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Vázquez-Lozano, JE.; Martínez Abietar, AJ. (2018). Optical Chirality in Dispersive and Lossy Media. *Physical Review Letters*. 121(4):043901-1-043901-7.  
<https://doi.org/10.1103/PhysRevLett.121.043901>



The final publication is available at

<http://doi.org/10.1103/PhysRevLett.121.043901>

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# Optical Chirality in Dispersive and Lossy Media

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(Dated: May 15, 2018)

Several dynamical properties such as energy, momentum, angular momentum, and optical helicity have been recently reexamined in dispersive and lossless media. Here, we address a parallel derivation for the optical chirality, extending it so as to include dissipative effects as well. To this end, we first elaborate on the most complete form of the conservation law for the optical chirality, without any restrictions on the nature of the medium. As a result we find a general expression for the optical chirality density both in lossless and lossy dispersive media. Our definition is perfectly consistent with that originally introduced for electromagnetic fields in free space, and is applicable to any material system, including dielectrics, plasmonic nanostructures, and left-handed metamaterials.

*Introduction.*—Local dynamical properties such as energy, linear momentum, and angular momentum, among others, are conserved quantities for electric and magnetic fields in vacuum [1, 2]. In fact, leaving aside the physical meaning, there exists an infinity class of conserved quantities for free electromagnetic fields [3, 4]. In particular, in 1964 Lipkin demonstrated the existence of a set containing ten new independent conservation laws for electromagnetic radiation in vacuum [5]. Originally, these tensorial quantities were merely conceived as mathematical entities theretofore unknown, and having no ready physical significance. That is why they were collectively referred to as the *ij-zilches* (which literally means “nothingness”), where *i* and *j* stand for the labels indicating the tensor indices. Since then, there have been many efforts in searching for a physically meaningful picture for these quantities [6–8].

Recent advances in near-field optics attempting to achieve full spatiotemporal control of light-matter interactions [9] has led to a renewed interest in Lipkin’s zilches as a measure of the handedness, or knottedness, of the streamlines describing highly contorted optical fields [10]. In this regard, and motivated by the possibility for enhancing the chiroptical effects (such as circular dichroism (CD) [11]), which leads to enantioselective signals far larger than that due to circularly polarized light (CPL), Tang and Cohen introduced the 00-zilch as a measure of the local density of optical chirality [12]:

$$C_{\text{vacuum}} \equiv \frac{1}{2} [\varepsilon_0 \mathcal{E} \cdot (\nabla \times \mathcal{E}) + \mu_0 \mathcal{H} \cdot (\nabla \times \mathcal{H})], \quad (1)$$

where  $\varepsilon_0$  and  $\mu_0$  are the permittivity and permeability of vacuum, respectively, and  $\mathcal{E}(\mathbf{r}, t)$  and  $\mathcal{H}(\mathbf{r}, t)$  are the local, time-dependent electric and magnetic fields. Shortly after, this definition for the optical chirality was successfully used in enhanced CD spectroscopic measurements for the experimental detection and characterization of chiral biomolecules [13], thus confirming its physical significance, and highlighting the feasibility for practical applications. The extremely high sensitivity in the chiroptical responses (enhancement factors up to 6 orders of magnitude were reported)

was attributed to the presence of the incipiently postulated *superchiral fields* [14]. However, as pointed out in Refs. [15, 16], on account of the energy conservation, there should be an upper bound lowering those enhancements. In this respect, it was argued that this fundamental restriction ought to limit the enhancement factor up to two orders of magnitude [15]; the other four orders should come from the highly twisted evanescent near-field modes [16, 17]. It then follows that, essentially, the main requirement for the occurrence of strengthened chiroptical influence in light-matter interaction relies on the complexity in the structure of the electromagnetic field distribution [10, 17, 18]. For this reason, metallic nanostructures represent ideal candidates for investigating chirality-based applications and functionalities in nanophotonics [19–26]. It is certainly surprising, however, that, most of the previous studies on this issue build on the earliest definition for the optical chirality density [5], which is only valid for monochromatic optical fields in free space [12, 14, 16]. Still, there are few works attempting to extend the definition of the optical chirality density to linear [15], gyrotropic [27], or dispersive lossless media [28].

Inspired by the latest theoretical results concerning the dispersive features of the electromagnetic energy-momentum, the optical orbital and spin angular momentum [29–31], and the electromagnetic helicity [32], in this Letter we report on the optical chirality in lossless and lossy dispersive media. Special emphasis is placed on the role of the mathematical structure of the corresponding conservation law. Indeed, building on previous approaches addressing the electromagnetic energy density considering dispersion as well as dissipation [33], we put forward a complete description for the optical chirality conservation law valid for arbitrarily structured optical fields. The only restriction we need to impose relies on the electromagnetic characterization of the medium, which must be fitted by Lorentzian line shapes. Hence, our results are completely general [34], and are applicable to any material system, including dielectrics, semiconductors, metals, as well as metamaterials, even with negative refractive index.

Further, our findings are perfectly consistent with the ones so far established for optical fields in free space [12, 16], thus showing its physical meaningfulness.

*Conservation law for the optical chirality.*— Conservation laws and symmetry properties of a physical system are, arguably, among the most important cornerstones of modern physics [35]. More in-depth insights would require the standard Lagrangian formulation [36]. Indeed, appealing to the principle of least action, the Noether's theorem states that, in the absence of sources (or sinks), conserved quantities and symmetries can be regarded as equivalent features [37]. These theoretical concepts are mathematically described via continuous or discrete symmetry groups, which are in turn related to the corresponding physical transformations [38]. Typical examples of continuous symmetries lead to the conservation of energy, linear momentum and angular momentum, which are associated with the invariance under the universal space-time transformations, i.e., translations and rotations. An insightful picture of the conserved quantities, reminiscent of the quantum formalism [39], allows one to deal with the conserved quantities as differential operators representing the generators of the corresponding infinitesimal symmetry transformations. For the above dynamical properties, the generators simply involve first derivatives with respect to the space-time coordinates acting on the electromagnetic fields, and are given explicitly by  $\{i\partial_t, i\nabla\}$ , for the space-time translations [29], and  $i(\mathbf{r} \times \nabla)$ , for the spatial rotations [30]. Furthermore, it was recently demonstrated that the conservation of the optical chirality is underpinned by  $i(\partial_t \nabla \times)$  [28], which must be applied on the vector potentials rather than on the electromagnetic fields. Importantly, these generators can be used to find the eigenstates of the aforementioned conserved quantities. In this regard, just as the plane waves are the eigenstates of the energy-momentum differential operator, it has been shown that the corresponding eigenstates associated to the optical chirality are the circularly polarized plane waves [28].

The above scheme for identifying continuous conserved quantities only holds in the absence of sources. In the presence of charges and/or currents, conservation laws are to be expressed through the continuity equations [40, 41]. Within the electromagnetic field theory, the most well-known example is perhaps the Poynting's theorem, accounting for the energy conservation [1, 2]:

$$\nabla \cdot \mathbf{S} = -[\mathcal{E} \cdot \partial_t \mathcal{D} + \mathcal{H} \cdot \partial_t \mathcal{B} + \mathcal{J} \cdot \mathcal{E}], \quad (2)$$

where  $\mathbf{S} \equiv \mathcal{E} \times \mathcal{H}$  is the Poynting vector, which represent the energy flux density, and  $\mathcal{D}$ ,  $\mathcal{B}$ , and  $\mathcal{J}$  are the time-dependent electric displacement, magnetic induction, and electric current density (or charge flux), respectively. This expression is generally valid, and can be readily obtained by taking the divergence of

the energy flux density. Likewise, we can derive the time-dependent conservation law for the optical chirality from the corresponding chirality flux density [5, 12, 16]:

$$\mathcal{F} \equiv [\mathcal{E} \times (\nabla \times \mathcal{H}) - \mathcal{H} \times (\nabla \times \mathcal{E})]/2. \quad (3)$$

With the aid of the structural Maxwell's equations and the vector identity,  $\nabla \cdot (\mathcal{A} \times \mathcal{B}) = \mathcal{B} \cdot (\nabla \times \mathcal{A}) - \mathcal{A} \cdot (\nabla \times \mathcal{B})$ , it follows that

$$\nabla \cdot \mathcal{F} = -[\mathcal{E} \cdot \partial_t (\nabla \times \mathcal{D}) + \mathcal{H} \cdot \partial_t (\nabla \times \mathcal{B}) + \mathcal{S}_{\mathcal{J}}]/2, \quad (4)$$

where  $\mathcal{S}_{\mathcal{J}} = \mathcal{E} \cdot (\nabla \times \mathcal{J})$  is the current-related source-like contribution. Taking into account the general structure of the continuity equation [33], Eq. (4) can be recast as

$$\nabla \cdot \mathcal{F} + \partial_t \mathcal{C} = \mathcal{S}, \quad (5)$$

where

$$\mathcal{C} \equiv \frac{1}{2} [\mathcal{E} \cdot (\nabla \times \mathcal{D}) + \mathcal{H} \cdot (\nabla \times \mathcal{B})], \quad (6)$$

$$\mathcal{S} \equiv \frac{1}{2} [\partial_t \mathcal{E} \cdot (\nabla \times \mathcal{D}) + \partial_t \mathcal{H} \cdot (\nabla \times \mathcal{B}) - \mathcal{S}_{\mathcal{J}}], \quad (7)$$

are the optical chirality density and the source-like terms, respectively. It is worth remarking that the above expressions represent the most general result for the conservation law of optical chirality, without any restrictions on the nature of the medium, i.e., they are valid regardless of the linearity, homogeneity, isotropy, or dispersion. However, they differ significantly from the previously established continuity equation [5, 12, 16, 26],

$$\nabla \cdot \mathcal{F} + \partial_t \mathcal{C}_{\text{vacuum}} = \mathcal{S}_{\text{vacuum}}, \quad (8)$$

where  $\mathcal{C}_{\text{vacuum}}$  is the optical chirality density as defined in Eq. (1), and  $\mathcal{S}_{\text{vacuum}} \equiv -[\mathcal{J} \cdot (\nabla \times \mathcal{E}) + \mathcal{E} \cdot (\nabla \times \mathcal{J})]/2$  is the source-like term in free space. As discussed in Sec. II of the Supplemental Material [33], the essential discrepancy arises on account of the dispersion-related terms. In particular, it is easy to prove that  $\mathcal{C} = \mathcal{C}_{\text{vacuum}} + \mathcal{C}_{\text{medium}}$ , where  $\mathcal{C}_{\text{medium}} \equiv [\mathcal{E} \cdot (\nabla \times \mathcal{P}) + \mu_0 \mathcal{H} \cdot (\nabla \times \mathcal{M})]/2$ , and  $\mathcal{P}$  and  $\mathcal{M}$  are the macroscopic polarization and magnetization fields. Strikingly, up to our knowledge, these considerations have never been properly analyzed in previous approaches [12, 15, 16, 28, 42]. In fact, even though both the dispersion-related and the dissipation terms are explicitly disregarded in Eq. (8), it has been widely used for investigating chirality and chiroptical effects in media where the permittivity is highly dispersive, including plasmonic nanostructures as well as metamaterials [19–26]. Thus, as shown below, the dispersion of the material systems brings about important corrections into the original expressions for the optical chirality density [compare Eqs. (1) and (6)] and the source-like terms of the continuity equation [compare Eqs. (5) and (8)], and hence, it must be generally considered.

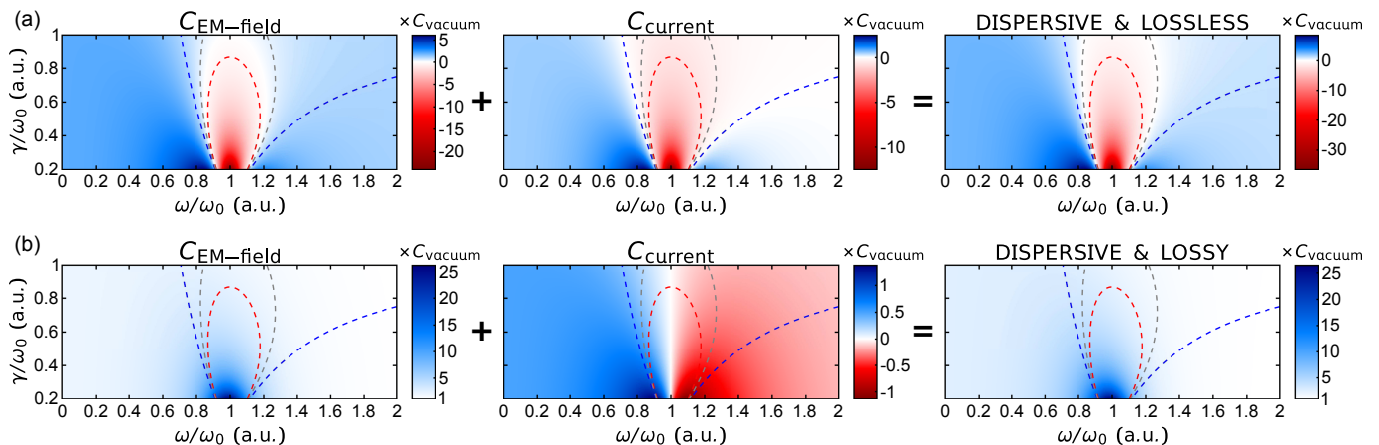


FIG. 1. Optical chirality density in (a) lossless and (b) lossy dispersive media. Material parameters correspond to a nonmagnetic medium ( $\mu = 1$ ) whose permittivity is described by a single Lorentz pole with  $\omega_p = \omega_0$ . In all the cases, red, gray and blue dashed lines indicate the curves where the total contribution of the optical chirality in the lossless case is  $-1$ ,  $0$  and  $1$ , respectively.

*Optical chirality density in lossless dispersive media: Brillouin's approach.*—For monochromatic electric and magnetic fields in free space,  $\mathcal{E}(\mathbf{r}, t) = \text{Re}[\mathbf{E}(\mathbf{r})e^{-i\omega t}]$  and  $\mathcal{H}(\mathbf{r}, t) = \text{Re}[\mathbf{H}(\mathbf{r})e^{-i\omega t}]$ , the time-averaged optical chirality density is given by [12, 16]

$$C_{\text{vacuum}} = \frac{\omega}{2c^2} \text{Im}[\mathbf{E} \cdot \mathbf{H}^*], \quad (9)$$

where bold letters stand for complex field amplitudes and the asterisk denotes complex conjugation. A straightforward calculation allows us to show that, for freely propagating electromagnetic plane waves, the maximum value of  $C$  is achieved for CPL:

$$C_{\text{vacuum}}^{(\pm)\text{CPL}} = \pm \frac{\omega}{2c^2} \frac{1}{Z_0} |\mathbf{E}|^2, \quad (10)$$

where  $Z_0 \equiv \sqrt{\mu_0/\epsilon_0}$  is the vacuum impedance, and the signs  $+$  and  $-$  correspond to left- and right-handed CPL.

In general, CPL is considered as the paradigmatic example of field displaying optical chirality, and has been widely used for chiroptical measurements [13, 14]. Unfortunately, mainly due to the mismatch between the scales of the wavelength of light and the typical size of chiral objects [10], chiral responses are inherently very small [11, 43]. To overcome this drawback, several efforts have been undertaken in the last decades for improving the detection schemes [14, 18, 44, 45], with special emphasis on metallic nanostructures, which are regarded as well suited platforms for strengthening chiroptical light-matter interactions [19–26].

Metals are inherently absorptive and highly dispersive. Something similar happens with semiconductors at energies around the band gap. These features are characterized in terms of the electric permittivity  $\epsilon$  (and eventually with the magnetic permeability  $\mu$ ) depending on the frequency  $\omega$ . According to the Kramer-Kronig relations [1], dispersion is necessarily tied to dissipation.

Thus, in order to avoid misleading outcomes, the analysis of the local dynamical properties have to be carefully carried out from a material standpoint as well. This is well known for the electromagnetic field energy in metals, for which a general treatment has been developed [1, 2]. Indeed, in a lossless dispersive medium the energy density is described by the Brillouin's formula [46, 47]. Following a similar procedure we may then obtain a closed expression for the optical chirality density. For simplicity, we will assume a linear, homogeneous and isotropic medium such that  $\mathbf{D} = \epsilon_0 \epsilon(\omega) \mathbf{E}$  and  $\mathbf{B} = \mu_0 \mu(\omega) \mathbf{H}$ . From the continuity equation as given in Eq. (4), and using the Fourier transforms, the instantaneous distribution of the optical chirality density can be obtained by integrating  $\mathcal{E} \cdot \partial_t (\nabla \times \mathbf{D})$  and  $\mathcal{H} \cdot \partial_t (\nabla \times \mathbf{B})$  over time. It should be noted that the integral convergence is constrained by the *slowly varying amplitude approximation* [2]. Within this assumption, the electric contribution reads as

$$C^{\text{elec}} = \frac{i}{2c^2} \iint \left[ \frac{\omega^2 \epsilon_{\omega} \mu_{\omega}}{\omega' + \omega} \right] \mathbf{E}_{\omega'} \cdot \mathbf{H}_{\omega} e^{-i(\omega' + \omega)t} d\omega' d\omega, \quad (11)$$

where the subscripts denote the frequency dependence. By proceeding in the same way for the magnetic contribution, summing up both expressions, and integrating them properly over the frequencies  $\omega$  and  $\omega'$  [33], we can get the time-averaged optical chirality density in a lossless dispersive media:

$$C_{\text{lossless}} = \text{Re}[n(\omega)\tilde{n}(\omega)] C_{\text{vacuum}} = \frac{\omega}{2} \frac{\text{Im}[\mathbf{E} \cdot \mathbf{H}^*]}{v_p(\omega)v_g(\omega)}, \quad (12)$$

where  $v_p(\omega) \equiv c/\text{Re}[n(\omega)]$  and  $v_g(\omega) \equiv c/\text{Re}[\tilde{n}(\omega)]$ , are the phase and group velocities [48, 49], respectively, which are in turn expressed in terms of the complex-valued phase refractive index  $n(\omega) \equiv \sqrt{\epsilon(\omega)\mu(\omega)}$  and the corresponding dispersion-modified

group index  $\tilde{n}(\omega) \equiv n(\omega) + \omega [\partial n(\omega)/\partial \omega]$ . A detailed description of the above derivation as well as the current-related contribution can be found in Sec. III.B of the Supplemental Material [33].

It should be noted that the same expression for the optical chirality density was previously obtained, but using a more complicated approach (see Eq. (33) in Ref. [28]). Importantly, this definition [Eq. (12)] reduces to the standard result [Eq. (9)] for freely propagating optical fields, i.e., when  $n = 1$ . Furthermore, it is important to emphasize the dependence of Eq. (12) on the dispersion-related phase and group velocities. From this simple relation, it is easy to realize that we may enhance the optical chirality in artificially engineered materials directly by lowering both velocities [48, 49]. This is specifically accomplished in the vicinity of the resonance frequency, i.e., in the anomalous dispersion region [see upper panel of Fig. 1]. However, in a dispersive and lossy media, there are certain frequency ranges where the precise physical meaning of the group velocity turns out to be somewhat unclear [46, 47], and then the result given by Eq. (12) may not be valid.

*Optical chirality density in lossy dispersive media: Loudon's approach.*—A more physically realistic description of dispersive media requires careful considerations of dissipative effects. In this regard, as previously reported (see, e.g., Refs. [50, 51]), the expression for the energy density in dispersive and lossy media crucially depend on the specific model characterizing the medium. This information is enclosed within the material parameters,  $\epsilon$  and  $\mu$  [1, 2].

In classical theory,  $\epsilon$  can be modeled as a collection of Lorentz oscillators [2, 52]:

$$\epsilon_{\text{Drude-Lorentz}}(\omega) = 1 - \sum_n \frac{f_n \omega_p^2}{\omega^2 - \omega_n^2 + i\omega\gamma_n}, \quad (13)$$

where  $f_n$ ,  $\omega_p$ ,  $\omega_n$ ,  $\omega$  and  $\gamma_n$  are, respectively, the relative strength of the oscillators, the plasma frequency, the  $n$ th resonance frequency, the excitation frequency, and the  $n$ th damping constant. This multi-resonant model has been proved to fit very well with the experimental data [34, 53, 54], and thus, it can be regarded as a generic approach for characterizing the electric response of any material system for any frequency and bandwidth. A similar expression can also be introduced for the magnetic permeability  $\mu$ , e.g., when describing negative-index metamaterials consisting of arrays of split-ring resonators or fishnet-like structures [55–57]. It should be noted that the latter expression for  $\epsilon$  follows from the dynamic equation of the polarization field:

$$\frac{\partial^2 \mathcal{P}_n}{\partial t^2} + \gamma_n \frac{\partial \mathcal{P}_n}{\partial t} + \omega_n^2 \mathcal{P}_n = \epsilon_0 f_n \omega_p^2 \mathcal{E}_{\text{loc}}, \quad (14)$$

with  $\mathcal{E}_{\text{loc}}$  being the time-varying external electric field. This relation between the electric and the polarization

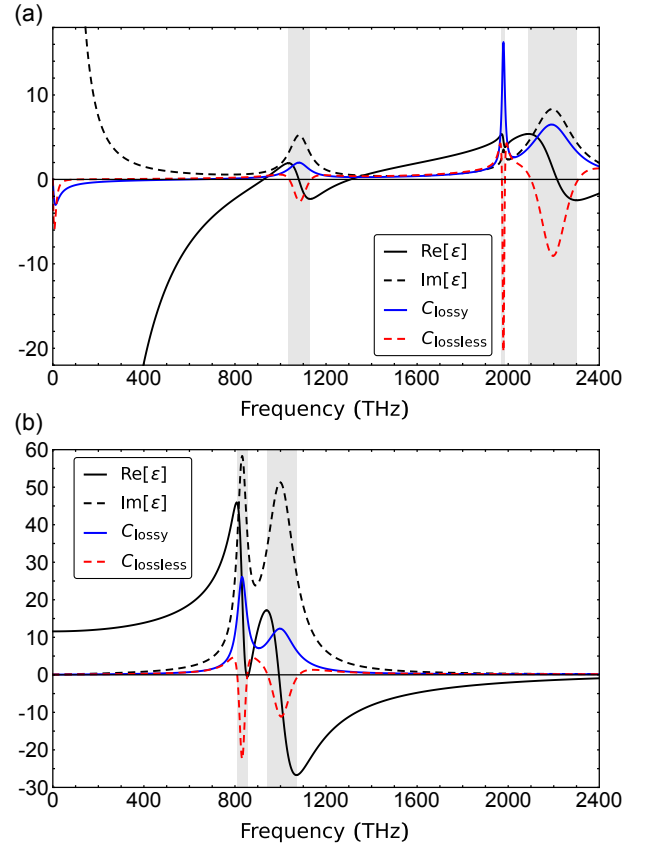


FIG. 2. Optical chirality density for (a) silver [Ag], and (b) silicon [Si]. Material parameters describing the permittivities are taken from Refs. [53] and [54], respectively. For comparison we represent the results for lossless (red dashed lines) [Eq. (12)] and lossy (blue solid lines) [Eq. (17)] dispersive media. The gray shaded areas indicate the spectral ranges with anomalous dispersion behavior.

field is actually the key point to get the general form of the energy density [50, 51]. Likewise, taking into account the underlying mathematical structure of the continuity equation [33], we can use Eq. (14) (and the corresponding one for the magnetization field) to identify the electric (and the magnetic) contribution of the optical chirality density in dispersive and lossy media. To this aim, we start again from the continuity equation as given in Eq. (4). Attempting to find the total time derivative for the electric contribution, we have to express  $\mathcal{E} \cdot \partial_t (\nabla \times \mathcal{D})$  in terms of the electric and the polarization fields (and similarly for the magnetic contribution [33]):

$$\mathcal{E} \cdot \partial_t (\nabla \times \mathcal{D}) = [\epsilon_0 \mathcal{E} \cdot \partial_t (\nabla \times \mathcal{E}) + \mathcal{E} \cdot \partial_t (\nabla \times \mathcal{P})]. \quad (15)$$

In this way, we can also account for the influence of the medium on the chirality density. In the latter expression, the first term of the right-hand side can be rewritten as

$$\mathcal{E} \cdot \partial_t (\nabla \times \mathcal{E}) = \partial_t [\mathcal{E} \cdot (\nabla \times \mathcal{E})] - \partial_t \mathcal{E} \cdot (\nabla \times \mathcal{E}), \quad (16)$$

thereby leading to a total time derivative plus a residual term. As shown at the end of Sec. III.A



in the Supplemental Material [33], this residual term exactly cancel with the one appearing for the magnetic contribution in vacuum, thus allowing us to recover the usual expression for the optical chirality in free space [Eq. (1)]. On the other hand, the second term in the right-hand side of Eq. (15) can be addressed by using the dynamic equation for the polarization field given in Eq. (14) (see Sec. III.A in the Supplemental Material [33] for further details on this derivation). Following the same procedure for the magnetic contribution and summing up both expressions, we finally find that the time-averaged optical chirality density in a lossy dispersive medium is

$$C_{\text{lossy}} \frac{\omega}{4c^2} \text{Im}[(\varepsilon(\omega)\mu_{\text{eff}}(\omega) + \varepsilon_{\text{eff}}(\omega)\mu^*(\omega)) \mathbf{E} \cdot \mathbf{H}^*]. \quad (17)$$

where  $\varepsilon_{\text{eff}}$  and  $\mu_{\text{eff}}$  are the real-valued effective material parameters, which are defined as

$$\varepsilon_{\text{eff}}(\omega) \equiv 1 + \sum_n (\chi'_n + 2\omega\chi''_n/\gamma_n), \quad (18a)$$

$$\mu_{\text{eff}}(\omega) \equiv 1 + \sum_n (\xi'_n + 2\omega\xi''_n/\tilde{\gamma}_n), \quad (18b)$$

with  $\chi = \chi' + i\chi'' \equiv \varepsilon - 1$  and  $\xi = \xi' + i\xi'' \equiv \mu - 1$  being the electric and magnetic susceptibilities. Furthermore, as pointed out in the Supplemental Material, there are also a current-related contribution which should be included.

As shown in Fig. 1, both of the above approaches yield different results. Indeed, whereas  $C_{\text{lossless}}$  [Eq. (12)] can display both positive and negative values, the total contribution of  $C_{\text{lossy}}$  [Eq. (17)], remains always positive, with a minimum value of  $C_{\text{vacuum}}$  that is reached in the high-frequency limit. The largest discrepancies occur close to the resonance frequency. Still, the peaks for both approaches are almost equal in absolute value. These signatures can also be appreciated in Fig. 2, where we plot the optical chirality density of silver and silicon, as examples of metal and semiconductor, respectively. Both materials have been modeled using Eq. (13) with parameters taken from Refs. [53] and [54]. From the results shown in Fig. 2 it is worth emphasizing that  $C_{\text{lossless}}$  overlaps almost exactly with  $C_{\text{lossy}}$  for all frequencies, except in the vicinity of the region of anomalous dispersion, i.e., where  $d\varepsilon'/d\omega < 0$ . There, the curves turn out to be drastically separated from each other, thereby highlighting the importance of considering dissipative effects.

Equation (17) is the main result of this work. To the best of our knowledge, it provides the most general definition for the optical chirality density in dispersive and lossy media, being applicable to any material system including plasmonic nanostructures [26] and left-handed metamaterials [57]. Yet, our definition differs significantly from the standard formula for optical fields in free space [12, 16], and even from previous suggestions attempting to tackle optical chirality in

dispersive media [28, 42]. As discussed in Sec. III of the Supplemental Material [33], the distinction between our result and those found in the aforementioned approaches essentially arises from considering properly the dynamic response of the time-dependent electromagnetic fields within a dispersive medium. In this regard, it should be noted that the time derivative of the fields  $\mathcal{D}$  and  $\mathcal{B}$ , must be expressed as convolution integrals in the time domain. Furthermore, in this particular case, regarding lossy dispersive media, the mathematical structure of the continuity equation plays a central role in the identification of the optical chirality density as a conserved dynamical property.

*Summary.*— We have carried out a theoretical analysis of the conservation law for the optical chirality. Taking advantage from previous approaches addressing the electromagnetic energy density, we have also provided a parallel derivation for the optical chirality both in lossless and lossy dispersive media. Remarkably, our description is completely general, i.e., is valid for arbitrarily varying radiation fields, and can be applied to any medium, including dielectrics, semiconductors, as well as highly lossy material systems such as metals and metamaterials with negative refractive index. In view of the growing interest on chirality and chiral light-matter interaction, we hope that these results will aid the development of plasmonic and metamaterial nanostructures for advanced chiroptical applications [17, 57], especially in the context of enhanced enantioselectivity, detection and characterization of chiral biomolecules via specifically designed chiral and nonchiral structures [58, 59].

The authors are grateful to C. García-Meca for valuable comments and discussions. This work was supported by fundings from Ministerio de Economía y Competitividad (MINECO) of Spain under Contract No. TEC2014-51902-C2-1-R.

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