized light can pass thorough the structure without a change in its polarization.

We used the method that is introduced in (29) for retrieving the effective parameters of our proposed chiral metamaterials. The real and imaginary parts of chirality and also the real and imaginary parts of refractive indices of the RCP and LCP waves for the case of the structure with two and three layers of nanodisc crosses are shown in Figures 4(b)–(d) and 5(b)–(d), respectively. As can
Figure 5. Characteristic curves of the structure with three layers of nanodisc crosses: (a) The polarization azimuth rotation, \( \theta \), and the ellipticity angle, \( \eta \). The real and imaginary parts are shown for the following parameters. (b) chirality, (c) refractive index for incident RCP wave, (d) refractive index for incident LCP wave, (e) permeability and (f) permittivity. (The colour version of this figure is included in the online version of the journal.)

Figure 6. A 5-layer structure (without the middle dielectrics): (a) top view, (b) side view.

Figure 7. The real part of the normal components of electric fields \( E_z \) on the two nanodisc crosses corresponding to the structure with two layers for LCP illumination at the resonant frequency, \( f = 508 \) THz. (The colour version of this figure is included in the online version of the journal.)
be seen in Figure 4(c)–(d), the real part of refractive indices are approaching to zero and even goes below zero for RCP incident wave in a part of the simulation spectrum, but, this is not a noticeable achievement. For the structure with three layers of nanodisc crosses, the refractive index is negative for incident RCP wave from \( f = 468 \) to \( f = 477 \) THz and for incident LCP wave from \( f = 504 \) to \( f = 512 \) THz, as can be seen in Figure 5(c)–(d). For a three-layer structure and right-handed circularly polarized light, the real part of the effective index of refraction is \(-1\) and its imaginary part is 2.18 at the frequency of 471 THz. So, the figure of merit (FOM = \(|\text{Re}(n)/\text{Im}(n)|\)) is 0.46. Also, for the same structure and for left-handed circularly polarization, the real part of the effective index of refraction is \(-1\) and its imaginary part is 2.11 at \( f = 509 \) THz which corresponds to FOM = 0.47. These FOMs are not suitable. This issue relates to the high loss in chiral metamaterials, as mentioned earlier, apart from relatively high metal losses at optical frequencies. Also, the real and imaginary parts of the permeability and the permittivity of the structure with two and three layers of nanodisc crosses are shown in Figures 4(e)–(f) and 5(e)–(f), respectively.

We have studied the proposed structure with four and five layers of nanodisc crosses and found that for a four-layer structure, the chirality coefficient is less than the three-layer case and more than the two-layer case. In the four-layer case, only one of the circular polarization has negative index of refraction. In the case of five-layer structure, the chirality coefficient is reduced further, such that none of the two circularly polarization has negative index of refraction. The explanation for these observations is that if the number of layers is increased beyond a number (herein 3), the structure asymmetry is weakened and the structure becomes more symmetric. This results in decreasing the chirality coefficient. Therefore, increasing the number of layers beyond three, makes the structure more complex and also corresponds to weaker results. For example, a five-layer structure is shown in Figure 6. To clarify the above explanation, the middle dielectrics are not shown in the figure.

In the case of closely spaced nanoparticles, these nanoparticles can be modelled as electric point dipoles (23). On the other hand, the surface charges are related to normal components of electric fields. So, for better understanding the excitation of the longitudinal modes corresponding to localized surface plasmons, we set some monitors in our simulations to investigate the normal components of electric fields. As an example, for the structure with two layers of nanodisc crosses, the real part of the normal component of electric field on the two nanodisc crosses are sketched in Figure 7, for LCP illumination at its resonant frequency, \( f = 508 \) THz.

At last, we make a comparison between the proposed structure and a conventional structure. The conventional structure is similar to our proposed structure but, it has continuous strips instead of arrays of aligned nanodiscs. The results of this comparison are described in detail in a supplementary file. In summary, the resonant frequency of the conventional structure cannot increase beyond a limit. In fact, by shortening the lengths of strips, they gradually act as nanoparticles. In this case, the structure can be considered as an array of nanoparticles with one element. The resonant behaviour of this one-element array is not strong enough to create negative refraction.

4. Conclusion

In this paper, we used the resonant frequency of surface plasmons in nanoparticles to shift the resonant frequency of metamaterial based on bilayer cross wires (6) to optical domain. In fact, we used crosses of nanodiscs instead of continuous wires. Our proposed chiral metamaterial provides significant optical activity, large circular dichorism and also negative refractive index in the optical spectrum. First, we realized the proposed structure by two layers of nanodisc crosses but, only strong circular dichorism and optical activity were obtained and the results about the refractive index were not remarkable. When we added another layer, negative refractive index was achieved too. In a thickness of only 100 nm, our proposed structure provides the same rotary power as a gyrotropic crystal of quartz of 1-mm thickness. The foundation of the proposed structure to obtain negative refractive index is based on the creation of different coupling between longitudinal modes of localized surface plasmons under the incidence of RCP and LCP waves. Meanwhile, we developed a formula for the twist angle of the nanodisc crosses between adjacent layers under which the maximum chirality is achieved. Also, we mentioned some points for changing the resonant frequency of a chiral metamaterial in a wide range of optical spectrum.

Acknowledgements

The authors would like to thank Miss Fahimeh Armin for her valuable comments.

Disclosure statement

No potential conflict of interest was reported by the authors.

References

(1) Engheta, N.; Ziolkowski, R. *IEEE Trans. Microwave Theory Tech.* **2005**, *53*, 1535–1556.
(2) Pendry, J.B.; Schurig, D.; Smith, D.R. *Science* **2006**, *312*, 1780–1782.
(3) Tretyakov, S.; Nefedov, I.; Sihvola, A.; Maslovski, S.; Simovski, C.J. *Electromagn. Waves Appl.* **2003**, *17*, 695–706.
(4) Pendry, J.B. *Science* **2004**, *306*, 1353–1355.
(5) Plum, E.; Zhou, J.; Dong, J.; Fedotov, V.A.; Koschny, T.; Soukoulis, C.M.; Zheludev, N.I. *Phys. Rev. B* **2009**, *79*, 035407–035412.
(6) Zhou, J.; Dong, J.; Wang, B.; Koschny, T.; Kafesaki, M.; Soukoulis, C.M. *Phys. Rev. B* **2009**, *79*, 121104–121107.
(7) Wu, Z.; Zeng, B.Q.; Zhong, B. *J. Electromagn. Waves Appl.* **2010**, *24*, 983–992.
(8) Zari, D.; Soleimani, M.; Naryeri, V. *J. Electromagn. Waves Appl.* **2012**, *26*, 251–263.
(9) Kenanakis, G.; Zhao, R.; Stavrinidis, A.; Konstantinidis, G.; Katsarakis, N.; Kafesaki, M.; Soukoulis, C.M.; Economou, E.N.; *Opt. Mater. Express* **2012**, *2*, 1702–1712.
(10) Zhukovsky, S.V.; Chigrin, D.N.; Kremers, C.; Lavrinenko, A.V. *Photonics Nanostruct.* **2013**, *4*, 353–361.
(11) Kenanakis, G.; Zhao, R.; Katsarakis, N.; Kafesaki, M.; Soukoulis, C.M.; Economou, E.N. *Opt. Express* **2014**, *22*, 12149–12159.
(12) Decker, M.; Klein, M.W.; Wegener, M.; Linden, S. *Opt. Lett.* **2007**, *32*, 856–858.
(13) Ruther, M.; Krieger, C.E.; Zhou, J.; Soukoulis, C.M.; Linden, S.; Wegener, M. *Opt. Lett.* **2009**, *34*, 2501–2503.
(14) Plum, E.; Fedotov, V.A.; Schwancke, A.S.; Zheludev, N.I.; Chen, Y. *Appl. Phys. Lett.* **2007**, *90*, 223113–223115.
(15) Decker, M.; Zhao, R.; Soukoulis, C.M.; Linden, S.; Wegener, M. *Opt. Lett.* **2010**, *35*, 1593–1595.
(16) Giloan, M.; Astilean, S. *Opt. Commun.* **2014**, *315*, 122–129.
(17) Fang, F.; Gao, L.; Liao, H. *J. Modern Opt.* **2016**, *63*, 190–194.
(18) Shin, H.; Fan, S. *Phys. Rev. Lett.* **2006**, *96*, 073907–073910.
(19) Verhagen, E.; Waele, R.; Kuipers, L.; Polman, A. *Phys. Rev. Lett.* **2010**, *105*, 223901–223904.
(20) Dionne, J.A.; Verhagen, E.; Polman, A.; Atwater, H.A. *Opt. Express* **2008**, *16*, 19001–19017.
(21) Alu, A.; Salandrino, A.; Engheta, N. *Opt. Express* **2006**, *14*, 1557–1567.
(22) Johnson, P.B.; Christy, R.W. *Phys. Rev. B* **1972**, *6*, 4370–4379.
(23) Maier, S. *Plasmonics: Fundamentals and Applications*; Springer, New York, 2007.
(24) Monzon, J.C. *IEEE Trans. Antennas Propag.* **1990**, *38*, 227–235.
(25) Pendry, J.B. *Phys. Rev. Lett.* **2000**, *85*, 3966–3969.
(26) Lindell, I.V.; Sihvola, A.H.; Tretyakov, S.A.; Viitanen, A.J. *Electromagnetic Waves in Chiral and Bi-isotropic Media*; Artech House Publishers, Boston, 1994.
(27) Jackson, J.D. *Classical Electrodynamics*3rd ed.; Wiley, New York, 1998.
(28) Gennes, P.G. *The Physics of Liquid Crystals*; Clarendon Press, Oxford, 1974.
(29) Zhao, R.; Koschny, T.; Soukoulis, C.M. *Opt. Express* **2010**, *18*, 14553–14567.