

Spectral changes of light produced by scattering from disordered anisotropic media

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We investigate theoretically changes in the spectrum of polychromatic light scattered by a disordered, birefringent medium. We derive an expression for the spectrum of scattered light for ordinary and extraordinary incident waves within the accuracy of the first Born approximation. Using this result, we analyze the changes in the spectrum of light due to the combined action of disorder and anisotropy in the scattering process. [S1063-651X(99)03909-4]

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I. INTRODUCTION

It has been over a decade since Wolf [1] predicted that the far-zone spectrum of polychromatic light emitted by a quasi-homogeneous source can differ from the source spectrum. These spectral changes, caused by correlations in the source, are now known as the Wolf effect. Because of the close analogy between the processes of radiation and scattering, similar changes in the spectrum of light scattered from random media have attracted a great deal of interest [2–6]. The first theoretical investigations focused on the spectral shifts generated by weak scattering in isotropic media [3]. Very recently, similar shifts have been studied both experimentally and theoretically in the static scattering of light from a random distribution of particles [7]. Considerable spectral changes were predicted in the multiple scattering of light from randomly rough surfaces [8,9]. Much of this work is reviewed in Ref. [10].

To date, the studies of spectral changes induced by scattering have dealt with optically isotropic media. In such media, the essential features of the spectral changes are well described by the scalar theory. In contrast, for an anisotropic medium, the vector nature of electromagnetic field plays a crucial role and must be taken into account. The scattering of *monochromatic* light in anisotropic media has been studied for both solid [11,12] and liquid crystals [13]. Therefore, it seems timely to address the problem of the spectral changes occurring in the scattering of *polychromatic* light in a disordered anisotropic medium. As in the isotropic case, the spectral information may provide useful characteristics of the scattering system.

In this paper we theoretically study the spectral changes induced by the scattering of polychromatic light in a birefringent dielectric medium with weak static disorder. We present the exact Green's tensor formulation of the scattering problem for a disordered anisotropic medium. We then use the first Born approximation to derive an expression for the far-zone spectrum of the scattered field. We find that this approximation captures the main effects introduced by the combined action of disorder and anisotropy, and yields pronounced spectral shifts. In addition, we demonstrate that two types of waves existing in a birefringent medium (ordinary and extraordinary) display different spectral changes due to

the scattering of these waves into secondary ordinary and extraordinary waves.

II. SCATTERING PROBLEM: PHYSICAL SYSTEM, SPECTRUM OF THE INCIDENT FIELD, AND BASIC EQUATIONS

We consider a static, disordered, anisotropic medium, described by the frequency-independent dielectric tensor,

$$\epsilon_{\alpha\beta}(\mathbf{r}) = \epsilon_{\alpha\beta}^{(0)} + \delta\epsilon_{\alpha\beta}(\mathbf{r}). \quad (1)$$

The unperturbed dielectric tensor $\epsilon_{\alpha\beta}^{(0)}$ describes a birefringent medium whose optical axis lies along the unit vector $\hat{\mathbf{s}}$, i.e.,

$$\epsilon_{\alpha\beta} = \epsilon_{\parallel} s_{\alpha} s_{\beta} + \epsilon_{\perp} (\delta_{\alpha\beta} - s_{\alpha} s_{\beta}), \quad (2)$$

where ϵ_{\perp} and ϵ_{\parallel} are the principal dielectric constants. We assume that the perturbation $\delta\epsilon_{\alpha\beta}(\mathbf{r})$ is isotropic, i.e., that $\delta\epsilon_{\alpha\beta}(\mathbf{r}) = \delta_{\alpha\beta} \delta\epsilon(\mathbf{r})$, and that $\delta\epsilon(\mathbf{r})$ is a zero-mean, stationary, Gaussian random process, defined by the correlation function,

$$\langle \delta\epsilon(\mathbf{r}) \delta\epsilon(\mathbf{r}') \rangle = B^2 e^{-|\mathbf{r}-\mathbf{r}'|^2/2\sigma^2}, \quad (3)$$

where B and σ are positive constants characterizing the strength and the effective correlation width of disorder, respectively. The angle brackets $\langle \rangle$ denote an average over the ensemble of realizations of disorder. Form (3) implies that the random scattering potential, defined by $\delta\epsilon_{\alpha\beta}(\mathbf{r})$, is statistically homogeneous and isotropic.

The scattering geometry is shown in Fig. 1. Our formulation of the problem of scattering of polychromatic light from disorder in an anisotropic medium is analogous to the isotropic case [3,6,7]. Since the medium is static, each frequency component $\mathbf{E}(\mathbf{r}, \omega)$ of the electric field is scattered independently of the others, and the scattering is described by the integral equation

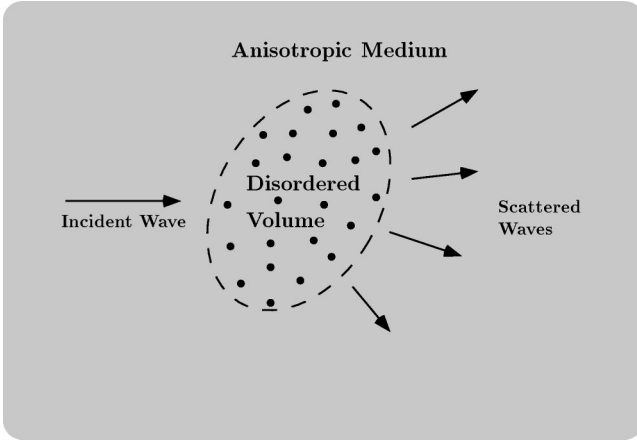


FIG. 1. The scattering geometry.

$$E_{\alpha}(\mathbf{r}, \omega) = E_{\alpha}^{(i)}(\mathbf{r}, \omega) + \frac{\omega^2}{c^2} \int_V d^3 r' G_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; \omega) \delta\epsilon_{\beta\gamma}(\mathbf{r}', \omega) E_{\gamma}(\mathbf{r}', \omega). \quad (4)$$

Here V is the scattering volume, the Greek indices assume the values 1, 2, 3, and summation over repeated Greek indices is implied throughout the paper, $G_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; \omega)$ is the electromagnetic Green's tensor for the unperturbed birefringent medium [11], and $E_{\alpha}^{(i)}(\mathbf{r}, \omega)$ is the α Cartesian component of the incident field. The incident field is assumed to be either an ordinary or an extraordinary plane wave:

$$\mathbf{E}_{1,2}^{(i)}(\mathbf{r}, \omega) = A(\omega) \mathbf{e}_{1,2}^{(i)} e^{i\mathbf{k}_{1,2}^{(i)} \cdot \mathbf{r}}, \quad (5)$$

where

$$\mathbf{k}_1^{(i)} = (\omega/c) \sqrt{\epsilon_{\perp}} (\sin \theta^{(i)} \cos \phi^{(i)}, \sin \theta^{(i)} \sin \phi^{(i)}, \cos \theta^{(i)}), \quad (6a)$$

$$\mathbf{k}_2^{(i)} = (\omega/c), (\sqrt{\epsilon_{\parallel}} \sin \theta^{(i)} \cos \phi^{(i)}, \sqrt{\epsilon_{\parallel}} \sin \theta^{(i)} \sin \phi^{(i)}, \sqrt{\epsilon_{\perp}} \cos \theta^{(i)}), \quad (6b)$$

define the wave vector of the incident wave with the lower index ‘‘1’’ corresponding to the ordinary wave and index ‘‘2’’ corresponding to the extraordinary wave here and throughout the paper. The polarization vectors are given by

$$\mathbf{e}_1^{(i)}(\mathbf{k}) = \epsilon_{\perp}^{-1/2} (-\sin \phi^{(i)}, \cos \phi^{(i)}, 0), \quad (7a)$$

$$\mathbf{e}_2^{(i)}(\mathbf{k}) = [(\cos^2 \theta^{(i)} / \epsilon_{\perp}) + (\sin^2 \theta^{(i)} / \epsilon_{\parallel})]^{-1/2} \times (\cos \theta^{(i)} \cos \phi^{(i)} / \epsilon_{\perp}, \cos \theta^{(i)} \sin \phi^{(i)} / \epsilon_{\perp}, -\sin \theta^{(i)} / \epsilon_{\parallel}). \quad (7b)$$

The spherical polar ($\theta^{(i)}$) and azimuthal ($\phi^{(i)}$) angles specify the direction of propagation of the incident wave with respect to the optical axis $\hat{\mathbf{s}}$ (chosen to be the z axis). The random function $A(\omega)$ in Eq. (5) defines the position-independent spectral density $S^{(i)}(\omega)$ of the statistically stationary incident field [14],

$$\langle A^*(\omega) A(\omega') \rangle_A = S^{(i)}(\omega) \delta(\omega - \omega'), \quad (8)$$

where the angle brackets $\langle \rangle_A$ denote an average over the ensemble of realizations of the field.

III. SPECTRUM OF THE SCATTERED FIELD

We now consider how the spectrum $S^{(i)}(\omega)$ of the incident ordinary or extraordinary polychromatic wave changes due to scattering by disorder. The spectrum $S^{(s)}(\mathbf{r}, \omega)$ of the scattered field $\mathbf{E}^{(s)}(\mathbf{r}, \omega)$ is defined by the relation [14]

$$\langle \langle E_{\alpha}^{(s)}(\mathbf{r}, \omega) E_{\alpha}^{*(s)}(\mathbf{r}, \omega') \rangle \rangle = S^{(s)}(\mathbf{r}, \omega) \delta(\omega - \omega'), \quad (9)$$

where the double brackets denote an average over the ensembles of realizations of disorder and of the incident field. The scattered field $\mathbf{E}^{(s)}(\mathbf{r}, \omega)$ is given by the second, integral term on the right-hand side of Eq. (4). We calculate it within the accuracy of the first Born approximation,

$$E_{\alpha}^{(s)}(\mathbf{r}, \omega) = \frac{\omega^2}{c^2} \int_V d^3 r' G_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; \omega) \delta\epsilon_{\beta\gamma}(\mathbf{r}', \omega) E_{\gamma}^{(i)}(\mathbf{r}', \omega). \quad (10)$$

This expression gives the dominant contribution to spectral shifts when the number of scattering events is small, and when $|\mathbf{E}^{(s)}(\mathbf{r}, \omega)| \ll |\mathbf{E}^{(i)}(\mathbf{r}, \omega)|$. Therefore, we expect this approximation to describe the main features caused by the combined action of disorder and anisotropy. We consider the far zone, where the expression for the Green's tensor $G_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; \omega)$ of the unperturbed birefringent medium takes the asymptotic form [11],

$$G_{\alpha\beta}(\mathbf{r}, \mathbf{r}', \omega) = \frac{\epsilon_{\perp}}{4\pi} \frac{e^{ik_1^{(s)} r}}{r} e^{-i\mathbf{k}_1^{(s)} \cdot \mathbf{r}'} e_{1\alpha}^{(s)} e_{1\beta}^{(s)} + \frac{\epsilon_{\parallel}}{4\pi} \frac{e^{ik_2^{(s)} \tilde{r}}}{\tilde{r}} e^{-i\mathbf{k}_2^{(s)} \cdot \tilde{\mathbf{r}}'} e_{2\alpha}^{(s)} e_{2\beta}^{(s)}, \quad (11)$$

where the vectors $\tilde{\mathbf{r}}$, $\mathbf{e}_{1,2}^{(s)}$, and $\mathbf{k}_{1,2}^{(s)}$ are expressed in terms of the spherical polar angles $\theta^{(s)}$ and $\phi^{(s)}$ for the observation point $\mathbf{r} = (r \sin \theta^{(s)} \cos \phi^{(s)}, r \sin \theta^{(s)} \sin \phi^{(s)}, r \cos \theta^{(s)})$. Further,

$$\tilde{\mathbf{r}}' = (r \sqrt{\epsilon_{\parallel}} \sin \theta^{(s)} \cos \phi^{(s)}, r \sqrt{\epsilon_{\parallel}} \sin \theta^{(s)} \sin \phi^{(s)}, \times r \sqrt{\epsilon_{\perp}} \cos \theta^{(s)}) / [\epsilon_{\perp} \cos^2 \theta^{(s)} + \epsilon_{\parallel} \sin^2 \theta^{(s)}]^{1/2},$$

$$\tilde{r} = r \sqrt{\cos^2 \theta^{(s)} + (\epsilon_{\parallel} / \epsilon_{\perp}) \sin^2 \theta^{(s)}},$$

the wave vectors $\mathbf{k}_1^{(s)}$ and $\mathbf{k}_2^{(s)}$ are given by Eqs. (6b) with the superscript i replaced by s , and the explicit expressions for the polarization vectors $\mathbf{e}_{1,2}^{(s)}$ are

$$\mathbf{e}_1^{(s)} = \epsilon_{\perp}^{-1/2} (-\sin \phi^{(s)}, \cos \phi^{(s)}, 0), \quad (12a)$$

$$\mathbf{e}_2^{(s)} = [\epsilon_{\perp} \cos^2 \theta^{(s)} + \epsilon_{\parallel} \sin^2 \theta^{(s)}]^{-1/2} \times (\cos \theta^{(s)} \cos \phi^{(s)}, \cos \theta^{(s)} \sin \phi^{(s)}, -\sin \theta^{(s)}). \quad (12b)$$

We next substitute $G_{\alpha\beta}$ from Eq. (11), and $(\mathbf{E}_1)_\gamma$ (for the incident ordinary wave) or $(\mathbf{E}_2)_\gamma$ (for the incident extraordinary wave) from Eq. (5), into Eq. (10). Using the resulting expression for the scattered field in Eq. (9), and taking the averages in Eq. (9) with the aid of Eqs. (3) and (8), we obtain the far-zone spectral density of the scattered field

$$S_j^{(s)}(\mathbf{r}, \omega) = \frac{VB^2}{(4\pi r)^2} \frac{\omega^4}{c^4} S^{(i)}(\omega) \times \left\{ \epsilon_\perp^2 |\mathbf{e}_j^{(i)} \cdot \mathbf{e}_1^{(s)}|^2 e^{-|\mathbf{k}_1^{(s)} - \mathbf{k}_j^{(i)}|^2 \sigma^2 / 2} + \frac{\epsilon_\parallel^2}{\cos^2 \theta^{(s)} + (\epsilon_\parallel / \epsilon_\perp) \sin^2 \theta^{(s)}} \times |\mathbf{e}_j^{(i)} \cdot \mathbf{e}_2^{(s)}|^2 e^{-|\mathbf{k}_2^{(s)} - \mathbf{k}_j^{(i)}|^2 \sigma^2 / 2} \right\}, \quad (13)$$

where $j=1$ if the incident wave is ordinary and $j=2$ if it is extraordinary. Equation (13) is the main result of the present paper. In the isotropic case ($\epsilon_\parallel = \epsilon_\perp$) it agrees with the earlier results of Refs. [3,7]. In the next section we analyze a combined action of disorder and anisotropy and illustrate our results by numerical examples.

IV. DISCUSSION AND NUMERICAL EXAMPLES

Formula (13) shows how the spectrum $S^{(i)}(\omega)$ of the incident field is modified by interaction with the scattering medium. The factors that determine these modifications are (i) Rayleigh scattering contribution ω^4 , (ii) spatial correlations of disorder, (iii) the type of the incident wave (ordinary or extraordinary), and (iv) the spatial orientation of the wave vectors and polarization vectors of the incident and scattered waves. We now illustrate the role of each of these factors quantitatively.

We assume a Gaussian form for the spectrum of the incident field,

$$S^{(i)}(\omega) = S_0 e^{-(\omega - \omega_0)^2 / 2\Gamma^2}, \quad (14)$$

where S_0 is a positive constant, and ω_0 and Γ represent the central frequency and the effective width of the spectrum $S^{(i)}(\omega)$, respectively. In numerical calculations, we will use the values $\omega_0 = 3.93 \times 10^{15} \text{ sec}^{-1}$ and $\Gamma = 0.05\omega_0$, corresponding to the parameters used in a recent experiment [7]. The normalized spectrum $S^{(i)}(\omega)$ calculated for these values is plotted in Figs. 2 and 3 in solid lines. The angles $\theta^{(i)}$ and $\phi^{(i)}$ defining the polarization of the incident wave and its direction of propagation with respect to the optical axis are taken to be 0° . The degree of anisotropy of the medium is characterized by the ratio $\epsilon_\parallel / \epsilon_\perp = 1.7$ [15].

We first demonstrate the importance of disorder correlations in the spectral changes. Assuming the incident wave to be extraordinary, we fix the direction of observation at $\theta^{(s)} = 60^\circ$ and $\phi^{(s)} = 0^\circ$ and calculate the spectrum of the scattered light from Eq. (13). The results are presented in Figs. 2 and 3 in dashed lines for two different correlation lengths of disorder: $\sigma\sqrt{\epsilon_\perp}\omega_0/c = 4$ for Fig. 2 and $\sigma\sqrt{\epsilon_\perp}\omega_0/c = 0.25$ for

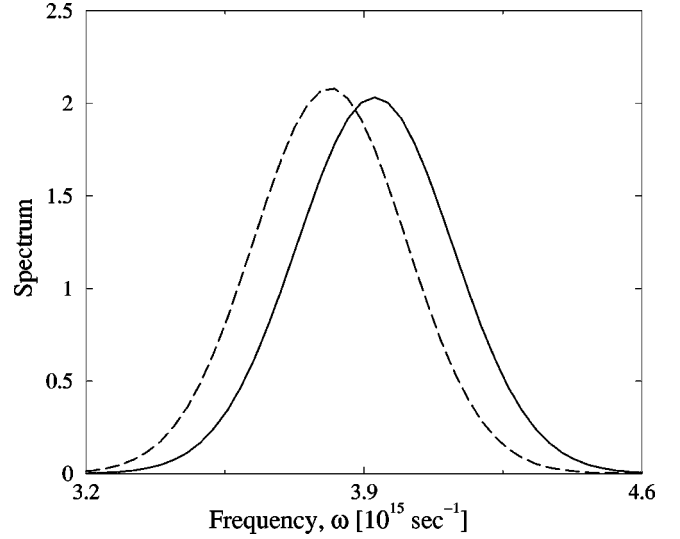


FIG. 2. Illustration of a scattering-induced red spectral shift. Dashed line: spectrum of light scattered in the direction ($\theta^{(s)} = 60^\circ$, $\phi^{(s)} = 45^\circ$) for an incident extraordinary wave with ($\theta^{(i)} = 0^\circ$, $\phi^{(i)} = 0^\circ$). Solid line: spectrum of the incident wave. The parameters used in calculation are $\epsilon_\parallel / \epsilon_\perp = 1.7$, $\omega_0 = 3.93 \times 10^{15} \text{ sec}^{-1}$, $\Gamma = 0.05\omega_0$, and $\sigma\sqrt{\epsilon_\perp}\omega_0/c = 4$.

Fig. 3. Figure 2 illustrates that in the case when the correlation length of disorder exceeds the typical wavelength of the polychromatic field, the Gaussian correlation functions play a dominant role in Eq. (13) causing a correlation-induced red spectral shift. In contrast, for correlation lengths smaller than the wavelength, the ω^4 factor in Eq. (13) is dominant, causing a blue spectral shift, as illustrated in Fig. 3. These conclusions are in qualitative agreement with the results of Refs. [3,6,7].

We next analyze the role of anisotropy in the formation of spectral shifts. To illustrate this role, we consider in-plane scattering $\phi^{(s)} = \phi^{(i)} = 0^\circ$ of an incident ordinary or extraordinary wave with the same spectrum as in Figs. 2 and 3. For the in-plane scattering the scattered wave is of the same type

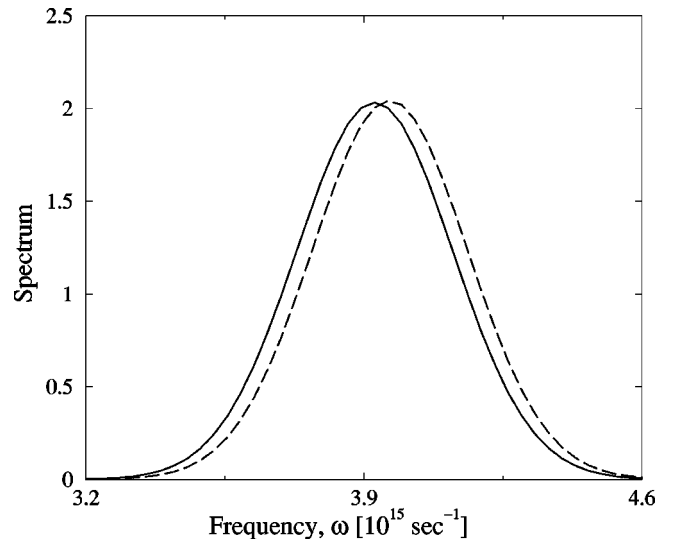


FIG. 3. Illustration of a scattering-induced blue spectral shift. The meaning of the curves and the values of the input parameters are the same as in Fig. 2, with the exception of $\sigma\sqrt{\epsilon_\perp}\omega_0/c = 0.25$.

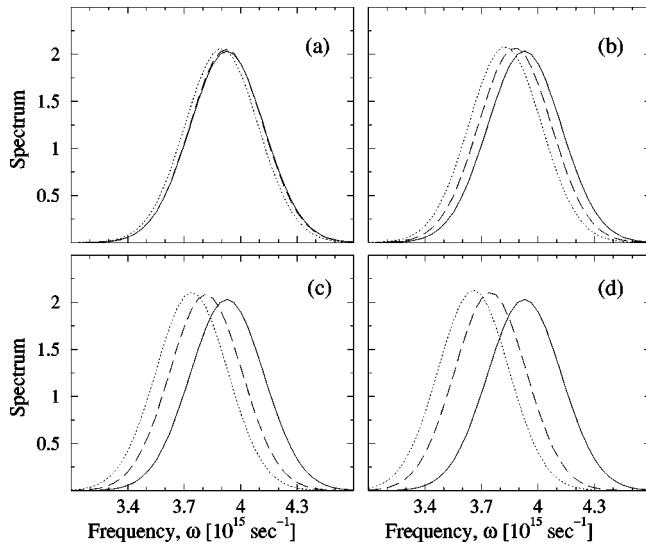


FIG. 4. Illustration of the role of anisotropy. Spectral shifts for in-plane scattering ($\phi^{(i)} = \phi^{(s)} = 0^\circ$), when the type of the scattered wave is the same as that for the incident wave. Solid lines, spectra of the incident waves; dashed lines, spectra of the scattered ordinary waves; dotted lines, spectra of the scattered extraordinary waves. (a)–(d) are plotted for four different polar scattering angles: (a) $\theta^{(s)} = 30^\circ$, (b) $\theta^{(s)} = 45^\circ$, (c) $\theta^{(s)} = 60^\circ$, and (d) $\theta^{(s)} = 75^\circ$. The other parameters are the same as in Fig. 2.

as the incident wave. Figures 4(a)–4(d), plotted for four different angles of scattering, clearly demonstrate that the changes in the spectra of ordinary and extraordinary waves are different. In particular, we see for the parameters used in our calculations that the spectral shifts for the extraordinary waves increase faster with increasing scattering angle than do the shifts for the ordinary waves. The large magnitude and strong dependence of the spectral shifts on the scattering angle would undoubtedly facilitate their experimental studies.

Further analysis shows that the two shifted spectral lines due to the two terms in the curly brackets of Eq. (13) could yield not merely a spectral shift, but a structural change of the incident wave spectrum (splitting of the spectral line). The observation of this effect would require, however, very

large values of the anisotropy parameter $\epsilon_{\parallel}/\epsilon_{\perp}$. This effect can also possibly be observed in the multiple-scattering regime, which is yet to be studied on the basis of exact Eq. (4).

V. CONCLUSIONS

We have studied spectral changes of light scattered by a static disorder in an anisotropic medium. Within the accuracy of the first Born approximation we obtained an expression for the spectrum [Eq. (13)] of scattered light for ordinary and extraordinary incident waves. We employed a simple model of disorder (isotropic, continuous) to illustrate the main effects introduced by anisotropy without complicating the presentation. However, it is straightforward to extend the result given in Eq. (13) to other types of disorder, e.g., orientational disorder in nematic liquid crystals — the only essential modification will be due to the change of the spatial correlation function $\langle \delta\epsilon_{\alpha\beta}(\mathbf{r}) \delta\epsilon_{\mu\nu}(\mathbf{r}') \rangle$. Spectrum (13) of the scattered field, derived in the single-scattering approximation, illustrates that scattering-induced spectral changes in complex systems, such as disordered anisotropic media, are determined by the interplay of many factors. This opens a possibility to control spectral properties by a proper choice of input parameters (type of the incident wave, disorder correlation length, etc.), which could be important in practical applications. Since the degree of anisotropy proved to play an important role in the formation of spectral changes produced by scattering, further studies of the Wolf effect in strongly anisotropic media are expected to be rewarding. For example, the Wolf effect may be used to study the fluctuations near the critical point of a phase transition in liquid crystals [16]. Finally, we hope that the analysis presented in this paper will stimulate further investigations of spectral changes in disordered anisotropic media.

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