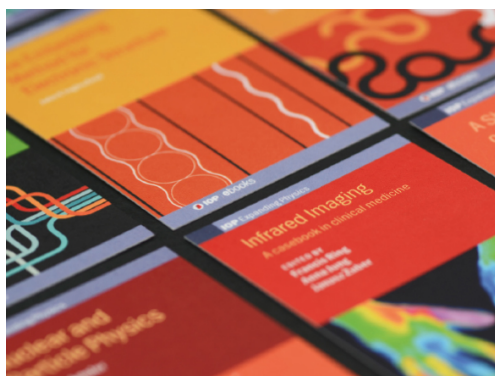


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On the differences between helicity and chirality

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Abstract

The optical helicity and the optical chirality are two quantities that are used to describe chiral electromagnetic fields. In a monochromatic field, the two quantities are proportional to one another, and the distinction between the two is therefore largely unimportant. However, in a polychromatic field, no such proportionality holds. This paper explicitly examines both the helicity and chirality densities in various polychromatic fields: the superposition of two circularly polarised plane-waves of different frequencies, a chirped pulse of circularly polarised light, and an ‘optical centrifuge’ consisting of two oppositely chirped circularly polarised beams of opposite handedness. Even in the simplest case, there can be significant qualitative differences between the two quantities—they may have opposite signs, or one may be zero while the other is not. The origin of these differences lies in the different frequency scaling of the two quantities, which is made relevant by the presence of multiple frequency components in the fields.

Keywords: optical helicity, optical chirality, optical centrifuge

(Some figures may appear in colour only in the online journal)

1. Introduction

If an electromagnetic field has a definite handedness, then it can interact differently with the different enantiomeric forms of chiral matter. In the study of chiral light–matter interactions, it is useful to have some quantitative measure of the chirality of an electromagnetic field, that in some way indicates the expected size of such differential interactions. It was proposed by Tang and Cohen [1] that a quantity, originally introduced by Lipkin under the name ‘zilch’ [2], could serve such a role, and this quantity is therefore often referred to as the ‘optical chirality’. It is a pseudoscalar that is locally conserved in the free field, and is defined by

$$\chi = \frac{\epsilon_0}{2} (\mathbf{E} \cdot \nabla \times \mathbf{E} + c^2 \mathbf{B} \cdot \nabla \times \mathbf{B}), \quad (1)$$

where \mathbf{E} and \mathbf{B} are the (real) electric and magnetic field vectors. Tang and Cohen motivated the use of this density by


showing that the differential excitation rate for a small chiral molecule in a monochromatic field is proportional to the chirality density of the field in which it is immersed.

Another closely related quantity is the electromagnetic helicity. Like the chirality, the helicity is locally conserved in free space [3]. It is similar in form, but is defined in terms of electromagnetic potentials, rather than the fields directly. The helicity density is defined by [3–6]

$$h = \frac{1}{2} \left(\sqrt{\frac{\epsilon_0}{\mu_0}} \mathbf{A} \cdot \nabla \times \mathbf{A} + \sqrt{\frac{\mu_0}{\epsilon_0}} \mathbf{C} \cdot \nabla \times \mathbf{C} \right), \quad (2)$$

where \mathbf{A} and \mathbf{C} are divergenceless vector potentials defined by $\nabla \times \mathbf{A} = \mathbf{B}$ and $\nabla \times \mathbf{C} = -\epsilon_0 \mathbf{E}$ [7, 8]. The volume integral of the helicity density is proportional to the difference in the number of left- and right-handed circularly polarised photons in the field [9], linking the concept to the particle physics notion of helicity as the component of spin angular momentum in the direction of propagation. The inclusion of both the \mathbf{A} and \mathbf{C} potentials in the definition is important in order to ensure that the helicity is locally conserved [3, 6].

The concepts of helicity and chirality have found use in the analysis of a wide range of electrodynamics problems.

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These include the enhancement of differential scattering rates for chiral molecules adsorbed on plasmonic nanostructures [10], the conversion of orbital to spin optical angular momentum in the far-field of a beam by scattering from a nanoparticle in the beam’s focus [11], and general studies of the propagation of light through dispersive or negative-index media [12], to name only three examples. In these, and in many other situations that the two measures are used to study, the fields concerned are monochromatic and the distinction between the two quantities is largely unimportant. However, while both quantities are proportional to one another in monochromatic fields, they are in general distinct—and the conservation of each actually arises from a different symmetry of the free electromagnetic field. The local conservation of helicity is related by Noether’s theorem to the duality symmetry of the free-space Maxwell equations [3, 4, 13, 14]—the invariance of the free space Maxwell equations under a mixing between electric and magnetic fields [15]. The local conservation of chirality, on the other hand, is linked to the infinitesimal symmetry transformation $\mathbf{A}' = \mathbf{A} + \theta(\nabla \times \frac{\partial \mathbf{A}}{\partial t})$, with θ an infinitesimal parameter [16].

This paper examines how even simple examples of polychromatic fields expose the differences between the chirality and the helicity. First, we examine the case of two co-propagating circularly polarised plane waves, of opposite handedness and different frequencies, and find that the helicity and chirality do not agree in sign. We then examine the positions of the maxima of helicity and chirality in a chirped circularly polarised pulse, showing that each is slightly displaced from the intensity maximum of the pulse in opposite directions. Finally, we consider the fields of an ‘optical centrifuge’, formed by two co-propagating circularly polarised pulses of opposite handedness, one positively and one negatively chirped [17]. Here, we find both features: that the measures disagree in sign, as for the unchirped plane waves, and that the helicity and chirality have different time dependencies, as for the chirped pulses. In all of the cases considered, the differences between the two measures of handedness can be understood by considering a frequency decomposition of the field, and the different frequency scaling of the measures.

2. Helicity and chirality in a superposition of two plane waves

Perhaps the simplest case in which the polychromatic nature of an electromagnetic field can cause differences between the helicity and the chirality is a superposition of two co-propagating circularly polarised plane waves, of opposite handedness and different frequencies. This situation is considered in [6], and below a more detailed discussion is given.

For definiteness, let us consider the complex electric and magnetic fields given by

$$\mathbf{E} = E_0 e^{-i\omega_1 \eta}(\mathbf{x} + i\mathbf{y}) + E_0 e^{-i\omega_2 \eta}(\mathbf{x} - i\mathbf{y}), \quad (3)$$

$$\mathbf{B} = \frac{E_0}{c} e^{-i\omega_1 \eta}(-i\mathbf{x} + \mathbf{y}) + \frac{E_0}{c} e^{-i\omega_2 \eta}(i\mathbf{x} + \mathbf{y}), \quad (4)$$

where E_0 is the peak electric field amplitude of each plane wave, \mathbf{x} and \mathbf{y} are unit vectors and $\eta \equiv t - z/c$. This electromagnetic field is a superposition of a right-handed plane wave with frequency ω_1 , and a left-handed wave with frequency ω_2 . If the two frequencies are fairly close to one another, then the result can be viewed as a linearly polarised wave of frequency $\frac{(\omega_1 + \omega_2)}{2}$, with the plane of polarisation slowly rotating at a frequency $\frac{(\omega_1 - \omega_2)}{2}$. In this sense, it is like the static limit of the optical centrifuge discussed in [17]—in our case, the rotation does not accelerate.

As we are considering the free field, we may choose a gauge in which the scalar potential is 0, and use the relationships $\mathbf{E} = -\frac{\partial \mathbf{A}}{\partial t}$ and $\mathbf{B} = -\frac{1}{\epsilon_0 c^2} \frac{\partial \mathbf{C}}{\partial t}$ to obtain the following choices for \mathbf{A} and \mathbf{C} ¹

$$\mathbf{A} = \frac{E_0}{\omega_1} e^{-i\omega_1 \eta}(i\mathbf{x} - \mathbf{y}) + \frac{E_0}{\omega_2} e^{-i\omega_2 \eta}(i\mathbf{x} + \mathbf{y}), \quad (5)$$

$$\mathbf{C} = -\sqrt{\frac{\epsilon_0}{\mu_0}} \frac{E_0}{\omega_1} e^{-i\omega_1 \eta}(\mathbf{x} + i\mathbf{y}) - \sqrt{\frac{\epsilon_0}{\mu_0}} \frac{E_0}{\omega_2} e^{-i\omega_2 \eta}(-\mathbf{x} + i\mathbf{y}). \quad (6)$$

The helicity density of the combined field can then be written

$$h = \epsilon_0 E_0^2 \left(\frac{1}{\omega_1} - \frac{1}{\omega_2} \right) (1 + \cos \eta(\omega_1 + \omega_2)). \quad (7)$$

The chirality density, by contrast, is given by

$$\chi = \frac{\epsilon_0 E_0^2}{c} (\omega_1 - \omega_2) (1 + \cos \eta(\omega_1 + \omega_2)). \quad (8)$$

We observe that the helicity and chirality densities have similar forms but opposite signs: if the frequency of the right-handed wave is higher than that of the left-, then the helicity is negative and the chirality positive, and vice versa when $\omega_2 > \omega_1$.

The reason for this can be seen intuitively by considering the relationship between the helicity density, chirality density and energy density in a single circularly polarised plane wave. The ratio of helicity density to energy density in a circularly polarised wave of frequency ω is given by [4]

$$\frac{h}{u} = \pm \frac{1}{\omega}, \quad (9)$$

where h is the helicity density, u is the energy density and the positive and negative signs refer to right- and left-handed waves, respectively. This relationship is clearly compatible with the notion that a circularly polarised photon carries a helicity of $\pm \hbar$. The ratio of chirality density to energy density is

$$\frac{\chi}{u} = \pm \frac{\omega}{c}, \quad (10)$$

which corresponds to a chirality of $\pm \frac{\hbar \omega^2}{c}$ per photon.

¹ We have also chosen a gauge in which \mathbf{A} and \mathbf{C} are everywhere divergence-free. A judicious choice of gauge is important when discussing a local helicity density, as the local helicity density is an explicitly gauge-dependent quantity. However, only the divergence-free parts of \mathbf{A} and \mathbf{C} contribute to the total helicity [4] (and the total helicity is thus gauge-independent), so it is natural to define the local helicity density using divergence-free potentials.

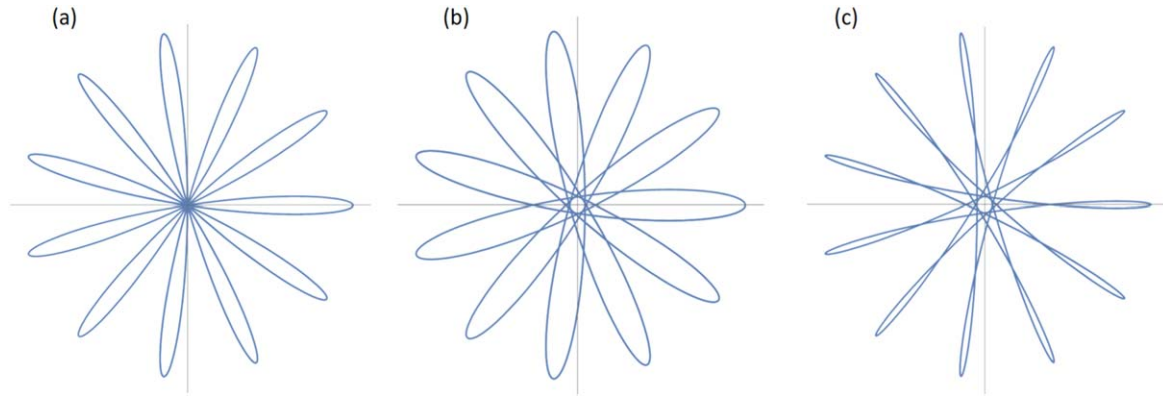


Figure 1. Illustrative plots showing the path traced by the electric field vector in the x - y plane at a point of fixed z , when two plane-waves are coherently superposed as described in the text. (a) The field given in (3), where both plane waves are of equal strength. (b) An example scaling where the average helicity is zero, and (c) where the average chirality is zero. For each of the figures, we use the somewhat impractical frequency relationship $\omega_1 = \frac{5}{6}\omega_2$, so that the rotation is fast enough that the polarisation can clearly be seen.

The origin of the difference in sign between the measures lies in this different frequency scaling—increasing the frequency, while keeping the energy density constant, decreases the helicity but increases the chirality density of a circularly polarised wave, and so the wave which makes the dominant contribution in the two-wave superposition is different for the two measures. Another way of putting this is that plane-waves of fixed energy density contain more photons if they are of lower frequency—and the helicity-per-photon is independent of frequency, while the chirality-per-photon is not.

The superposition that we have been considering consists of two waves with the same energy, and hence the same peak E-field amplitude. In a sense, it is this requirement that causes the different sized helicity contributions from the two waves—if we had instead formed a superposition of two plane waves with the same number of photons in each, then we would find the helicities from each the two individual waves equal and opposite, and the cycle-averaged helicity of the superposition would be zero.

In general, if the peak E -field amplitude is allowed to differ between the two waves, then in place of a linear polarisation with a rotating plane one obtains an elliptical polarisation with a rotating major axis. Changing the relative amplitudes will change both the helicity and chirality of the superposition, and it is possible to select amplitudes such that they have the same sign as one another, or such that the average helicity or chirality are zero. The expressions for the helicity and chirality densities when the two plane waves have amplitudes E_1 and E_2 are given by

$$h = \epsilon_0 \left(\frac{E_1^2}{\omega_1} - \frac{E_2^2}{\omega_2} + E_1 E_2 \left(\frac{1}{\omega_1} - \frac{1}{\omega_2} \right) \cos \eta (\omega_1 + \omega_2) \right), \tag{11}$$

$$\chi = \frac{\epsilon_0}{c} (E_1^2 \omega_1 - E_2^2 \omega_2 + E_1 E_2 (\omega_1 - \omega_2) \cos \eta (\omega_1 + \omega_2)). \tag{12}$$

We see that the average helicity is equal to zero if

$$\frac{E_1^2}{\omega_1} = \frac{E_2^2}{\omega_2}, \tag{13}$$

while the average chirality is equal to zero if

$$E_1^2 \omega_1 = E_2^2 \omega_2. \tag{14}$$

We note in passing that the conditions for changes in sign of helicity and chirality are not the same as those for which the handedness of the ‘elliptical polarisation’ changes (this is governed by which is larger of E_1 and E_2), nor do they correspond to changes of the sense of rotation of the major axis (which is governed by which is larger of ω_1 and ω_2). We note also that while certain amplitude scalings can make either the helicity or chirality separately zero, the only way to have both simultaneously zero is to have both $\omega_1 = \omega_2$ and $E_1 = E_2$, which corresponds to ordinary linear polarisation. Figure 1 shows the path traced by electric field vector in the x - y plane for some example field scalings.

3. The maxima of helicity and chirality in a chirped pulse

Another situation in which the different frequency scaling of helicity and chirality can cause a qualitative difference between the two measures is the case of a chirped circularly polarised pulse. The electric and magnetic fields of a pulse with a linear frequency chirp can be written [18]

$$\mathbf{E} = E_0 (\mathbf{x} + i\mathbf{y}) f(\eta) e^{-i(\omega_0 \eta + b \eta^2)}, \tag{15}$$

$$\mathbf{B} = \frac{E_0}{c} (-i\mathbf{x} + \mathbf{y}) f(\eta) e^{-i(\omega_0 \eta + b \eta^2)}, \tag{16}$$

where b is a parameter that determines the rate of the chirp, and $f(\eta)$ is a function determining the envelope of the pulse. Here, we will consider a Gaussian envelope of the form $f(\eta) = e^{-\frac{(\eta-a)^2}{2\sigma^2}}$, with a and σ the central position and standard deviation of the envelope. Note that the expressions for a linear frequency chirp given here are somewhat artificial, as a real linear chirp will extend over only a relatively narrow

spectral range, fixed by the bandwidth of the laser that produces the pulse. It must therefore be borne in mind that for these fields to be physically reasonable, a and σ will be such that the region of the pulse (in physical space) with appreciable field strengths is appropriately narrow, and far from the point at which the instantaneous frequency becomes negative.

The chirality density of the pulse is given by

$$\chi = \frac{\epsilon_0 E_0^2}{c} e^{-\frac{(\eta-a)^2}{\sigma^2}} (2b\eta + \omega_0). \quad (17)$$

Rather than being proportional to the frequency, as in the case of an unchirped plane wave, we see that it is proportional to the instantaneous frequency, $\omega_{inst}(\eta) = 2b\eta + \omega_0$. By differentiating with respect to η , we find that the maximum chirality density occurs at

$$\eta_{\chi_{max}} = \frac{2ba - \omega_0 + \sqrt{(2ba + \omega_0)^2 + 8b^2\sigma^2}}{4b}. \quad (18)$$

Note that the quadratic which results from differentiating the chirality density has two roots, but we choose the higher one, as the lower root corresponds to a local minimum in the unphysical region of negative instantaneous frequency. The maximum energy density occurs at the maximum of the envelope, a , and so the difference between the two is equal to

$$\eta_{\chi_{max}} - a = \frac{-(2ba + \omega_0) + \sqrt{(2ba + \omega_0)^2 + 8b^2\sigma^2}}{4b}. \quad (19)$$

We thus see that the maximum of chirality occurs after the maximum of energy. This makes intuitive sense, as the instantaneous frequency of the chirped pulse is higher at later times. This displacement is small: as typical examples of the frequency, pulse envelope and strength of the chirp, we can consider a pulse lasting around 50 ps, with an initial wavelength of 800 nm and a final wavelength of 780 nm (these values are chosen to be of comparable magnitude to those used in existing optical centrifuge experiments [19]). This would correspond to $\sigma \approx 25$ ps, $b \approx 6.0 \times 10^{23} \text{ s}^{-2}$, and $2ba + \omega_0 = 2.38 \times 10^{15} \text{ s}^{-1}$, making the time between maxima 0.16 ps².

In order to write the helicity density of the pulse, we can again obtain expressions for the vector potentials using the relationships $\mathbf{E} = -\frac{\partial \mathbf{A}}{\partial t}$ and $\mathbf{B} = -\frac{1}{\epsilon_0 c^2} \frac{\partial \mathbf{C}}{\partial t}$. However, the resulting indefinite integrals are quite cumbersome, and our analysis can equally well proceed by making the following approximations for the potentials:

$$\mathbf{A} \approx \frac{(-i\mathbf{x} + \mathbf{y})E_0 e^{-\frac{(\eta-a)^2}{2\sigma^2}} e^{-i(\omega_0\eta + b\eta^2)}}{\omega_0 + 2b\eta}, \quad (20)$$

² A difference of this size would not be seen in the actual pulse generation scheme used (for example) by [19]. They produce their chirped pulses effectively by splitting in Fourier space a beam with a Gaussian frequency distribution, and then delaying different frequency components. This means that the resulting pulses do not have a symmetrical Gaussian envelope, but are most intense initially, and decay as the chirp progresses.

$$\mathbf{C} \approx \sqrt{\frac{\epsilon_0}{\mu_0}} \frac{(-\mathbf{x} - i\mathbf{y})E_0 e^{-\frac{(\eta-a)^2}{2\sigma^2}} e^{-i(\omega_0\eta + b\eta^2)}}{\omega_0 + 2b\eta}. \quad (21)$$

These are simply the vector potentials of a circularly polarised plane-wave, but with the frequency in the denominator replaced by the instantaneous frequency, and with the inclusion of the appropriate frequency chirp and Gaussian envelope. A more rigorous treatment, finding the exact potentials by integrating (15) and (16), is sketched in the [appendix](#).

To check that (20) and (21) are reasonable approximations, we may examine the electric and magnetic fields derived from these potentials:

$$\mathbf{E} = -\frac{\partial \mathbf{A}}{\partial t} \approx E_0(\mathbf{x} + i\mathbf{y})e^{-i(\omega_0 + b\eta^2)} \times \left[-\frac{i(\eta - a)}{\sigma^2(\omega_0 + 2b\eta)} - \frac{2bi}{(\omega_0 + 2b\eta)^2} + 1 \right], \quad (22)$$

$$\mathbf{B} = -\frac{1}{\epsilon_0 c^2} \frac{\partial \mathbf{C}}{\partial t} \approx \frac{E_0}{c}(-i\mathbf{x} + \mathbf{y})e^{-i(\omega_0 + b\eta^2)} \times \left[-\frac{i(\eta - a)}{\sigma^2(\omega_0 + 2b\eta)} - \frac{2bi}{(\omega_0 + 2b\eta)^2} + 1 \right]. \quad (23)$$

The first two terms in the square brackets are always much less than 1, because of the optical frequencies in the denominators, showing that these potentials are indeed good approximations to the exact vector potentials.

The helicity calculated from these approximate vector potentials is given by

$$h = \epsilon_0 E_0^2 e^{-\frac{(\eta-a)^2}{\sigma^2}} \frac{1}{2b\eta + \omega_0}, \quad (24)$$

so the maximum helicity occurs at

$$\eta_{h_{max}} = \frac{2ba - \omega_0 + \sqrt{(2ba + \omega_0)^2 - 8b^2\sigma^2}}{4b}, \quad (25)$$

where again we have taken the higher root, as the lower one corresponds to a region of negative instantaneous frequency. The difference between the maxima of helicity and energy density is then given by

$$\eta_{h_{max}} - a = \frac{-(2ba + \omega_0) + \sqrt{(2ba + \omega_0)^2 - 8b^2\sigma^2}}{4b}, \quad (26)$$

confirming that the maximum of helicity occurs slightly before the maximum of energy. These results are illustrated in figure 2, which shows the calculated energy, helicity and chirality densities for a positive-helicity circularly polarised plane wave pulse. The parameters used in the figure are a standard deviation of $\sigma = 0.2$ ns, a central frequency of $\omega_0 + 2b a = 2.15 \times 10^{15} \text{ rad s}^{-1}$, and a chirp strength of $b = 5.6 \times 10^{24} \text{ s}^{-2}$, which have been chosen in order to make the differences between the densities clearly visible.

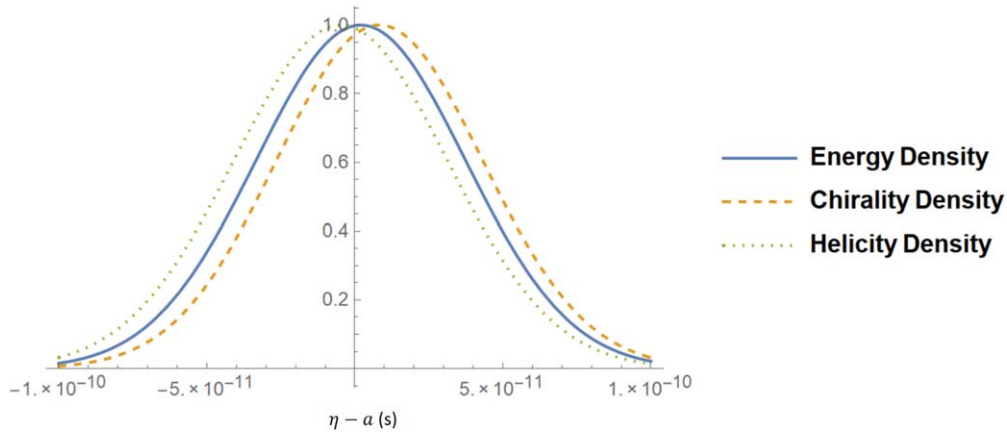


Figure 2. Calculated energy, chirality and helicity densities for a positive-helicity circularly polarised plane wave pulse, normalised so that each has a maximum of 1. The parameters used are a standard deviation of $\sigma = 0.2$ ns, a central frequency of $\omega_0 + 2ba = 2.15 \times 10^{15}$ rad s⁻¹, and a chirp strength of $b = 5.6 \times 10^{24}$ s⁻². These parameters describe a pulse significantly longer and broader in frequency than the more realistic ones discussed in the text, but are chosen to illustrate the behaviour.

4. Helicity and chirality in an optical centrifuge

An optical centrifuge is a superposition of a left- and a right-handed circularly polarised plane wave, as in section 1, but with each wave linearly chirped—one chirped up and the other down. This means that the frequency difference between the two waves increases linearly with time—and so if the resulting superposition is viewed as linear polarisation with a rotating plane of polarisation, the angular speed of this rotation increases at a constant rate. These fields can be used to excite molecules to very high rotational states, and even dissociate heavy molecules. The action of an optical centrifuge in inducing molecular rotation can be analysed classically, as in the original paper of Karczmarek *et al* [17], or viewed quantum-mechanically as driving the molecules up a successive ladder of rotational transitions [20, 21].

Because of this practical use, it seems worthwhile to extend the reasoning of sections 1 and 2 to examine the net helicity and chirality in such a field configuration. The fields of an optical centrifuge are given by the real parts of

$$\mathbf{E} = E_0 f(\eta) [(\mathbf{x} + i\mathbf{y})e^{-i(\omega_0\eta + b\eta^2)} + (\mathbf{x} - i\mathbf{y})e^{-i(\omega_0\eta - b\eta^2)}], \quad (27)$$

$$\mathbf{B} = \frac{E_0}{c} f(\eta) [(-i\mathbf{x} + \mathbf{y})e^{-i(\omega_0\eta + b\eta^2)} + (i\mathbf{x} + \mathbf{y})e^{-i(\omega_0\eta - b\eta^2)}]. \quad (28)$$

From these, it is straightforward to find the chirality density

$$\chi = \frac{8\epsilon_0 E_0^2}{c^2} e^{-\frac{(\eta-a)^2}{\sigma^2}} b\eta \cos^2(\omega_0\eta). \quad (29)$$

To find the helicity density, it is most straightforward to use the following approximations to the potentials, similar to the approximations made in section 2

$$\mathbf{A} \approx E_0 e^{-\frac{(\eta-a)^2}{2\sigma^2}} \left[\frac{(-i\mathbf{x} + \mathbf{y})e^{-i(\omega_0\eta + b\eta^2)}}{\omega_0 + 2b\eta} + \frac{(-i\mathbf{x} - \mathbf{y})e^{-i(\omega_0\eta - b\eta^2)}}{\omega_0 - 2b\eta} \right], \quad (30)$$

$$\mathbf{C} \approx \sqrt{\frac{\epsilon_0}{\mu_0}} E_0 e^{-\frac{(\eta-a)^2}{2\sigma^2}} \left[\frac{(-\mathbf{x} - i\mathbf{y})e^{-i(\omega_0\eta + b\eta^2)}}{\omega_0 + 2b\eta} + \frac{(\mathbf{x} - i\mathbf{y})e^{-i(\omega_0\eta - b\eta^2)}}{\omega_0 - 2b\eta} \right]. \quad (31)$$

We then find, using $h = \frac{1}{2} \left(\sqrt{\frac{\epsilon_0}{\mu_0}} \text{Re}[\mathbf{A}] \cdot \text{Re}[\mathbf{B}] - \sqrt{\epsilon_0\mu_0} \text{Re}[\mathbf{C}] \cdot \text{Re}[\mathbf{E}] \right)^3$

$$h = \frac{-8\epsilon_0 c E_0^2}{(\omega_0 + 2b\eta)(\omega_0 - 2b\eta)} e^{-\frac{(\eta-a)^2}{\sigma^2}} b\eta \cos^2(\omega_0\eta). \quad (33)$$

The energy density in the centrifuge is given by

$$u = 4E_0^2 \epsilon_0 e^{-\frac{(\eta-a)^2}{\sigma^2}} \cos^2(\omega_0\eta), \quad (34)$$

so, as in section 2, we have the result that the helicity and the chirality have opposite signs, with the chirality dominated by the higher frequency wave and the helicity by the lower frequency one. Due to the time-varying frequencies, the helicity and chirality densities are no longer proportional to the energy density—the ratios are

$$\frac{h}{u} = \frac{-2cb\eta}{(\omega_0 + 2b\eta)(\omega_0 - 2b\eta)}, \quad (35)$$

³ It would also be possible to use $h = \frac{1}{2} \left(\sqrt{\frac{\epsilon_0}{\mu_0}} \text{Re}[\mathbf{A}] \cdot \text{Re}[\nabla \times \mathbf{A}] + \sqrt{\frac{\mu_0}{\epsilon_0}} \text{Re}[\mathbf{C}] \cdot \text{Re}[\nabla \times \mathbf{C}] \right)$. One might argue that this is more self-consistent, as here we consistently use the approximate potentials, rather than mixing between exact fields and approximate potentials. If we do this, we obtain

$$h = \frac{-8\epsilon_0 c E_0^2 e^{-\frac{(\eta-a)^2}{\sigma^2}}}{(\omega_0 + 2b\eta)(\omega_0 - 2b\eta)} b\eta \cos^2(\omega_0\eta) + \frac{8\epsilon_0 c E_0^2 e^{-\frac{(\eta-a)^2}{\sigma^2}}}{2(\omega_0 + 2b\eta)^2(\omega_0 - 2b\eta)^2} b\omega_0 \sin(2\omega_0\eta). \quad (32)$$

The first term is the same as (33), and the second is much smaller, as it contains the squares of the instantaneous frequencies in the denominator. This question does not arise for the single chirped pulse considered in section 3: both procedures lead identically to (24).

$$\frac{\chi}{u} = \frac{2b\eta}{c^2}. \tag{36}$$

Taking the ratio with the energy density in this way removes the effect of the pulse envelope and the rapid oscillations at ω_0 . This makes clear the main qualitative difference between the measures—they have opposite signs, and different dependences on the instantaneous frequencies of the two beams.

5. Conclusion

This paper has examined the helicity and chirality densities in various polychromatic fields, and shown how the polychromaticity leads to significant differences between the two measures. It is striking that the quantities, which both in a sense indicate the ‘handedness’ of the field, may differ in sign in even such simple cases. The easiest way to understand this is that the frequency dependence of the two quantities can be a decisive factor when multiple frequency components are present, while in the monochromatic case it leads only to each quantity being multiplied by a different constant.

This behaviour seems counter-intuitive if the helicity or chirality densities are thought of, in some sense, as measures of the ‘amount’ of handedness in an electromagnetic field. However, there can be no single property that captures this idea of handedness, as it is impossible in general to define a quantitative measure of chirality which applies to all geometrical systems. For any particular pseudoscalar measure that might be supposed to indicate the ‘amount’ of chirality associated with a configuration of points, there will exist chiral configurations for which the measure is zero [22, 23]. This can be seen by considering that it is always possible to smoothly deform a configuration into its enantiomer without passing through an achiral configuration on the way. If the measure is a continuous function of the positions of the points, it must pass through a zero during this deformation, and therefore assign a zero to a chiral configuration.

It is therefore clear that, in the absence of globally satisfactory criteria, any quantification of chirality—of the electromagnetic field, or otherwise—must be performed with a view towards context and applications. In connection with this, we may note that the helicity is more transparently connected to physical quantities of the field, such as the spin angular momentum and photon number, than the chirality [4]. Ultimately, the appropriateness of either of the measures studied here as a description of a polychromatic light field will depend on the extent to which they are found useful in describing the interaction of the field with matter.

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Appendix. Exact expressions for the vector potentials in a chirped pulse and an optical centrifuge

When considering chirped circularly polarised pulses in the body of the article, we made use of approximations to the vector potentials, and avoided explicitly evaluating the indefinite integrals $\mathbf{A} = -\int \mathbf{E} dt$ and $\mathbf{C} = -c^2 \int \mathbf{B} dt$. Here we show an analytical method of treating these integrals. The integrals of (15) and (16) can be found using the standard integral [24]

$$\int e^{-(\alpha x^2 + 2\beta x + \gamma)} dx = \frac{1}{2} \sqrt{\frac{\pi}{\alpha}} e^{\frac{\beta^2 - \alpha\gamma}{\alpha}} \operatorname{erf}\left(\sqrt{\alpha}x + \frac{\beta}{\sqrt{\alpha}}\right), \tag{37}$$

where the error function is defined by $\operatorname{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-t^2} dt$, and is extended to complex arguments by analytic continuation. With the identifications $\alpha = \frac{1}{2\sigma^2} + ib$, $\beta = \frac{1}{2}\left(-\frac{a}{\sigma^2} + i\omega_0\right)$ and $\gamma = \frac{a^2}{2\sigma^2}$, we find the \mathbf{A} potential associated with the chirped circularly polarised pulse of (15) and (16) to be

$$\mathbf{A} = -E_0(\mathbf{x} + i\mathbf{y}) \int e^{-\frac{(\eta-a)^2}{2\sigma^2}} e^{-i(\omega_0\eta + b\eta^2)} d\eta \tag{38}$$

$$= -\frac{1}{2}E_0(\mathbf{x} + i\mathbf{y}) \sqrt{\frac{\pi}{\alpha}} e^{\frac{\beta^2 - \alpha\gamma}{\alpha}} \operatorname{erf}\left(\sqrt{\alpha}\eta + \frac{\beta}{\sqrt{\alpha}}\right). \tag{39}$$

Corresponding results can be found for the \mathbf{C} potential, and for down-chirped pulses or pulses of the opposite handedness. From these expressions for the potentials associated with chirped pulses, potentials corresponding to the optical centrifuge can also be straightforwardly obtained. Numerical values of the helicity density can be found by taking the real part of the complex potentials, using (for example) a series expansion for the complex error functions as given in [24].

There is a slight complication: the helicity (unlike the chirality) is non-local in the fields, in the sense that the helicity density at a point depends not only on the \mathbf{E} and \mathbf{B} fields and their derivatives at that point, but on the fields at other points as well [3]. This means that behaviour associated with the unphysical aspects of the infinite linear chirp is not necessarily avoided by considering the helicity density only in regions of physical interest. If the helicity density of the optical centrifuge is formed directly from the real part of (38) and corresponding results for the other waves and potentials, one finds rapid oscillations superimposed on the expected helicity density, which decrease in prominence when the envelope is reduced or the centres of the pulses concerned are moved further from the regions of negative instantaneous frequency.

Related to this is the fact that the \mathbf{A} and \mathbf{C} potentials are only defined up to a constant. While one is at liberty to choose any constant without affecting the physical fields, or the total helicity density, the choice can substantially affect the local density. This can be clearly seen if one considers calculating the helicity density of a circularly polarised plane wave, but adding a constant to the usually chosen \mathbf{A} potential: one would find rapid oscillation in the $\mathbf{A} \cdot \nabla \times \mathbf{A}$ term, caused by

the product of the oscillatory $\nabla \times \mathbf{A}$ and the added constant in \mathbf{A} .

For localised fields, this ambiguity is usually avoided by choosing the constant such that the potentials vanish at large distances (where the fields approach 0). However, the expressions for the potentials given above do not approach 0 as η approaches ∞ or $-\infty$, as $\lim_{x \rightarrow \pm\infty} \operatorname{erf}(x) = \pm 1$ when the argument of x is strictly less than $\frac{\pi}{4}$ (as is always the case in (39) with large η). Furthermore, no constant that can be added that will make these potentials approach 0 at both positive and negative ∞ . This feature is connected with the unphysical nature of an infinite linear frequency chirp.

The appropriate choice of constants to remove the rapid oscillation are those that make the potentials of the up-chirped pulse vanish at large positive times, and the potentials of the down-chirped pulse vanish at large negative times. It must be remembered that these are the large η limits in which the expressions for the respective chirps are still physically sensible, so it seems reasonable to be concerned with the behaviour of the potentials in these limits rather than the complementary ones⁴.

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⁴ The oscillatory helicity densities are still of course 'physically sensible' in the sense that they are locally conserved, and give the correct total helicity when integrated over all space. Adding a constant to \mathbf{A} is nothing other than an example of the simplest gauge transformation, and does not affect the physics of the system. However, there is always ambiguity when electromagnetic quantities are represented by local densities. If we are concerned with the choice that has the clearest physical meaning, choosing a constant to remove the rapid oscillation seems appropriate.