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# 3D digital technology differentiation of high-quality and low-quality organic polymers

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## ABSTRACT

A method of azimuthally invariant 3d Mueller-matrix mapping of the distributions of the parameters of phase and amplitude anisotropy of partially depolarizing layers of high-quality (group 1 - high density) and low-quality (group 2 - low density) polyethylene polymer films has been proposed and substantiated. layer-by-layer coordinate distributions of the magnitude of the set of Mueller-matrix invariants (MMI) of polymer films of both types were obtained in the volume of polymer samples.

**Keywords:** polarization; 3D Mueller matrix mapping; organic polymers; complex amplitude

## 1. INTRODUCTION

At present, optics is actively developing methods and means of polarimetric diagnostics of the structure of polymeric materials, which includes a number of original directions including computer-assisted methods:

- Mueller-matrix polarimetry<sup>1,2,3</sup>;

- two-dimensional Muller-matrix mapping in the framework of various model approximations<sup>4, 5, 6</sup>.

Our article is aimed at the development and experimental testing of a set of methods of Stokes-polarimetry and interferometry using algorithms for digital holographic reconstruction of the amplitude-phase structure of object fields<sup>7</sup> for differential diagnostics of layers of high-quality (group 1-high density) and low-quality (group 2 - low density) of films of polymer polyethylene by obtaining 3D distributions of Mueller-matrix invariants<sup>7,8,9</sup>.

## 2. BRIEF THEORY OF THE METHOD

### 2.1 Mueller-matrix invariants

For the Muller matrix  $\{M\}$ , the azimuthally invariant, independent of the angle ( $\Theta$ ) of rotation of the sample of the biological layer, are the following elements  $M_{ik}$  and their combinations:

$$\{M\} = \begin{vmatrix} 1 & M_{12} & M_{13} & M_{14} \\ M_{21} & M_{22} & M_{23} & M_{24} \\ M_{31} & M_{32} & M_{33} & M_{34} \\ M_{41} & M_{42} & M_{43} & M_{44} \end{vmatrix} \quad (1)$$

The MMI that characterizes the optical anisotropy of organic layers include<sup>10, 11, 12</sup>:

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- Matrix elements

$$M_{11}; M_{14}; M_{41}; M_{44} \quad (2)$$

- Combination of matrix elements

$$\Sigma \equiv (M_{22} + M_{33}) \quad (3)$$

$$\aleph \equiv (M_{23} - M_{32}) \quad (4)$$

- Lengths of matrix vectors

$$\begin{cases} A_h = \sqrt{M_{12}^2 + M_{13}^2}; \\ A_v = \sqrt{M_{21}^2 + M_{31}^2}; \\ B_h = \sqrt{M_{42}^2 + M_{43}^2}; \\ B_v = \sqrt{M_{24}^2 + M_{34}^2} \end{cases} \quad (5)$$

- Angles

$$\cos(B_h, B_v) = \frac{-\sqrt{(M_{42}^2 + M_{43}^2)}}{\sqrt{(M_{24}^2 + M_{34}^2)}} \quad (6)$$

$$\begin{cases} \{A_h\} = \frac{1}{\sqrt{M_{12}^2 + M_{13}^2}} \begin{pmatrix} M_{12}^2 - M_{13}^2 \\ 2M_{12}M_{13} \end{pmatrix}; \\ \{A_v\} = \frac{1}{\sqrt{M_{21}^2 + M_{31}^2}} \begin{pmatrix} M_{21}^2 - M_{31}^2 \\ 2M_{21}M_{31} \end{pmatrix}; \\ \{B_h\} = \frac{1}{\sqrt{M_{42}^2 + M_{43}^2}} \begin{pmatrix} M_{42}^2 - M_{43}^2 \\ 2M_{42}M_{43} \end{pmatrix}; \\ \{B_v\} = \frac{1}{\sqrt{M_{24}^2 + M_{34}^2}} \begin{pmatrix} M_{24}^2 - M_{34}^2 \\ 2M_{24}M_{34} \end{pmatrix} \end{cases} \quad (7)$$

$$G = \sqrt{(M_{22} - M_{33})^2 + (M_{23} - M_{32})^2} \quad (8)$$

The use of the MMI set will provide conditions for the dissemination of methods of experimentally reproducible Mueller-matrix mapping to serial, screening investigations<sup>13, 14, 15</sup>.

It is based on the use of a reference wave of laser radiation, which in the scheme of an optical interferometer is superimposed on a polarization-inhomogeneous image of a polymer film. The resulting interference pattern is recorded using a digital camera. With the help of diffraction integrals, the digital holographic reproduction of the distributions of the complex amplitudes  $\{E_x(x, y); E_y(x, y)\}$  of the object field of the polymer layer takes place<sup>16</sup>.

The set of elements of the Muller matrix is calculated by the following Stokes-polarimetric relations for the Stokes vectors of linearly polarized probing beams  $S^0(0^\circ); S^0(90^\circ)$ .

Traditionally, measuring the magnitude of a set of elements of the Mueller matrix includes the following experimental steps:

- formation of a series of probing linear and circularly polarized laser beams;

- for each of the probing beams, polarization filtering is carried out and the value of 4 parameters of the Stokes vector is calculated analytically;

- a series of Stokes vector parameters calculated for each of the probing beams serves as an analytical basis for determining 16 elements of the Mueller matrix of a phase-inhomogeneous layer:

$$\left[ \begin{array}{l} S^0(0^0) = \{M\} \begin{pmatrix} 1 \\ 1 \\ 0 \\ 0 \end{pmatrix} \rightarrow S(0^0) = \begin{pmatrix} M_{11} + M_{12} \\ M_{21} + M_{22} \\ M_{31} + M_{32} \\ M_{41} + M_{42} \end{pmatrix} \\ S^0(90^0) = \{M\} \begin{pmatrix} 1 \\ -1 \\ 0 \\ 0 \end{pmatrix} \rightarrow S(90^0) = \begin{pmatrix} M_{11} - M_{12} \\ M_{21} - M_{22} \\ M_{31} - M_{32} \\ M_{41} - M_{42} \end{pmatrix} \end{array} \right] \Rightarrow M_{ik} = \begin{vmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \\ M_{31} & M_{32} \\ M_{41} & M_{42} \end{vmatrix} \quad (9)$$

- for the Stokes vectors of linearly polarized probing beams  $S^0(45^0)$ ;  $S^0(135^0)$ :

$$\left[ \begin{array}{l} S^0(45^0) = \{M\} \begin{pmatrix} 1 \\ 0 \\ 1 \\ 0 \end{pmatrix} \rightarrow S(45^0) = \begin{pmatrix} M_{11} + M_{13} \\ M_{21} + M_{23} \\ M_{31} + M_{33} \\ M_{41} + M_{43} \end{pmatrix} \\ S^0(135^0) = \{M\} \begin{pmatrix} 1 \\ 0 \\ -1 \\ 0 \end{pmatrix} \rightarrow S(135^0) = \begin{pmatrix} M_{11} - M_{13} \\ M_{21} - M_{23} \\ M_{31} - M_{33} \\ M_{41} - M_{43} \end{pmatrix} \end{array} \right] \Rightarrow M_{ik} = \begin{vmatrix} M_{11} & M_{13} \\ M_{21} & M_{23} \\ M_{31} & M_{33} \\ M_{41} & M_{43} \end{vmatrix} \quad (10)$$

- and for the Stokes vectors of right and left circularly polarized probing beams  $S^0(\otimes)$ ;  $S^0(\oplus)$ :

$$\left[ \begin{array}{l} S^0(\otimes) = \{M\} \begin{pmatrix} 1 \\ 0 \\ 0 \\ 1 \end{pmatrix} \rightarrow S(\otimes) = \begin{pmatrix} M_{11} + M_{14} \\ M_{21} + M_{24} \\ M_{31} + M_{34} \\ M_{41} + M_{44} \end{pmatrix} \\ S^0(\oplus) = \{M\} \begin{pmatrix} 1 \\ 0 \\ 0 \\ -1 \end{pmatrix} \rightarrow S(\oplus) = \begin{pmatrix} M_{11} - M_{14} \\ M_{21} - M_{24} \\ M_{31} - M_{34} \\ M_{41} - M_{44} \end{pmatrix} \end{array} \right] \Rightarrow M_{ik} = \begin{vmatrix} M_{11} & M_{14} \\ M_{21} & M_{24} \\ M_{31} & M_{34} \\ M_{41} & M_{44} \end{vmatrix} \quad (11)$$

## 2.2 Methods for objective assessment of polarization maps

At the same time, such information is integrally averaged over the entire volume of the studied polymer<sup>17</sup>. Therefore, to analyze statistically distributed values of polarization parameters in a phase-inhomogeneous object field, an approach is used based on determining a set of statistical moments of the 1st - 4th orders, which characterize the mean, variance, asymmetry, and kurtosis of distributions of polarization parameters.

For an objective assessment of layer-by-layer polarization maps  $S(\theta_k, x, y)$ , the statistical moments of the first ( $Z_1$ ), second ( $Z_2$ ), third ( $Z_3$ ) and fourth ( $Z_4$ ) orders were used, which were calculated by the following algorithms<sup>7</sup>:

$$\begin{aligned}
Z_1 &= \frac{1}{N} \sum_{j=1}^N S(\theta_k, x, y)_j; \\
Z_2 &= \sqrt{\frac{1}{N} \sum_{j=1}^N (S^2(\theta_k, x, y))_j}; \\
Z_3 &= \frac{1}{Z_2^3} \frac{1}{N} \sum_{j=1}^N (S^3(\theta_k, x, y))_j; \\
Z_4 &= \frac{1}{Z_2^4} \frac{1}{N} \sum_{j=1}^N (S^4(\theta_k, x, y))_j,
\end{aligned} \tag{12}$$

where  $N$  – number of pixels of the photosensitive area of the CCD camera.

To carry out statistically reliable studies of the possibility of polarization differential diagnostics of polycrystalline films of organic polymers, representative groups of such samples were formed. Representativeness was assessed by cross-correlation analysis, which was based on achieving a standard deviation level of less than 0.025.

### 3. PRINCIPLES OF DIFFERENTIAL DIAGNOSIS OF POLYETHYLENE POLYMER FILMS

Begin the Introduction two lines below the Keywords. The manuscript should not have headers, footers, or page numbers. It should be in a one-column format. References are often noted in the text<sup>1</sup> and cited at the end of the paper.

In order to determine the diagnostic efficiency of the 3D Müller-matrix mapping method in differentiating layers of high-quality (group 1-high density) and low-quality (group 2 - low density) of polyethylene polymer films, two groups of partially depolarizing (degree of depolarization  $\Lambda \leq 50\%$ ) layers were formed:

- 26 samples – group 1 (attenuation coefficient  $0.79 < \tau < 0.85$ ,  $43\% < \Lambda < 48\%$ );
- 26 samples – group 2 ( $0.81 < \tau < 0.84$ ,  $45\% < \Lambda < 47\%$ ).

In the general case, the experimental use of Müller-matrix mapping of phase-inhomogeneous polymer layers turns out to be limited by the azimuthal dependence of the magnitude of most of the Müller matrix elements when the sample plane is rotated relative to the direction of the probe beam.

The search for and use of a series of so-called Müller matrix invariants as diagnostic parameters can become a solution to this technological problem.

These invariants include some matrix elements, combinations of elements, and mathematical vectors, the parameters of which are the elements of the Mueller matrix.

Optical technology for differential diagnosis of such samples includes the following steps:

1. Determination of a series of "phase" layer-by-layer images of 3D MMI distributions  $\{M_{44}; \Delta M; M_{41}; M_{14}\}(\varphi_1=0, 3; 2\varphi_1, \dots, 6\varphi_1)$  characterizing volumetric polarization manifestations of phase and amplitude anisotropy within both groups of samples.
2. For each "phase" section of 3D distributions of the MMI value, a set of statistical moments of the 1st - 4th orders is calculated  $Z_{i=1;2;3;4}\{[M_{44}; \Delta M; M_{41}; M_{14}](\varphi_k, x, y)\}$ .
3. For samples of group 1 and group 2, "phase" dependences  $Z_{i=1;2;3;4}\{[M_{44}; \Delta M; M_{41}; M_{14}](\varphi_1, \varphi_2, \dots, \varphi_k)\}$  of the magnitude of each statistical moment are plotted.
4. The "phase" planes ( $\varphi^*$ ) are determined in 3D MMI distributions, where the maximum differences between the values of the statistical moments ( $\Delta Z_{i=1;2;3;4}^* \equiv \Delta Z_{i=1;2;3;4}(\varphi^*) \rightarrow \max$ ), which characterize the distributions of the values of matrix elements  $M_{44}; \Delta M; M_{41}; M_{14}$  in these planes, are realized.
5. In the "phase" plane  $\varphi^*$ , the average  $\Delta Z_{i=1;2;3;4}^*$  and the error  $\sigma(\Delta Z_{i=1;2;3;4}^*)$  are determined within the polymer films from group 1 and group 2 (see Table 1).

Table 1. Statistical criteria for the differentiation of polymer films based on phase anisotropy.

Parameters	Group 1		Group 2		Accuracy, A, %	
	$M_{44}$	$\Delta M$	$M_{44}$	$\Delta M$	$M_{44}$	$\Delta M$
$Z_1 (\varphi^* = 0.45)$	$0,29 \pm 0,017$	$0,12 \pm 0,007$	$0,44 \pm 0,029$	$0,07 \pm 0,004$	85	82
$Z_2 (\varphi^* = 0.45)$	$0,21 \pm 0,012$	$0,15 \pm 0,008$	$0,14 \pm 0,006$	$0,11 \pm 0,005$	81	79
$Z_3 (\varphi^* = 0.45)$	$0,46 \pm 0,029$	$0,63 \pm 0,041$	$0,69 \pm 0,037$	$0,92 \pm 0,055$	91	89
$Z_4 (\varphi^* = 0.45)$	$0,57 \pm 0,033$	$0,88 \pm 0,053$	$1,03 \pm 0,059$	$1,39 \pm 0,084$	92	87

#### 4. CONCLUSIONS

A method of azimuthally invariant 3D Mueller matrix mapping of the distributions of the parameters of phase and amplitude anisotropy of partially depolarizing layers of qualitative (group 1-high density) and low-quality (group 2 - low density) polyethylene polymer films is proposed and substantiated.

Layer-by-layer coordinate distributions of the set of Mueller-matrix invariants of polyethylene were obtained in the volume of film samples.

The "phase" depends on the magnitude of the statistical moments of the 1st - 4th orders, characterizing the distributions of the MMI values of the polarization manifestations of the parameters of the linear and circular birefringence and dichroism of the polycrystalline component of various types of polyethylene layers have been determined.

The optimal conditions for the differentiation of polycrystalline structures of polymer layers of different densities - the range of phase cross-sections and the most sensitive parameters - statistical moments of the 3rd and 4th orders, characterizing the distributions of MMI are revealed.

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