Scattering mechanism for quadratic evolution of depolarization

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It was recently demonstrated theoretically, when the polarimetric properties of a material depend only upon the direction transverse to that of propagation (long coherence length regime), depolarization in transmission evolves quadratically with material thickness. This behavior was observed in several experimental studies. However, some of these studies unlikely satisfy the long coherence length condition under which the theory applies. Here, we demonstrate that abandoning a unidirectional approach to the propagation of light through a medium, i.e., introducing scatter, causes quadratic depolarization to occur in the short coherence length regime.

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The evolution of the intensity and polarization state of radiation transmitting through a medium is often treated in terms of a 4×4 Mueller matrix $\mathbf{M}(z)$ that evolves according to the differential equation [1–3]

$$\frac{\mathrm{d}\mathbf{M}(z)}{\mathrm{d}z} = \mathbf{m}\mathbf{M}(z), \tag{1}$$

where $\mathbf{m}$ is a 4×4 differential Mueller matrix, and z is the propagation coordinate. Implicit in the use of Eq. (1) is that the radiation is propagating in a single direction through the medium, as illustrated in Fig. 1(a). When $\mathbf{m}$ is independent of z, the solution is well known:

$$\mathbf{M}(z) = \exp(\mathbf{m}z), \tag{2}$$

where the matrix exponential is used [4], and the initial condition is $\mathbf{M}(0) = \mathbf{I}$ (the identity matrix). The behavior described by Eq. (2) motivates the logarithmic decomposition of a Mueller matrix,

$$\mathbf{L} = \log(\mathbf{M}), \tag{3}$$

in analogy with a Beer-Lambert law extinction coefficient [5, 6]. It is common to use the normalized Mueller matrix, $\mathbf{M}/M_{00}$, in place of $\mathbf{M}$ in Eq. (3), which just adds an additive component proportional to the identity matrix, and ensures $L_{00} = 0$.

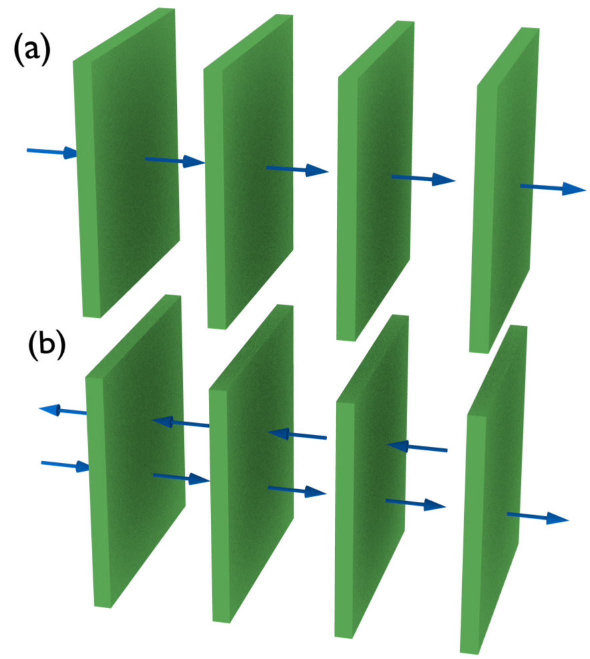

Fig. 1. Schematic illustrating the stacking of layers in transmission: (a) the traditional view, where only forward transmission is considered, and (b) the layers interacting through reflection or scattering.

Ossikovski and Arteaga found the physical interpretation of the matrix $\mathbf{m}$ to be related to the non-depolarizing mean properties $\langle\mathbf{m}\rangle$ and the depolarizing variances $\langle\Delta\mathbf{m}^2\rangle$ of the material properties [7]. Using elementary fluctuation theory and assuming that the material is homogeneous along the propagation direction (long coherence length limit), it was found that the differential Mueller matrix is given by [7, 8]

$$\mathbf{m} = \langle\mathbf{m}\rangle + \langle\Delta\mathbf{m}^2\rangle z. \tag{4}$$

The solution to Eq. (1) with Eq. (4) is approximately (since

$\langle \mathbf{m} \rangle$ and $\langle \Delta \mathbf{m}^2 \rangle$ do not, in general, commute) given by

$$\mathbf{M}(z) = \exp\left(\langle \mathbf{m} \rangle z + \frac{1}{2} \langle \Delta \mathbf{m}^2 \rangle z^2 \right). \tag{5}$$

Eq. (5) suggests that the depolarizing properties would behave quadratically with sample thickness in the long coherence length limit. In the short coherence length regime, the matrix $\mathbf{m}$ is independent of z [8], and depolarization would be expected to grow linearly.

Several studies have indeed observed quadratic evolution of depolarization, at least for small thicknesses [9–13]. However, in a number of these studies [9–11, 13], the materials being studied, or how their thickness was varied, would not have been expected to be in the long coherence length regime. Charbois and Devlaminck recognized this issue and developed an approach by which a linear or quadratic dependence can be observed [10]. They constructed measurements that showed an initial quadratic effect for thin samples and a linear effect for thick samples, in expectation of the transition from a thickness shorter than the coherence length to one greater than it. Agarwal *et al.* recognized that the results of their measurements were incompatible with a finite thickness of material being represented by the product of Mueller matrices for subdivided thin layers [9]. Yet, different sample thicknesses were created by stacking TiO_2-impregnated polyvinyl chloride blocks in [9] and by layering adhesive tape in [11]. Within fluctuation theory, if each layer of thickness z_0 is homogeneous along the propagation direction, the Mueller matrix of n independent layers (i.e., long coherence within a layer, but no coherence between layers) would be

$$\mathbf{M} = \exp\left(n \langle \mathbf{m} \rangle z_0 + \frac{n}{2} \langle \Delta \mathbf{m}^2 \rangle z_0^2 \right). \tag{6}$$

That is, if the fluctuation theory described the origin of depolarization in the layered samples, the growth of depolarization would have been linear with the number of layers.

In this paper, we suggest an alternative approach that explains the observed growth of depolarization with thickness, even in the short coherence length regime. As mentioned above, all of the previous analyses have assumed that radiation is only passing sequentially through the medium, as illustrated in Fig. 1(a). If the material is diffusely scattering, some radiation reflects backward, so that an approach illustrated in Fig. 1(b) would be more appropriate. The approach we take is a polarimetric extension of the theory of Kubelka and Munk (KM) for reflectance (but applied here in transmittance) and represents the simplest approximation to the radiative transfer equation [14, 15].

We begin by assuming that there are two streams, one propagating forward and one propagating backward, and that there is coupling between the two due to scattering. Following KM, but generalizing the absorption and scattering coefficients with matrices, we have

$$\frac{\mathrm{d}}{\mathrm{d}z} \begin{pmatrix} \mathbf{M}_+(z) \\ \mathbf{M}_-(z) \end{pmatrix} = \mathbf{m}' \begin{pmatrix} \mathbf{M}_+(z) \\ \mathbf{M}_-(z) \end{pmatrix}, \tag{7}$$

where

$$\mathbf{m}' = \begin{pmatrix} \mathbf{m} - \mathbf{a}(\mathbf{s}) & \mathbf{r}(\mathbf{s}) \\ -\mathbf{s} & \mathbf{r}[\mathbf{a}(\mathbf{s}) - \mathbf{m}] \end{pmatrix}, \tag{8}$$

$\mathbf{m}$ is the forward differential Mueller matrix [as for Eq. (1)], and $\mathbf{s}$ is a scattering Mueller matrix. The matrices $\mathbf{M}_+(z)$ and $\mathbf{M}_-(z)$ represent the Mueller matrices for forward and backward propagating radiation, respectively. The matrix function $\mathbf{a}(\mathbf{s})$ is needed to account for polarization-dependent losses in one direction as radiation is scattered into the other, and is a non-depolarizing, diattenuative, and lossy differential Mueller matrix:

$$\mathbf{a}(\mathbf{s}) = \begin{pmatrix} s_{00} & s_{01} & s_{02} & s_{03} \\ s_{01} & s_{00} & 0 & 0 \\ s_{02} & 0 & s_{00} & 0 \\ s_{03} & 0 & 0 & s_{00} \end{pmatrix}. \tag{9}$$

The matrix function $\mathbf{r}(\mathbf{m})$ relates the forward propagating optical properties to the backward propagating ones that result from the use of different coordinate systems for propagation in each direction [16]:

$$\mathbf{r}(\mathbf{m}) = \begin{pmatrix} m_{00} & m_{01} & -m_{02} & m_{03} \\ m_{10} & m_{11} & m_{12} & -m_{13} \\ -m_{20} & m_{21} & m_{22} & m_{23} \\ m_{30} & -m_{31} & m_{32} & m_{33} \end{pmatrix}. \tag{10}$$

The solution to Eq. (7) is

$$\begin{pmatrix} \mathbf{M}_+(z) \\ \mathbf{M}_-(z) \end{pmatrix} = \exp(\mathbf{m}'z) \begin{pmatrix} \mathbf{M}_+(0) \\ \mathbf{M}_-(0) \end{pmatrix}, \tag{11}$$

and we use the boundary conditions $\mathbf{M}_+(0) = \mathbf{I}$ (the incident beam), $\mathbf{M}_+(\Delta z) = \mathbf{M}_\mathrm{t}$ (the net Mueller matrix transmittance), $\mathbf{M}_-(0) = \mathbf{M}_\mathrm{r}$ (the net Mueller matrix reflectance), and $\mathbf{M}_-(\Delta z) = \mathbf{0}$ (representing the lack of an incident beam from the right). The thickness of the material is Δz. Reflectance and transmittance at the $z = 0$ and $z = \Delta z$ interfaces can be included with other boundary conditions but are not included here for simplicity.

As an illustrative example, we consider a material with absorption coefficient α and birefringence r, so that

$$\mathbf{m} = \begin{pmatrix} -\alpha & 0 & 0 & 0 \\ 0 & -\alpha & 0 & 0 \\ 0 & 0 & -\alpha & r \\ 0 & 0 & -r & -\alpha \end{pmatrix}, \tag{12}$$

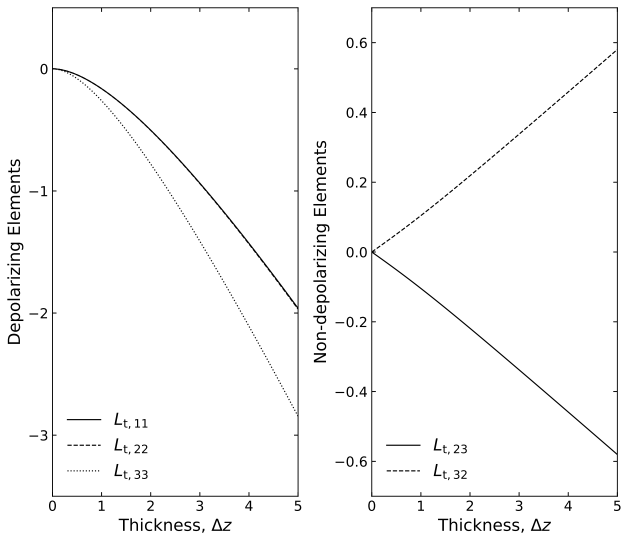

Fig. 2. The non-zero elements of the logarithmic decomposition $\mathbf{L}_t$ of the Mueller matrix transmittance $\mathbf{M}_t$ as a function of dimensionless thickness. The top frame shows the depolarizing elements $L_{t,11}$, $L_{t,22}$, and $L_{t,33}$, while the bottom frame shows the non-depolarizing elements $L_{t,23}$ and $L_{t,32}$. The curves for $L_{t,11}$ and $L_{t,22}$ are nearly identical.

and a scattering coefficient s and scattering depolarization c, so that

$$\mathbf{s} = \mathrm{diag}\left[s, sc, -sc, s(1-2c)\right], \tag{13}$$

which is the form expected for back-reflection [17]. Note here that, unlike Eq. (4), both $\mathbf{m}$ and $\mathbf{s}$ are local; they do not depend upon z. That is, they are being treated in the short coherence length regime. The only direct mechanism for depolarization is from scattering, if $c < 1$.

We further choose $\alpha = 10^{-6}$ (non-zero to prevent singular intermediate matrices), $r = 0.1$, $s = 1$, and $c = 0.7$. Non-zero birefringence ($r \neq 0$) is included here because some of the past measurements [11, 12] included samples with birefringence. Figure 2 shows the non-zero elements of the matrix $\mathbf{L}_t = \log(\mathbf{M}_t/M_{00})$. One immediately notices the curved nature of the depolarizing elements $L_{t,11}$, $L_{t,22}$, and $L_{t,33}$, which appear quadratic near zero thickness and become linear at larger thicknesses. This behavior was observed by Charbois and Devlaminck in their measurements of turbid liquids [10]. The non-depolarizing elements $L_{t,23}$ and $L_{t,32}$ are approximately linear with thickness, as has been observed in [11, 12].

Multiple scattering is the root of why depolarization behaves quadratically in this model. In order for scattering to affect radiation in transmission, there must be at least two scattering events: one to transfer radiation from the forward direction to the backward direction, and a second to transfer radiation back to the forward direction. The scattering parameter s is the product of the scatterer concentration and the mean backscattering cross section. The KM model described here exhibits a depolarization that also depends quadratically on s. This behavior was also found in [9], although it was expressed as the model-extracted standard deviation of the birefringence ($\langle \Delta \mathbf{m}^2 \rangle^{1/2}$, determined from Eq. (5)), found to be linear in particle concentration.

At this time, we do not know of a specific measurement that would distinguish between the behavior predicted by fundamental fluctuation theory [Eqs. (4) and (5)] and that resulting from scatter. It is expected that the measurements performed by stacking media [11, 12] would have achieved different results if the layers were spaced apart, since diffuse back reflections would be reduced. Furthermore, different results would be observed if the incident radiation were well collimated and only that radiation transmitted in the forward direction were collected, since contributions from scatter would be reduced. However, some of the artifacts that were studied, specifically those where materials were stacked to create different thicknesses (blocks of TiO_2 in polyvinyl chloride [9] or strips of adhesive tape [11]), would be expected to have correlation lengths shorter than the thinnest layer measured. That is, the polarimetric properties of the material could not have been coherent in the direction of propagation as blocks were stacked. Yet, those measurements observed the quadratic onset of depolarization.

Refs. [9, 11] acknowledged that, as layers were added, the radiation was attenuated. The KM theory described here not only predicts the polarization properties, but also net attenuation, which is found to be linear (that is, M_{00} is exponential) for small thicknesses. Attenuation can be included in the fluctuation theory through addition of an isotropic term in $\langle \mathbf{m} \rangle$, but attenuation does not arise naturally out of the fluctuation terms themselves.

One apparent paradox is solved by the present model: it was found that the results were not consistent with the multiplication of successive Mueller matrices implied by Fig. 1(a). Our resolution of that paradox is that scattering in the material causes radiation to propagate backward as well as forward. That is, the addition of a layer affects the radiation pattern in previous layers.

The polarimetric extension of the KM model we use here is highly simplified and does not represent a quantitative approach towards solving the radiative transfer equation [15]. Monte Carlo (MC) simulations better simulate the 3-dimensional nature of radiative transfer. In fact, quadratic depolarization was predicted by Monte Carlo simulations reported in [9] using scattering phase functions from Mie theory. Like the KM model described here, Monte Carlo simulations do not propagate radiation coherently, demonstrating that quadratic depolarization can be a scattering phenomenon, rather than a coherent one, like the fluctuation theory. We presented the KM approach, because it illustrates how simply adding scattering into the calculation results in a behavior similar to that observed in the measurements.

One of the experimental studies measured transmitted depolarization for different thicknesses sliced from bio-mimetic skin equivalents and focused on birefringent dermal regions. [12] These materials had thicknesses in the range 2 μm to 15 μm. Because the different samples were each obtained subtractively from blocks of material (in contrast to those studies which investigated different thickness by additively stacking layers), the condition of homogeneity of the polarimetric properties in the direction of propagation may have been satisfied. Thus, part or all of the quadratic behavior observed in the depolarization may indeed have been a result of the unidirectional approach that the authors used to interpret their data.

In this paper, we have shown that one does not need to assume longitudinal homogeneity (long coherence length) to observe quadratic evolution of depolarization in transmittance. Instead, the presence of scattering prevents application of a unidirectional application of Mueller matrices, illustrated in Fig. 1(a). By simply considering backward propagating radiation and scattering, quadratic evolution of depolarization can occur in the short coherence length limit. These results are important for interpreting depolarization in turbid media. Because there are two competing mechanisms that predict quadratic depolarization, extracting physical information from data may be difficult. It should be borne in mind that, because both scattering and depolarization result from inhomogeneities in a turbid media, scattering and depolarization are necessarily intertwined phenomena.

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