Tunable electromagnetic chirality induced by graphene inclusions in multi-layered metamaterials

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We theoretically investigate the electromagnetic response of a novel class of multi-layered metamaterials obtained by alternating graphene sheets and dielectric layers, the whole structure not exhibiting a plane of reflection symmetry along the stacking direction. We show that the electromagnetic response of the structure is characterized by a magneto-electric coupling described by an effective chiral parameter. Exploiting the intrinsic tunability of the graphene-light coupling, we prove that one can tune both the dielectric and the chiral electromagnetic response by varying the graphene chemical potential through external voltage gating.

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Graphene, a one-atom thick layer of carbon atoms arranged in a honeycomb lattice, shows a wide range of unique properties [1]. For example, graphene exhibits high thermal and electric conductivity, high optical damage threshold and high third-order optical nonlinearities [2]. Recently, many graphene-based photonic and optoelectronic devices have been proposed and developed such as plasmonic waveguides [3–5], frequency multipliers [6], modulators [7], photodetectors [8] and polarizers [9]. In the context of metamaterials, A. Vakil et al. [10] have theoretically proposed a setup where a graphene sheet is a one-atom-thick platform for achieving the desired infrared metamaterials and transformation optical devices. On the other hand, several researchers have investigated multilayer structures composed of stacked graphene sheets separated by thin dielectric layers [11–14]. A newsworthy advantage of such proposed metamaterials is the overall tunability of the electromagnetic response which is entailed by the dependence of the graphene conductivity on the chemical potential. For example, the graphene-based metamaterial response can be tailored from elliptic birefringent to hyperbolic by varying the graphene chemical potential through an external gate voltage [12].

In this Letter, we propose a novel class of graphene-based metamaterials exhibiting a marked chiral electromagnetic response and we demonstrate that such nonlocal effect can be tuned by varying the chemical potential of graphene sheets. More precisely, we consider propagation of transverse magnetic (TM) waves through a multilayer periodic structure not exhibiting a plane of reflection symmetry whose unit cell comprises $N$ layers of different dielectric materials alternated with $N$ graphene sheets. Exploiting a suitable multiscale approach where the period to wavelength ratio is the small expansion parameter, we obtain the constitutive equations describing the spatially nonlocal metamaterial response. Specifically we refine the standard effective medium theory by deriving higher order contributions predicting, in particular, an overall medium chiral response for those layer thicknesses not fully assuring homogenization. Generally, a reciprocal or chiral magneto-electric coupling is a consequence of the medium 3D or 2D chirality, namely the underlying constituents (organic molecules, proteins, ”meta-molecules”, etc.) exhibit mirror asymmetry [15]. Chirality can produce newsworthy effects such as optical rotation and negative refraction [16]. On the other hand, the configuration we consider in this Letter has a 1D chirality which is in turn tunable due to the presence of the graphene sheets. It is worth stressing that usually considered bilayer metal-dielectric structures [17,18] and graphene-based metamaterials (considered in Refs. [12–14] where the metamaterial unit cell consists of a graphene sheet placed on top of a dielectric material) show electromagnetic response strongly affected by second order spatial dispersion which, however, does not yield electromagnetic chirality since the structure geometry admits plane of mirror symmetry.

Let us consider TM waves propagating in a graphene-based metamaterial whose underlying multilayered structure has a unit cell obtained by stacking along the $z$-axis, $N$ graphene sheets separated by $N$-layers of different media of thicknesses $d_j$ ($j = 1, 2, 3, ..., N$) (see Fig.1 where the case $N = 2$ is reported). The electromagnetic field amplitudes $\mathbf{E} = E_x(x,z)\hat{\mathbf{e}}_x + E_z(x,z)\hat{\mathbf{e}}_z$, $\mathbf{H} = H_y(x,z)\hat{\mathbf{e}}_y$ associated with monochromatic TM waves (the time dependence $\exp(-i\omega t)$ has been assumed where $\omega$ is the angular frequency) satisfy Maxwell’s equations

$$
\partial_z E_x - \partial_x E_z = i\omega \epsilon_0 \mu_0 H_y,
$$
$$
\partial_z H_y = i\omega \epsilon_0 \epsilon_x(z) E_x,
$$
$$
\partial_z H_y = -i\omega \epsilon_0 \epsilon_z(z) E_z,
$$

(1)

where $\epsilon_x$ and $\epsilon_z$ are the $x$-component and the $z$-
component of the dielectric permittivity tensor, respectively and both are periodic functions of period \( d = \sum_{j=1}^{N} d_j \). Here, the \( j \)-th graphene sheet response is described by the surface conductivity \( \sigma_j \) so that the surface current \( K_{xz} = \sigma_j E_z \) yields a delta-like contribution to \( \varepsilon_x \) which is \( i\sigma_j/(\omega\varepsilon_0)\delta(z - z_j) \), \( z_j \) being the sheet position \([13]\) (see below). Note that the surface conductivities \( \sigma_j \) can assume different values in order to encompass the relevant situation where the graphene can be locally tuned or substituted with more general bi-dimensional hetero-structures.

In order to obtain an effective medium description of the electromagnetic propagation in the regime where the ratio between the period \( d \) and the wavelength \( \lambda \) is small, we exploit a standard and rather general multiscale technique \([19, 20]\) holding for very general \( \varepsilon_x(z) \) and \( \varepsilon_z(z) \) periodic profiles (which we will later specialize to considered graphene-based multi-layer). Accordingly we introduce the parameter \( \eta = d/\lambda \ll 1 \) and the fast coordinate \( Z = z/\eta \) and, aimed at isolating the slowly and rapidly varying contributions, we consider the Fourier series of \( \varepsilon_x \) and \( \varepsilon_z^{-1} \), namely \( \varepsilon_x = \langle \varepsilon_x \rangle + \delta \varepsilon_x, \varepsilon_z^{-1} = \langle \varepsilon_z^{-1} \rangle + \delta \varepsilon_z^{-1} \) where \( \langle f \rangle \) is the mean value of the function \( f \) and

\[
\delta \varepsilon_x = \sum_{n \neq 0} a_n \exp \left( \frac{2\pi i n x}{d} \right) = \sum_{n \neq 0} a_n \exp (ink_0 Z),
\]
\[
\delta \varepsilon_z^{-1} = \sum_{n \neq 0} b_n \exp \left( \frac{2\pi i n z}{d} \right) = \sum_{n \neq 0} b_n \exp (ink_0 Z).
\]

where \( k_0 = 2\pi/\lambda \). The basic Ansatz of our approach is given by

\[
E_x(x, z, Z) = \bar{E}_x(x, z) + \eta \delta E_x(x, z, Z),
\]
\[
E_z(x, z, Z) = \bar{E}_z(x, z) + \delta E_z(x, z, Z),
\]
\[
H_y(x, z, Z) = \bar{H}_y(x, z) + \delta H_y(x, z, Z),
\]

where \( \bar{A}(x, z) \) and \( \delta A(x, z, Z) \) are the slowly (averaged) and rapidly varying part of each electromagnetic field component \( A \), respectively \( (A = E_x, E_z, H_y) \). The considered Ansatz, where each field component is a Taylor expansion up to first order in \( \eta \), has been suitably chosen to self-consistently assure that finite and not trivial results are obtained in the asymptotic \( \eta \to 0 \) limit. Substituting the Fourier series of \( \varepsilon_x \) and \( \varepsilon_z^{-1} \) and the Ansatz of Eqs.(3) into Maxwell equations (1), after separating the slowly and rapidly varying contributions, we obtain the coupled equations

\[
\partial_z \bar{E}_z - \partial_z \bar{E}_x = i\omega \mu_0 \bar{H}_y,
\]
\[
\partial_z \bar{H}_y = i\omega \varepsilon_0 \left( \langle \varepsilon_x \rangle \bar{E}_z + \eta \langle \delta \varepsilon_x \delta E_z \rangle \right),
\]
\[
\bar{E}_z = \frac{1}{\omega \varepsilon_0} \left( \langle \varepsilon_z^{-1} \rangle \partial_z \bar{H}_y + \eta \langle \delta \varepsilon_z^{-1} \delta \varepsilon_x \partial_z \bar{H}_y \rangle \right)
\]

and

\[
\partial_Z \delta E_x - \partial_Z \delta E_z = 0,
\]
\[
\partial_Z \delta H_y = i\omega \varepsilon_0 \delta \varepsilon_x \bar{E}_z, \delta E_z = \frac{\omega \mu_0}{\varepsilon_0} \delta \varepsilon_z^{-1} \partial_Z \bar{H}_y.
\]

It is important stressing that no terms have been neglected when deriving Eqs.(4) whereas only the leading contributions (the lowest powers of \( \eta \)) has been retained to obtain Eqs.(5). After integration on \( Z \) and using Eqs.(2), Eqs.(5) yield the rapidly varying parts of the field amplitudes as functions of the slowly ones, i.e.

\[
\delta E_x = \frac{1}{k_0 \omega \varepsilon_0} \sum_{n \neq 0} b_n e^{i k_0 n Z} \bar{E}_z^2 \delta \bar{H}_y,
\]
\[
\delta E_z = -\frac{1}{i \omega \varepsilon_0} \sum_{n \neq 0} b_n e^{i k_0 n Z} \partial_z \bar{E}_z \bar{H}_y,
\]
\[
\delta H_y = \frac{\omega \mu_0}{k_0} \sum_{n \neq 0} \bar{a}_n e^{i k_0 n Z} \bar{E}_z.
\]

Finally, substituting Eqs.(6) into Eqs.(4), we get

\[
\partial_z \bar{E}_z - \partial_z \bar{E}_x = i\omega \mu_0 \bar{H}_y,
\]
\[
\partial_z \bar{H}_y = i\omega \varepsilon_0 \left( \langle \varepsilon_x \rangle \bar{E}_z - \chi^{(\text{eff})}\frac{Z_0}{k_0^2} \partial_z^2 \bar{H}_y \right),
\]
\[
\partial_z \bar{H}_y = -i\omega \varepsilon_0 \left( \langle \varepsilon_x \rangle \bar{E}_z + \frac{\chi^{(\text{eff})}}{k_0^2} \partial_z \bar{E}_z \right),
\]

where \( Z_0 = \sqrt{\mu_0/\varepsilon_0} \) is the vacuum impedance, and

\[
\chi^{(\text{eff})} = \frac{\eta \varepsilon_0}{\mu_0} \sum_{n \neq 0} \frac{a_n}{n} b_n.
\]

It is evident that in the limit \( \eta \to 0 \) the parameter \( \chi^{(\text{eff})} \) vanishes and the multiscale approach considered in this Letter reproduces the results of the well known standard effective medium theory (EMT) \([17]\). Furthermore, it is worth noting that in the case where the structure admits mirror symmetry with respect a specific plane \( z = z_0 \), i.e. the relations \( \varepsilon_x(z) = \varepsilon_x(z + 2z_0) \) and \( \varepsilon_z(z) = \varepsilon_z(z + z_0) \) hold, it is straightforward proving that the dielectric Fourier coefficients are such that

\[
a_{-n} = \exp(i2\pi n z_0/d) a_n \text{ and } b_{-n} = \exp(i2\pi n z_0/d) b_n.
\]
that \( a_n b_n = a_n b_{-n} \) and the series of Eq.(8) provides a vanishing \( \chi^{(eff)} \). Therefore, the slowly varying and leading electromagnetic field can experience the effect of the novel terms proportional to \( \chi^{(eff)} \) in the effective Maxwell equations of Eq.(7) only if the multi-layer does not exhibit an inversion center i.e. if it is chiral. Comparing the second and the third of Eqs.(7) with the standard equations \( \partial_x H_y = i \omega D_z, \partial_z H_y = -i \omega q D_z \) and using the third of Eqs.(7) to substitute for the magnetic field derivative we obtain

\[
\begin{align*}
\bar{D}_x &= \epsilon_0 \left( \frac{\epsilon_z^{(eff)}}{k_0} E_z - \frac{\chi^{(eff)} k_0^2}{\epsilon_z^{(eff)}} \partial_x^2 E_z \right), \\
\bar{D}_z &= \epsilon_0 \left( \frac{\epsilon_z^{(eff)}}{k_0} E_z + \frac{\chi^{(eff)}}{k_0} \partial_x E_z \right).
\end{align*}
\]

which are the structure effective constitutive relations. Note that Eqs.(9) contain term proportional to the first and second \( x \)-spatial derivative of the field components, term usually arising when dealing with weakly spatially nonlocal medium. Exploiting the fact that the effective Maxwell’s equations are invariant with respect to transformation \( \bar{D}_x = D_x - \partial_x Q, \bar{D}_z = D_z + \partial_z Q \) and \( H_y = \bar{H}_y - i \omega Q \) (where \( Q(x, z) \) is an arbitrary function), after setting \( Q = -\epsilon_0 \chi^{(eff)} / k_0 E_z \), we obtain the equivalent effective constitutive relations

\[
\begin{align*}
\bar{D}_x' &= \epsilon_0 \left( \left( \frac{\epsilon_z^{(eff)}}{k_0} + \chi^{(eff)} \right) E_z - \frac{\chi^{(eff)} k_0^2}{\epsilon_z^{(eff)}} \partial_x^2 E_z \right) + \frac{i \chi^{(eff)}}{c} \bar{H}_y', \\
\bar{D}_z' &= \epsilon_0 \epsilon_z^{(eff)} E_z, \\
\bar{B}_y &= \frac{\epsilon_0}{c} \bar{H}_y - \frac{i \chi^{(eff)}}{c} E_z.
\end{align*}
\]

Therefore, in the limit \( \eta \ll 1 \) of quasi-homogenization regime, the effect of the multi-layer mirror asymmetry can be reinterpreted to yield an effective chiral magneto-electric coupling (through the terms proportional to \( \chi^{(eff)} \) in Eqs.(10)), a correction \( \chi^{(eff)} \) to the \( x \)-component of the dielectric permittivity and a second order dispersion effect (see terms proportional to the second-order derivatives of electric field in the first of Eqs.(10)).

In the contexts of chiral multilayers, graphene can play a twofold significant role since its sheets can turn a standard achiral structure into a chiral one whose response, described by the above theory, can be further tuned by varying the graphene chemical potential. In order to discuss this point, we consider a bilayer structure whose unit cell comprises two dielectric layers separated by a graphene sheet. The dielectric permittivities of such structure can be written, within the unit cell \( 0 \leq z < d \) as \( \epsilon_x = \Xi(z) + \frac{\delta(z - d_1)}{\epsilon_0} \) and \( \epsilon_z = \Xi(z) \) where \( \Xi(z) = \cos \left( \frac{\pi x}{a} \right) \) and \( \Xi(z) = \cos \left( \frac{\pi x}{a} \right) \) is the rectangular function \( \Pi(z) = 0 \) if \( \vert z \vert > 1/2, \Pi(z) = 1/2 \) if \( \vert z \vert = 1/2, \Pi(z) = 1 \) if \( \vert z \vert < 1/2, \delta(z) \)

is the Dirac delta function and \( \sigma_1 \) is the surface conductivity of the graphene layer. In this model, the graphene sheet is infinitesimally thin and the current it supports is along the \( x \)-direction thus solely affecting the bilayer \( x \)-component of the permittivity tensor. Note that the considered structure is chiral since the permittivity component \( \epsilon_z(z) \) does not show along the stacking direction a plane of mirror symmetry and it is worth stressing that this due to the graphene sheets. The structure effective parameters are easily evaluated and are

\[
\begin{align*}
\epsilon_x^{(eff)} &= \frac{1}{\delta} \left( d_1 \epsilon_1 + d_2 \epsilon_2 + \frac{\delta}{\epsilon_0} \right), \\
\epsilon_z^{(eff)} &= d \left( \frac{\epsilon_1}{\epsilon_2} + \frac{\epsilon_2}{\epsilon_1} \right)^{-1}, \\
\chi^{(eff)} &= \frac{i d_1 d_2^2 \delta^{(eff)}_{z}}{2 \epsilon_0 \epsilon_d^2 (\epsilon_z^{(eff)} \epsilon_1 \left( \frac{1}{\epsilon_2} - \frac{1}{\epsilon_1} \right),
\end{align*}
\]

where the expression of \( \chi^{(eff)} \) is obtained after the straightforward summation of the series in Eq.(8). Evidently \( \chi^{(eff)} \) vanishes if there is no graphene \( (\sigma_1 = 0, \delta \text{ discussed in Refs. [17,18]} \) or if the dielectrics are identical \( (\epsilon_1 = \epsilon_2 \text{ discussed in Refs. [12--14]} \) since in both situations the structure is achiral. In the following numerical examples, we choose the wavelength \( \lambda = 10.71 \mu m \) and the layer dielectric permittivities \( \epsilon_1 = -1.87 + 0.16i, \epsilon_2 = 2.25 \text{ associated to silicon carbide (SiC) [21] and PMMA [22]}, \text{respectively. In addition we adopt the semi-classical expression for the graphene conductivity } \sigma_1 \text{ holding if } \vert \mu_c \vert \gg K_b T \text{ (} K_b \text{ is the chemical potential, } K_b \text{ is the Boltzmann’s constant and } T \text{ is the temperature) and obtained by taking into account the inter- and intra-band contributions (see Eqs.(4) and (5) in Ref. [23])}. \text{Note that the graphene surface conductivity depends on the frequency } \omega, \text{ the chemical potential } \mu_c, \text{ the temperature } T \text{ and the phenomenological scattering rate } \Gamma. \text{ Here we assume } T = 300 K \text{ and } \Gamma = 0.43 \text{ meV. In addition, setting } d_1 = d_2, \text{ the effective permittivity } z \text{-component } \epsilon_z^{(eff)} = -18.43 + i9.60 \text{ and it is not affected by the chemical potential, whereas the effective permittivity } x \text{-component } \epsilon_x^{(eff)} \text{ and the chiral parameter } \chi^{(eff)} \text{ can be tuned by varying the graphene chemical potential through external voltage gating.}

In Fig. 2, we report the real (solid line) and imaginary (dashed line) parts of \( \epsilon_z^{(eff)} \) and \( \chi^{(eff)} \), respectively, as functions of \( \mu_c \) for \( \eta = 1/15 \). The tunability of the overall electromagnetic response is evident and it also remarkable that in this case a transition from hyperbolic behavior to anisotropic negative dielectric one occurs: the real part of the \( x \)-component of the dielectric permittivity \( \epsilon_x^{(eff)} \) is positive in the region 0.1 eV < \( \mu_c < 0.2 \) eV (shadow area in Fig.2(a)) and it is negative in the region \( \mu_c > 0.2 \) eV, whereas \( \Re(\epsilon_z^{(eff)}) \) is negative everywhere.

In order to check and discuss the predictions of our multiscale approach, we here consider the scattering process of TM waves by a graphene-based metamaterial slab lying in the region 0 < \( z < L \) which, using the standard transfer matrix-method, admits full analytical description. Accordingly we evaluate the exact optical transfer function defined as \( OTF = H_y^{(i)} / H_y^{(i)} \) where \( H_y^{(i)} \),
In conclusion we have shown that a multilayer structure not exhibiting mirror symmetry along the stack-
ing direction (chiral structure), in the quasi-homogenized regime, provide first-order electromagnetic nonlocal re-
response. In particular, we have argued that graphene sheets suitably inserted within a achiral structure can
turn it into a chiral one whose nonlocal electromagnetic response is tunable through the graphene chemical poten-
tial.

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