## Incomplete Photonic Band Gap as Inferred from the Speckle Pattern of Scattered Light Waves

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Motivated by recent experiments on intensity correlations of the waves transmitted through disordered media, we demonstrate that the speckle pattern from disordered photonic crystal with incomplete band gap represents a sensitive tool for determination of the stop-band width. We establish the quantitative relation between this width and the *angular anisotropy* of the intensity correlation function.

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Introduction.—A wave propagating in a random medium undergoes multiple scattering and forms a complicated intensity pattern, commonly referred to as a speckle pattern. It is described in statistical terms, with the help of probability distributions and correlation functions. The intensity correlation function,  $\langle \delta I(\mathbf{r}) \delta I(\mathbf{r}') \rangle$ , where  $\delta I(\mathbf{r})$  is the deviation from the averaged intensity at point r, contains a short-range term [1],  $C_1(r, r')$ , which oscillates on a scale of the wavelength and exponentially decays beyond the mean free path, l. It also contains small long-range terms [2-4], which become dominant for  $|\mathbf{r} - \mathbf{r}'| \gg l$ . Although the theory of speckles was developed some time ago, it is only recently that the first experimental measurements of the spatial correlator  $C_1(\mathbf{r}, \mathbf{r}')$  were reported both for microwaves [5] and for optical waves [6,7]. These experiments were carried out for isotropic media; macroscopic isotropy was also assumed in the existing theories. In this isotropic situation, comparison with the theory has enabled the authors [6] to infer the effective refractive index, which is the only relevant parameter of the medium in the absence of disorder.

The main message of this Letter is that, in a medium with *underlying spatial structure*, the pattern of intensity correlations exhibits a vastly richer behavior as compared to the isotropic case. Moreover, additional features in  $C_1(\mathbf{r}, \mathbf{r}')$  carry *quantitative* information about this structure. As an example, we consider a disordered incomplete-band gap photonic crystal and demonstrate how the band-structure parameters can be extracted from the *angular anisotropy* of the correlator  $C_1$ .

Superficially, it may seem that a wave with frequency  $\omega$ , arriving from a distant source and after having been scattered by many random inhomogeneities, will lose all information about the crystal band structure. Our point, though, is that  $C_1(\mathbf{r}, \mathbf{r}')$  is essentially a *local* object: It is determined by the last scattering event, prior to the arrival to the points  $\mathbf{r}, \mathbf{r}'$ , and thus it is not sensitive to the "prehistory." In other words,  $C_1(\mathbf{r}, \mathbf{r}')$  can serve as a "microscope" for observation of the local interference picture (on a scale smaller than *l*), thus revealing the band

structure of the inherent photonic crystal. Such a microscope is particularly well suited for determination of the band structure of *realistic* photonic crystals since it is not affected by the long-range disorder. On the other hand, long-range disorder is a generic feature of realistic crystals, such as silica-based synthetic opals.

Disorder in synthetic opals.—Silica-based synthetic opals, which are the fcc self-assembled arrangement of almost monodispersed silica spheres, play a distinguished role in the fabrication of photonic band gap materials, the potential of which was first appreciated in Refs. [8,9]. This is because the opals constitute a template subsequently infiltrated with high refraction index material, while the spheres are selectively removed (see, e.g., [10–12]). The majority of studies on light propagation in photonic crystals has been carried out on opals [13].

In opals, the contrast of the dielectric constant is weak so that the photonic band gaps are not only incomplete, but also so that the corresponding stop bands are relatively narrow. For this reason, the photonic band structure of these materials is strongly obscured by the disorder. As a result, any reliable determination of the stop band, either from disorder-broadened reflection maxima or from transmission minima, which are strongly affected by the disorder, is highly ambiguous. In fact, in early studies, actual extraction of the stop-band width from the data was based on the "thumb rules" [14,15].

Later studies [13,16–18] have yielded a deeper insight into the microscopic origin of the disorder in opals. Namely, they indicated that one should distinguish three types of the disorder: (i) Short-range disorder due to point defects and spread in the sphere diameters. This type of disorder causes the mean free path [19]  $l \sim 15 \mu m$ . (ii) Stacking faults in [111] direction [16]. This disorder has a *one-dimensional* character and thus might give rise to the 1D ingap states [20] within the incomplete stop band. In fact, experimentally observed deep and sharp transmission minimum [16] near the stop-band center has been accounted for in these states. Subsequent interpretation of reflection and transmission [17] as well as diffraction data [18] also relied on the prominent role played by the stacking faults. (iii) Macroscopic domains that are  $\sim$ 50–100  $\mu$ m in size. Such domains unavoidably emerge in the course of self-assembly of thick enough photonic crystals [13]. Because of their presence, the stop bands are strongly *inhomogeneously* broadened—a serious complication for determination of photonic band structure.

Summarizing, the studies [13,16-18] have led to significant improvement of the understanding of the light propagation in strongly disordered incomplete-band gap photonic crystals. However, inferring the stop band width from reflection and transmission remains a problem. Below we demonstrate that this width naturally emerges in the *angular dependence* of the intensity correlator  $C_1$ .

Correlator  $C_1$  in a disordered photonic crystal.—The correlator  $C_1(\mathbf{r}, \mathbf{r}')$  is obtained by factorizing the product of four fields,  $\psi(\mathbf{r})$  in the average  $\langle I(\mathbf{r})I(\mathbf{r}')\rangle$ . This leads [2] to the relation  $C_1(\mathbf{r}, \mathbf{r}') = |C_{\psi}(\mathbf{r}, \mathbf{r}')|^2$ , where

$$C_{\psi}(\mathbf{r}, \mathbf{r}') = \frac{\langle \psi(\mathbf{r})\psi^{\star}(\mathbf{r}')\rangle}{\langle I(\mathbf{r})\rangle^{1/2}\langle I(\mathbf{r}')\rangle^{1/2}}$$
(1)

is the field correlation function. It follows from the Bethe-Salpeter equation that

$$C_{\psi}(\mathbf{r},\mathbf{r}') = \frac{4\pi}{l} \int d\mathbf{r}_1 \langle G(\mathbf{r},\mathbf{r}_1) \rangle \langle G^*(\mathbf{r}',\mathbf{r}_1) \rangle, \qquad (2)$$

where  $\langle G(\mathbf{r}, \mathbf{r}_1) \rangle$  is the disorder-averaged Green's function. The meaning of Eq. (2) is that a correlation between points  $\mathbf{r}, \mathbf{r}'$  is established via scattering on an intermediate impurity, at  $\mathbf{r}_1$ . Then Eq. (2) can be reduced to

$$C_{\psi}(\mathbf{r},\mathbf{r}') = -\frac{4\pi c^3}{\epsilon^{3/2}\omega} \operatorname{Im}\langle G(\mathbf{r},\mathbf{r}')\rangle, \qquad (3)$$

which has the following intuitive explanation: field correlation between a pair of points is due primarily to waves that are scattered in the vicinity of one point and arrive at the vicinity of the other point. The average amplitude to arrive from  $\mathbf{r}'$  to  $\mathbf{r}$  is  $\langle G(\mathbf{r}, \mathbf{r}') \rangle$ , so that  $C_{\psi}(\mathbf{r}, \mathbf{r}')$  should be a linear combination of  $\langle G(\mathbf{r}, \mathbf{r}') \rangle$  and  $\langle G^*(\mathbf{r}, \mathbf{r}') \rangle$ . Equation (3) is the right linear combination since it reduces to unity for  $\mathbf{r} = \mathbf{r}'$ . Using Eq. (3), we present the field correlator as

$$C_{\psi}(\mathbf{r},\mathbf{r}') = -\frac{4\pi c^3}{\epsilon^{3/2}\omega} \operatorname{Im}\sum_{\mu,k} \frac{\psi_{\mu,k}(\mathbf{r})\psi_{\mu,k}^*(\mathbf{r}')}{\omega^2 - \omega_{\mu,k}^2 + i\eta},\qquad(4)$$

where  $\eta = c\omega/\epsilon^{1/2}l$ , with  $\epsilon$  standing for the background dielectric constant, and  $\psi^*_{\mu,k}(\mathbf{r})$ ,  $\omega_{\mu,k}$  are the photonic crystal eigenmodes and eigenfrequencies, respectively. They are characterized by the wave vector  $\mathbf{k}$  of the first Brillouin zone and the band index  $\mu = \pm 1$ .

Suppose that stop band corresponds to the direction of propagation along the *z* axis. Then, the anisotropy of  $C_{\psi}(\mathbf{r}, \mathbf{r}')$  is expected within a narrow angular interval,  $\theta = \arctan(z'-z)/(\rho'-\rho) \sim \gamma^{1/2}$ , where  $\gamma = \delta \omega / \omega \ll 1$  is the dimensionless frequency width of the

stop band. For such  $\theta$ , the main contribution to the sum in Eq. (4) comes from the small domain of **k** space around the Bragg condition  $k_z \approx Q_B/2$ , where  $2\pi/Q_B$  is the period along z (see Fig. 1). Within this domain, the dispersion law,  $\omega_{\mu,k}$ , can be simplified as

$$\frac{\epsilon \omega_{\mu,k}^2}{c^2} = k_\perp^2 + \frac{Q_B^2}{4} + \mu Q_B \sqrt{\frac{1}{L_B^2} + \left(k_z - \frac{Q_B}{2}\right)^2}, \quad (5)$$

where  $L_B = 4\omega/Q_B\delta\omega$  is the Bragg length of the decay along z exactly at the Bragg condition. The corresponding eigenmodes,  $\psi_{\mu,k}$ , can also be simplified:

$$\psi_{\mu,k} = \mathcal{U}_{\mu}(z) \exp[i(k_z - Q_B/2)z + ik_{\perp}\rho], \quad (6)$$

where  $\mathcal{U}_{\mu}$  are the Bloch functions. It is seen from Eq. (5) that, for a given  $\omega_{\mu,k} = \omega$  and the same  $k_z$ , the values of  $k_{\perp}$  are very different for upper  $\mu = 1$  and lower  $\mu = -1$  bands. As a result, the contribution to  $C_{\psi}$  from  $\mu = 1$  and  $\mu = -1$  differs exponentially in parameter  $\exp(-|\mathbf{r} - \mathbf{r}'|/L_B)$ . Then, keeping only  $\mu = -1$  terms, we can express the correlator  $C_{\psi}$  as

$$C_{\psi}(\mathbf{r},\mathbf{r}') = -\frac{c^{3} \mathcal{U}_{-1}(z) \mathcal{U}_{-1}(z')}{2\pi^{3} \epsilon^{3/2} \omega} \int dk_{z} \int k_{\perp} dk_{\perp} \int d\phi \\ \times \operatorname{Im} \left\{ \frac{\exp[iz(k_{z} - Q_{B}/2) + i\rho k_{\perp} \cos\phi]}{\omega^{2} - \omega_{-1,k}^{2} + i\eta} \right\}.$$
(7)

Substituting Eq. (5) into the integral Eq. (7) and performing the integration over  $k_{\perp}$  and  $\phi$ , we arrive at

$$C_{\psi}(\mathbf{r},\mathbf{r}') = \left(\frac{\gamma}{4}\right) \mathcal{U}_{-1}(z) \mathcal{U}_{-1}(z') \operatorname{Re}\left\{\int dq \exp\left[iq\frac{\gamma \mathcal{R}}{4}\right] \times H_0\left(\frac{\theta \mathcal{R}\gamma^{1/2}}{2}\sqrt{\Delta + \sqrt{1+q^2}}\right)\right\},\tag{8}$$

where  $H_0$  is a Hankel function of zero order. The dimensionless length  $\mathcal{R}$  and "detuning"  $\Delta$  are defined as

$$\mathcal{R} = |\mathbf{r} - \mathbf{r}'|Q_B, \qquad \Delta = \frac{2\omega}{\delta\omega} - \frac{cQ_B}{\epsilon^{1/2}\delta\omega} + i\frac{L_B}{2l}.$$
 (9)



FIG. 1. Surface of constant frequency (solid lines)  $\omega(k_z, k_\perp) = \omega$  is shown schematically for the Bragg frequency  $\omega = cQB/2\epsilon 1/2$ .

For  $\theta \mathcal{R} \gamma^{1/2} \gg 1$ , i.e.,  $|\mathbf{r} - \mathbf{r}'| \gg L_B$ , the asymptotic expansion of the Hankel function can be used so that the integral Eq. (8) takes the form

$$C_{\psi}(\mathbf{r}, \mathbf{r}') = \mathcal{U}_{-1}(z) \mathcal{U}_{-1}(z') \left[ \frac{\gamma^{3/4}}{2(\pi \mathcal{R})^{1/2}} \right] \\ \times \operatorname{Re} \left\{ e^{-i\pi/4} \int dq \frac{\exp[i\mathcal{R}F(\theta, q)]}{(\Delta + \sqrt{1 + q^2})^{1/4}} \right\}, \quad (10)$$

where  $F(\theta, q) = \gamma q/4 + \theta(\gamma^{1/2}/2)[\Delta + \sqrt{1+q^2}]^{1/2}$ . The function  $F(\theta, q)$  has a special point at which the first and the second derivatives with respect to Q, for some value of  $\theta = \theta_c$ , are zero. The position of this critical point,  $(\theta_c, q_c)$ , depends on the detuning  $\Delta$  and can be parametrized by a complex number w as

$$\Delta = \frac{4}{27}w^3 - w, \quad \theta_c = 2\gamma^{1/2} \left(\frac{w}{3}\right)^{3/2}, \quad q_c = \sqrt{\frac{4w^2}{9}} - 1.$$

For  $\Delta = 0$  we have  $w = 3^{3/2}/2$ ,  $\theta_c = (3^{3/4}/2^{1/2})\gamma^{1/2}$ , and  $q_c = 2^{1/2}$ . To evaluate the integral, Eq. (10), for  $\theta$  close to the critical point,  $\theta \sim \theta_c$ , the function  $F(\theta, q)$  should be expanded up to the third order in  $(q - q_c)$ :

$$F(\theta, q) = \phi_0 + \frac{\delta\theta}{\theta_t} + \delta q \frac{\delta\theta}{\theta_d} - \frac{1}{3} (\delta q)^3, \quad (11)$$

where  $\delta\theta = \theta - \theta_c$ ,  $\delta q = (9/4w)(6/w)^{1/3}(q - q_c)$ ,  $\phi_0 = \gamma (4w^2 - 9)^{3/2}/108$ , and characteristic angles  $\theta_t$ and  $\theta_d$  are defined as

$$\theta_t = \frac{6}{\gamma^{1/2}} \left( \frac{3}{4w^3 - 9w} \right)^{1/2}, \qquad \theta_d = 6 \left( \frac{2w}{3\gamma} \right)^{1/6}.$$
 (12)

Substituting Eq. (11) into Eq. (10) and performing integration over  $\delta q$ , we obtain

$$C_{\psi}(\mathbf{r},\mathbf{r}') = \left[\frac{2\gamma^{1/5}}{3\mathcal{R}}\right]^{5/6} \mathcal{U}_{-1}(z) \mathcal{U}_{-1}(z') \operatorname{Re}\left\{\frac{\pi^{1/2} w^{1/3}}{(4w^2 - 9)^{1/4}} \times \exp\left(i\phi_0 + i\mathcal{R}\frac{\delta\theta}{\theta_t}\right) \operatorname{Ai}\left(-\mathcal{R}^{2/3}\frac{\delta\theta}{\theta_d}\right)\right\},\tag{13}$$

where Ai is the Airy function.

Discussion.—It is easy to see that Eq. (13) has a form similar to the field distribution near the caustics, which is the envelope of rays reflected by a curved surface [21]. This similarity is not accidental. Indeed, near the caustics the rays cross over from geometrically allowed directions to the "shadow." Equation (13) also describes a crossover from the directions  $\theta \gg \theta_c \sim \gamma^{1/2}$  of free propagation to the angular domain  $\theta \leq \theta_c$  where propagation is forbidden due to the stop band, which plays the role of shadow in the *k* space. Similar to the case of caustics, the behavior of  $C_{\psi}$  near the crossover  $\theta = \theta_c$  is described by the following exponential decay for  $\theta < \theta_c$ :

$$C_{\psi}(\theta) \sim \exp\left[-\mathcal{R}\left(\frac{\theta_c - \theta}{\theta_d}\right)^{3/2}\right] \cos\left[\mathcal{R}\frac{\theta_c - \theta}{\theta_t} + \phi_0\right],$$

which, in the case of intensity correlation, serves as an envelope of the fast oscillations. For  $\theta > \theta_c$ , this envelope oscillates itself as  $\cos[\mathcal{R}(\theta - \theta_c/\theta_d)^{3/2} - \pi/4]$ . Overall, there are three angular scales in the correlator  $C_{\psi}$ , namely, the critical angle  $\theta_c$ , the period of the envelope  $\theta_d/\mathcal{R}^{2/3}$ , and the period  $\theta_t/\mathcal{R}$  of the oscillations. At spatial distances  $|\mathbf{r} - \mathbf{r}'| \sim L_B$ , all three scales are of the same order  $\sim \gamma^{1/2}$ . For large  $|\mathbf{r} - \mathbf{r}'|$ , we have  $\theta_c \gg \theta_d/\mathcal{R}^{2/3} \gg \theta_t/\mathcal{R}$ . Numerical examples illustrating separation of the three scales are given in Fig. 2. Outside the stop band  $\theta \gg \gamma^{1/2}$ , the correlator  $C_1$  saturates at the value  $\propto \exp(-|\mathbf{r} - \mathbf{r}'|/l)$ , governed by the mean free path [1]. Obviously, our main result, Eq. (13), applies in the vicinity of each of the equivalent Bragg directions.

While large-scale disorder (domains) broadens significantly the reflectivity peak  $R(\theta)$ , the dependence  $C_1(\theta)$  with all fine details *remains unchanged*. Even for purely short-range disorder, the intensity correlations are much more sensitive to the periodic background than the reflectivity. To illustrate this, in Fig. 3 we present the dependence  $C_1(\theta)$  together with  $\mathbf{R}(\theta)$ , which near the Bragg condition is given by  $\mathbf{R} = |(r_0 + s)/(1 + r_0 s)|^2$ , with  $r_0 = (\sqrt{\epsilon} - 1)/(\sqrt{\epsilon} + 1)$  and  $s(\theta) = g(\theta) - \sqrt{1 + g^2(\theta)}$ , where  $g(\theta) = L_B/2l + iL_BQ_B\theta^2/\epsilon$ . The dependencies  $\mathbf{R}(\theta)$  and  $C_1(\theta)$  were calculated for the effective refractive index  $\epsilon^{1/2} = 1.7$  and  $L_B = l$ . Similar to  $C_1$ , the width of  $\mathbf{R}(\theta)$  is  $\theta \sim \gamma^{1/2}$ . However,  $\mathbf{R}(\theta)$  is strongly smeared



FIG. 2. Correlator  $C_1$  as a function of dimensionless angle  $\theta/2(Q_BL_B)^{-1/2}$  is shown for  $|\mathbf{r} - \mathbf{r}'| = 5L_B$  and for two dimensionless detunings,  $\Delta$ , defined by Eq. (9); (a)  $\Delta = 0.15i$  (i.e., frequency is equal to the Bragg frequency and  $L_B = 0.3l$ ); (b)  $\Delta = 0.5 + 0.15i$ .



FIG. 3. (a) Correlator  $C_1$  as a function of dimensionless angle  $\theta/2(Q_BL_B)^{-1/2}$  is shown for  $L_B = l$  and two distances  $|\mathbf{r} - \mathbf{r'}| = 2l$  (curve 1) and  $|\mathbf{r} - \mathbf{r'}| = 3l$  (curve 2); (b) Angular dependence of the reflectivity is shown for  $L_B = l$  and effective refractive index  $\epsilon^{1/2} = 1.7$ .

when  $L_B = l$ , whereas  $C_1(\theta)$  contains oscillations. Three oscillations are well pronounced for the distances  $|\mathbf{r} - \mathbf{r}'|$ equal to 2l and 3l, as shown in Fig. 3. Since these values are comparable to  $L_B$ , the argument of the Hankel function in Eq. (8) is ~1. For this reason,  $C_1 = |C_{\psi}|^2$  was calculated directly from (8) without using the asymptotic form Eq. (10). Therefore, even when  $L_B \sim l$ , comparison of the saturation values of  $C_1(\theta)$  at two  $|\mathbf{r} - \mathbf{r}'|$  allows one to determine the value of the mean free path. Then  $L_B$ and, correspondingly, the stop-band width,  $\delta \omega =$  $2c/\epsilon^{1/2}L_B$ , can be inferred from the oscillations in the following way. Knowledge of the oscillation period,  $\pi \theta_t(w, L_B)/\mathcal{R}$ , together with frequency and mean free path, i.e., the ratios  $\operatorname{Re}\Delta(w)/L_B$  and  $\operatorname{Im}\Delta(w)/L_B$ , allows one to deduce the complex parameter w, and, finally,  $L_B = 2l\{\operatorname{Im}\Delta(w)\}.$ 

Conclusion.-In the present Letter, we have demonstrated that the intensity correlations in a disordered photonic crystal with *incomplete* band gap uncover the underlying band structure that is obscured by the disorder, so that conventional approaches applicable to the "clean" photonic crystals [22,23] fail. Note that the sensitivity of a number of disorder-induced effects, such as coherent backscattering [19,24] and directionality of the exiting diffusing light [25], to the periodic background has already been discussed in the literature. This sensitivity has also been employed [19,25] for the estimation of the stopband width. In this regard, the intensity correlations constitute a more accurate tool. Indeed, in the shape of the coherent backscattering cone, the properties of periodic structure enter only through the convolution with the Cooperon, which describes the large-scale diffusive motion, whereas the correlator  $C_1$  comes from short distances. Another advantage of using intensity correlations over traditional methods is that  $C_1$  is insensitive to the long-range disorder due to domains that mask the photonic band structure [13]. Earlier, it was demonstrated [26] that isolating a single domain and measuring reflection from this domain results in a drastic narrowing of the reflection spectra. We emphasize that correlation analysis of the speckle pattern allows one to get rid of inhomogeneous broadening in a natural way. To measure a certain incomplete gap (say, in the direction [111]), the near-field pattern on the plane parallel to this direction [e.g., (-110)] should be analyzed.

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