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Optical Chirality

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Abstract: The building blocks of life comprise chiral molecular units such as amino acids and sugars, so biomacromolecules formed from these units also exhibit chirality on molecular and supramolecular scales. Chirally sensitive (chiroptical) spectroscopic techniques such as circular dichroism (CD), optical rotatory dispersion (ORD) and Raman optical activity (ROA) are therefore particularly incisive probes of the three-dimensional aspects of biomacromolecular structure and are widely used in biomolecular science [1, 2]. Chiroptical methods typically measure small differences (or dissymmetries) in the interaction of left- and right-circularly polarized light (the chiral probe) with a chiral material. However, the inherent weakness of these existing chiroptical phenomena usually restricts their application to samples at the microgram level. Recently, it has been postulated [3] that under certain circumstances superchiral electromagnetic fields could be produced that display greater chiral asymmetry than circularly polarized plane light waves. We have calculated that such superchiral electromagnetic fields are generated in the near fields of planar chiral metamaterials when we can produce standing chiral waves [4-6].

Key words: Missing

INTRODUCTION

The Maxwell equations for the macroscopic free electromagnetic fields, (without charge and current) are well known. We often write Maxwell's equations in terms of electric and magnetic fields **E**, **B**, **H** and **D**:

$$\nabla \times \mathbf{E} = -\frac{\partial}{\partial t}\mathbf{B}, \quad \nabla \cdot \mathbf{B} = 0, \quad \nabla \times \mathbf{H} = \frac{\partial}{\partial t}\mathbf{D}+\mathbf{J}, \quad \nabla \cdot \mathbf{D} = \rho; \tag{1}$$

These equations, however, are not complete. Six more equations, the constitutive relations, have to be added relating the electric field **E**, the magnetic induction **B**, the displacement field **D** and the magnetic field **H** to each other. These constitutive relations are completely independent of the Maxwell equations; the Maxwell equations involve only the fields and their sources. The constitutive relations are concerned with the equations of motion of the constituents of the medium in an electromagnetic field [7, 8].

We write Maxwell's equations in terms of electric and magnetic fields, **E** and **B**, defined by the non locality definitions of Born-Fedorov [9-12]: The Drude-Born-Fedorov constitutive relations of homogeneous, isotropic chiral media are [8-9]:

$$\mathbf{B} = \mu\left[\mathbf{H} + T^c(\nabla \times \mathbf{H})\right] \quad \mathbf{D} = \epsilon\left[\mathbf{E} + T^c(\nabla \times \mathbf{E})\right], \tag{2}$$

in which permittivity ε, permeability μ and chirality T^c are function of ω. With exp(iωt) implicit and the light velocity c = 1, the Maxwell equations become:

$$\nabla \times \mathbf{E} + i\,\omega\mu[\mathbf{H} + T^c]\,(\nabla \times \mathbf{H})] = 0,$$
$$\nabla \times \mathbf{H} + i\omega\varepsilon\,[\mathbf{E} + T^c\,(\nabla \times \mathbf{E})] = 0; \tag{3}$$

the solution for $k_{L,R}$ is $k_{L,R} = k / (1 \pm kT^c)$. When $kT^c \to 1$, $k > 0$, $T^c > 0$, $k_L = k / 2$ and $k_R = \omega / v_{phR} \to \infty$ when $v_{phR} \to 0$. With this approach we discuss the called circular dichroism CD.

Theory on Chiralityon Circular Dichroism of Light: In this section we present the theoretical framework on explaining optical activity of chiral molecules under circularly polarized light (CPL). Chirality of light, however, has not been under such examination so far. Here we start

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by proposing a quantitative measure of local handedness for arbitrary electromagnetic fields. Based on this quantity, we derive the existence of a superchiral solution to Maxwell's equations that is more effective at distinguishing chiral species of opposite chirality than CPL. Optical chirality: a measure of EM handedness Let's recall the definition of dissymmetry factor in conventional CD experiments:

$$g = \frac{A^+ - A^-}{(A^+ + A^-)/2} \leq 2, \tag{4}$$

where $A\pm$ is the molecular absorption rate in left (+) or right (-) CPL. We seek for a measure of optical chirality that may be embodied in the dissymmetry factor from first principles.

For non-plane wave EM fields, one expects the degree of dissymmetry to depend on local properties of the field. We begin by seeking a quantitative definition of the chirality of a vector field, independent of its interaction with matter. Chirality is time even and we rename "optical chirality":

$$C = \frac{1}{2}(\varepsilon_0(\mathbf{E}\cdot\nabla\times\mathbf{E}) + \mu_0(\mathbf{H}\cdot\nabla\times\mathbf{H})), \tag{5}$$

where ε_0 and μ_0 are the permittivity and permeability of free space, respectively and **E** and **H** are the time-dependent electric and magnetic fields. We see that the quantity, optical chirality, completes the fundamental measures in EM theory of scalar properties of energy density:

$$U = \frac{1}{2}(\varepsilon_0(\mathbf{E}\cdot\mathbf{E}) + \mu_0(\mathbf{H}\cdot\mathbf{H})). \tag{6}$$

Optical chirality is a time-even pseudo-scalar; ε_0, permittivity of free space; μ_0, permeability of free space.

Optical Chirality in the Interaction with Matter: Is optical chirality observable? In the standard theory of CD the dissymmetry factor, λ, measures the fractional difference in rates of excitation between left- and right-circularly polarized light at wavelength g(λ). In the section, we generalized the theory of CD to include pairs of arbitrary mirror-image fields. The response of a molecule to an EM perturbation may be calculated and based on a response function-based description We restrict attention to isotropic samples, for which the response tensors may be replaced by scalar response functions. The restriction to isotropic samples is necessary because in oriented samples even achiral molecules may show optical activity.

A chiral molecule subjected to monochromatic EM fields generates an electric dipole moment, p and a magnetic dipole moment, m, given by:

$$\bar{\mathbf{p}}(t) = \alpha\bar{\mathbf{E}} - T^c\nabla\times\bar{\mathbf{E}}, \quad \bar{\mathbf{m}} = \chi\bar{\mathbf{H}} + T^c\nabla\times\bar{\mathbf{H}}; \tag{7}$$

Here, we use equations (2) and (3) of Born-Fedorov relationship. Quantities with a - are complex, e.g., $\bar{\alpha} = \alpha' + i\alpha''$ and so on. The electric polarizability, magnetic polarizability and optical rotatory strength are given by $\bar{\alpha}$, $\bar{\chi}$ and $\bar{T^c}$, respectively. In equation (7), one should take the real part of each side to obtain physical quantities. In the most general time-periodic EM field, the electric and magnetic fields each describe an ellipse, with arbitrary relative phase and orientation. We consider a pair of fields with harmonic time dependence, from which we can build an arbitrary solution by Fourier superposition; the fields are interchanged by application of Parity:

$$\bar{\mathbf{E}}(t) = \pm\mathbf{e}_0\exp(-i\omega t), \quad \bar{\mathbf{H}}(t) = \mathbf{h}_0\exp(-i\omega t), \tag{8}$$

where again the real parts of $\bar{\mathbf{E}}(t)$ and $\bar{\mathbf{H}}(t)$ denoted **E** and **H**, describe the physical quantities, $\mathbf{e}_0$ and $\mathbf{h}_0$ are arbitrary complex vectors. **E** is odd under parity while **H** is even.

The rate of excitation of the molecule is:

$$A^\pm = \langle \mathbf{E}\cdot d\mathbf{p}/dt + \mathbf{H}\cdot d\mathbf{m}/dt \rangle = \frac{\omega}{2}\mathrm{Im}(\bar{\mathbf{E}}^*\cdot\bar{\mathbf{p}} + \bar{\mathbf{H}}^*\cdot\bar{\mathbf{m}}), \tag{9}$$

Such that the brackets indicate an average over time and * means the complex conjugate. Expanding the rate of excitation using eq. (7) we have:

$$A^\pm = \frac{\omega}{2}\mathrm{Im}(\alpha''|\bar{\mathbf{E}}|^2 + \chi''|\bar{\mathbf{H}}|^2 \pm T^{c}{}''\omega\,\mathrm{Im}\,\bar{\mathbf{E}}^*\cdot\bar{\mathbf{H}}), \tag{10}$$

where the term involving χ is negligibly small for most molecules and so we henceforth neglect it. We apply the identity $\omega\,\mathrm{Im}\bar{\mathbf{E}}^*\cdot\bar{\mathbf{H}}) = \mathbf{E}\cdot d\mathbf{H}/dt - \mathbf{H}\cdot d\mathbf{E}/dt$ to the term containing $T^{c''}\omega$ with $T^{c''} << T^{c'}$ and employing the Maxwell's equations in free space, we find that $A^\pm = \frac{2}{\varepsilon_0}(\omega U_e\alpha'' \mp CT^{c}{}''\omega)$, being $U_e = (\varepsilon_0/4)|\bar{\mathbf{E}}|^2$ the time-average electric energy density and C is defined in eq. (5).

For a monochromatic CPL plane wave, the dissymmetry factor is defined as $g = 2(A^+ - A^-)/A^+ + A^-$, where $A^{\pm}$ is the absorption rate in left (+) or right (-) CPL. We now generalize the definition of g to include any pair of EM fields interchanged by parity, whereupon we find:

$$g == \frac{T^{c}"}{\alpha"} \frac{2C}{U_e}; \qquad (11)$$

We note that when $\omega T^{c"} = G"$, we obtain the result of [3-6, 14, 15] given by:

$$g = \frac{G"}{\alpha"} \frac{2C}{\omega U_e}. \qquad (12)$$

Superchiral Solution to Maxwell's Equations: We may rewrite the general dissymmetry factor in equation (12) by incorporating the parameters $\alpha"$ and $G" = \omega T^{c"}$ into g_{CPL}, where g_{CPL} is the dissymmetry factor under circularly polarized light:

$$g = g_{CPL} \frac{cC}{2\omega U_e}; \qquad (13)$$

therefore, the search for superchiral solution to Maxwell's equations means that exist fields for which $|C| > 2U_e\ \omega/c$ in some regions of space, i.e., with greater chirality than that of a circularly polarized plane wave. We are here to present one "superchiral" solution to Maxwell's equations with surprising chiroptical properties.

The fields are everywhere circularly polarized, but modulated in amplitude in a standing wave pattern which is obtained from Sec. 2 with **E** ‖ **H**. The remarkable finding is that, as the difference in the A+ and A- amplitudes approaches zero, the chiral asymmetry at the nodes diverges. This divergence arises because, at a node, U_e goes to zero as $(E_1 - E_2)^2$, while *C* goes to zero as $(E_1^2 - E_1^2)$; the ratio C/U_e is thus proportional to $(E^1 + E_2) / (E_1 - E_2)$.

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