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Light intensity correlations in optically active media

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Abstract
We calculate the field–field correlation function in a disordered optically active medium. It is shown that this function exhibits characteristic spatial oscillations, with a period related to the chirality parameter of the medium. Similar oscillations show up in correlations of the polarization-resolved intensity. In contrast, correlations in the total intensity, i.e. summed over all polarizations, are not sensitive to chirality.

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In a weakly disordered dielectric medium, electromagnetic radiation propagates by diffusion, being multiply scattered by random inhomogeneities of the dielectric constant. As a result, a complex, apparently random, field and intensity pattern (a speckle pattern) is established in the sample. It is described in statistical terms, with the help of the correlation functions $\langle E_i(\mathbf{r}_1)E_j^*(\mathbf{r}_2)\rangle$ and $\langle \Delta I(\mathbf{r}_1)\Delta I(\mathbf{r}_2)\rangle$, where E_i is the ith component of the electric field, ΔI is the deviation of the intensity from its average value and the angular brackets denote averaging over the ensemble of disordered samples.

It was noticed long ago that the correlation functions exhibit oscillatory behaviour if one assumes that the field is a superposition of many contributions with random phases [1–3]. In a systematic approach, based on the diagram technique, one does not make assumptions on the statistical properties of the field but only on the properties of the medium. This approach reveals an exponential decay of the correlations, on a scale of the mean free path l, in addition to the rapid oscillations (on a scale of the wavelength) [4]. Lately there was a renewed interest in the subject of spatial correlations [5, 6], motivated by the recent experiments [7–9]. In particular, in [5] correlations in a weakly disordered photonic crystal were studied and it was shown that they contain useful information about the corresponding clean crystal. It is quite remarkable that the disorder induced effect of correlations reveals the properties of the underlying clean medium. The point is that the averaging process, involved in calculating the correlations, deletes the 'irrelevant', sample-specific information of the apparently random speckle pattern, so that the structure of the underlying clean medium emerges. In this paper, we obtain an expression for the field correlation function in an optically active (chiral) medium and show that the chirality parameter of the underlying clean medium can be extracted from that expression.

The electric field $\mathbf{E}(\mathbf{r})$ of a monochromatic wave propagating in a chiral disordered medium, with no sources inside the medium, satisfies the equation

$$\nabla \times \nabla \times \mathbf{E} - k_0^2 \left(1 + \frac{\delta\varepsilon(\mathbf{r})}{\varepsilon_0}\right) \mathbf{E} - k_0 \gamma \nabla \times \mathbf{E} = 0 \tag{1}$$

where k_0 is the wave number in the medium, ε_0 is the average dielectric constant, $\delta\varepsilon$ is its fluctuating part and $\gamma \ll 1$ is the chirality parameter. It is defined as the relative difference in the velocities, $(\Delta c)/c$, of two waves propagating (in the absence of disorder) with the same frequency and in the same direction, but with opposite circular polarizations—one clockwise, the other counterclockwise. For $\delta\varepsilon = 0$, equation (1) describes an isotropic chiral medium, which is characterized by a scalar dielectric constant and the chirality parameter. The random scatterers, which are responsible for the fluctuations in the dielectric constant, are assumed to be non-chiral. The sensitivity of the correlation function $\langle E_i(\mathbf{r}_1) E_j^*(\mathbf{r}_2)\rangle \equiv C_{ij}(\mathbf{r}_1 - \mathbf{r}_2)$ to the value of the chirality parameter γ can be explained as follows: in a clean system the two circular polarizations define two independent branches of the spectrum. Disorder couples the branches, introducing 'dephasing' between the two (circularly polarized) components of a linearly polarized wave. The corresponding 'dephasing length' is the mean free path l, i.e. the concept of branches retains its validity locally, at a scale of order l or smaller. In other words, the two components of the linearly polarized wave will move on their respective branches (and preserve their phases) at a distance of order l, after which these phases will get scrambled by interbranch scattering. Therefore, if at a distance $|\mathbf{r}_1 - \mathbf{r}_2| \equiv \mathbf{R}$ the polarization direction undergoes a significant change, then one can expect to observe it in the correlation function $C_{ij}(\mathbf{R})$. To make the argument more explicit, let us introduce the rotation distance, $L_\gamma = (4\pi/\gamma k_0)$, over which the polarization direction makes a full rotation. Then, for $L_\gamma \ll l$ the polarization direction will manage to make l/L_γ full rotations, so that $C_{ij}(\mathbf{R})$ will exhibit l/L_γ oscillations, with a period L_γ, as a function of $\mathbf{R}$. These oscillations are superimposed on the much more rapid oscillations, with the period of one wavelength, which also exist in the absence of chirality. In the opposite case, $L_\gamma \gg l$, there is rotation only by a small angle (over distance l) and no γ-related oscillations can occur.

We now turn to an analytical calculation of $C_{ij}(\mathbf{R})$. Let us first consider the Green's function of the clean system, i.e. $\delta\varepsilon = 0$. It is a 3×3 matrix in the polarization space and, in the momentum representation, it satisfies the equation

$$\left[\left(k_0^2 + \mathrm{i}\eta - p^2\right)\delta_{in} + p_i p_n + \mathrm{i}k_0 \gamma e_{imn} p_m\right] G_{nj}^0(\mathbf{p}) = \delta_{ij}, \tag{2}$$

where e_{imn} is the fully antisymmetric Levi-Civita tensor. The tensor $\mathrm{G}_{ij}^0(\mathbf{p})$ has a transverse and a longitudinal part with respect to the vector $\mathbf{p}$. The two parts describe, respectively, propagating and non-propagating (electrostatic) modes [10]. The latter correspond to the dipole field near the source. In the studies of propagation of electromagnetic waves in disordered media, one usually concentrates on the propagating modes [11–13]. Furthermore, the difference between $\mathrm{G}_{ij}^0(\mathbf{p})$ and the average Green's function, $\langle \mathrm{G}_{ij}(\mathbf{p})\rangle$, in a disordered medium amounts to the standard replacement of the infinitesimal η by k_0/l. This replacement is justified for the case of weak disorder, which is assumed throughout this paper. Furthermore, we assume white-noise Gaussian disorder, so that there is no difference between the single-particle mean free path l and the transport mean free path. This leads to the following expression for the transverse Green's function [11–13]:

$$\langle G_{ij}(\mathbf{p})\rangle = \frac{1}{2} \frac{\delta_{ij} - \hat{p}_i \hat{p}_j - \mathrm{i}e_{ijl}\hat{p}_l}{k_0^2 - p^2 + \gamma k_0 p + \mathrm{i}k_0/l} + \frac{1}{2} \frac{\delta_{ij} - \hat{p}_i \hat{p}_j + \mathrm{i}e_{ijl}\hat{p}_l}{k_0^2 - p^2 - \gamma k_0 p + \mathrm{i}k_0/l} \tag{3}$$

where the circumflex denotes a unit vector. The numerators in equation (3) are projection operators that filter out the transverse waves with circular polarization. The denominators are just the scalar propagators corresponding to the two circular polarizations. In what follows we will need the average Green's function in **r**-space:

$$\langle G_{ij}(\mathbf{R})\rangle = G_0(\mathbf{R})\left[(\delta_{ij}-\hat{R}_i\hat{R}_j)\cos\left(\frac{1}{2}\gamma k_0 R\right)+e_{ijl}\hat{R}_l\sin\left(\frac{1}{2}\gamma k_0 R\right)\right] \quad (4)$$

where $\mathbf{R}$ is the vector connecting the source $\mathbf{r}_2$ to the observation point $\mathbf{r}_1$ and

$$G_0(\mathbf{R}) = -\frac{1}{4\pi R}\exp\left(\mathrm{i}k_0 R - \frac{R}{2l}\right) \quad (5)$$

is the average propagator for the scalar field. Due to the chiral nature of the medium, $\langle G_{ij}(\mathbf{R})\rangle$ is not symmetric, either with respect to the change of the direction of $\mathbf{R}$ or with respect to the interchange between i and j. Note however that

$$\langle G_{ij}(\mathbf{R})\rangle = \langle G_{ji}(-\mathbf{R})\rangle, \quad (6)$$

as required by the time reversal symmetry.

The calculation of $C_{ij}(\mathbf{R})$ starts with the Bethe–Salpeter equation which has been widely used for light propagation in disordered media, including the effects of chirality [14]. Let us emphasize that in our treatment, the random scatterers are not chiral, only the underlying clean medium is. The Bethe–Salpeter equation in real space is

$$\langle E_i(\mathbf{r}_1)E_j^*(\mathbf{r}_2)\rangle = \langle E_i(\mathbf{r}_1)\rangle\langle E_j^*(\mathbf{r}_2)\rangle + \frac{6\pi}{l}\int \mathrm{d}\mathbf{r}\sum_{m,n}\langle G_{im}(\mathbf{r}_1,\mathbf{r})\rangle\langle G_{jn}^*(\mathbf{r}_2,\mathbf{r})\rangle\langle E_m(\mathbf{r})E_n^*(\mathbf{r})\rangle. \quad (7)$$

Setting $\mathbf{r}_1 = \mathbf{r}_2$, one obtains a close equation for $\langle E_i(\mathbf{r})E_j^*(\mathbf{r})\rangle$, whose solution is

$$\langle E_i(\mathbf{r})E_j^*(\mathbf{r})\rangle = \tfrac{1}{3}\langle I(\mathbf{r})\rangle\delta_{ij} \quad (8)$$

where $\langle I(\mathbf{r})\rangle = \langle|E(\mathbf{r})|^2\rangle$ is the total (i.e. summed over all polarization) intensity at point $\mathbf{r}$. ($\langle E_i(\mathbf{r})\rangle = 0$, due to rapid randomization of the phase of the field within the medium.) Since $\langle I(\mathbf{r})\rangle$ does not change on a scale of the mean free path, we obtain upon substitution of (8) into (7)

$$\begin{aligned}\langle E_i(\mathbf{r}_1)E_j^*(\mathbf{r}_2)\rangle &= \frac{6\pi}{l}\frac{1}{3}\langle I(\mathbf{r}_1)\rangle\int \mathrm{d}\mathbf{r}\sum_n\langle G_{in}(\mathbf{r}_1,\mathbf{r})\rangle\langle G_{jn}^*(\mathbf{r}_2,\mathbf{r})\rangle \\ &= \mathrm{i}\frac{\pi}{k_0}\langle I(\mathbf{r}_1)\rangle[\langle G_{ij}(\mathbf{r}_1,\mathbf{r}_2)\rangle - \langle G_{ji}^*(\mathbf{r}_2,\mathbf{r}_1)\rangle]. \end{aligned} \quad (9)$$

Thus, the field correlation in a speckle pattern, produced by the radiation propagating in a random medium, is related to the average Green's function in the medium and the local average intensity. Equation (9) implies that the field correlation between a pair of points is primarily due to waves that are scattered near one point and arrive at the vicinity of the other point. Combining equations (4) and (9) we arrive at the final expression for the field correlation function:

$$\begin{aligned}\langle E_i(\mathbf{r}_1)E_j^*(\mathbf{r}_2)\rangle = -\frac{2\pi}{k_0}\langle I(\mathbf{r}_1)\rangle\bigg[(\delta_{ij}-\hat{R}_i\hat{R}_j)\cos\left(\frac{1}{2}\gamma k_0 R\right) \\ + e_{ijl}\hat{R}_l\sin\left(\frac{1}{2}\gamma k_0 R\right)\bigg]\mathrm{Im}G_0(\mathbf{R}). \end{aligned} \quad (10)$$

Here the expression in square brackets describes the oscillations due to chirality, whereas the factor $\mathrm{Im}G_0(\mathbf{R})$ is responsible for the rapid oscillations and the exponential decay $\exp(-R/2l)$

well known for a non-chiral isotropic disordered medium. In order to observe the chirality-related oscillations, one needs the period of these oscillations to be much smaller than the mean free path, which amounts to the condition $L_\gamma \ll l$, as discussed above. A good candidate for observing these oscillations is the blue phase of liquid crystals. The chirality in this phase is three-dimensional (unlike the uniaxial chiral nematics) and is very strong; the length L_γ is only a few thousand angstroms, which is comparable to the optical wave length. Therefore, to have a clear separation between the two types of oscillations, one has to go to the ultraviolet region. The mean free path is a controllable parameter, depending on the degree of disorder, and the condition $L_\gamma \ll l$ is easily fulfilled.

In conclusion, the speckle pattern is highly irregular and sample specific. Averaging, involved in the calculation of the field correlation function, helps us 'decipher the speckle' and reveal the chiral nature of the underlying clean medium. The chirality of the medium manifests itself in spatial oscillations with a period $L_\gamma = (4\pi/\gamma k_0)$.

We concentrated on the field correlation function $\langle E_i(\mathbf{r}_1)E_j^*(\mathbf{r}_2)\rangle$. These correlations manifest themselves in a short-range term of the polarization-resolved intensity correlation, $\langle \Delta I_i(\mathbf{r}_1)\Delta I_j(\mathbf{r}_2)\rangle$, which is known as the C_1-term (see e.g. [7]) and is equal to $|\langle E_i(\mathbf{r}_1)E_j^*(\mathbf{r}_2)\rangle|^2$. Note, however, that the correlation function $\langle \Delta I(\mathbf{r}_1)\Delta I(\mathbf{r}_2)\rangle$, where $I(\mathbf{r}) = |E(\mathbf{r})|^2$ is the total intensity (i.e. summed over all polarizations), is not sensitive to chirality. Indeed, $I(\mathbf{r})$ is a true scalar (rather than a pseudoscalar) and, as such, should be invariant under space inversion. In contrast, the Poynting vector, $\boldsymbol{J}(\mathbf{r})$, is sensitive to chirality, so that the correlation function $\langle \boldsymbol{J}_i(\mathbf{r}_1)\boldsymbol{J}_j(\mathbf{r}_2)\rangle$ does exhibit chirality-related oscillations. Finally, let us note that no such oscillations are expected in the long-range intensity correlations, which are due to diffusion and cannot have any structure on a scale smaller than the mean free path.

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