Supplemental Document

Optics Letters

Eigenvalue calibration method for dual rotating-compensator Mueller matrix polarimetry: supplement

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Supplement DOI: https://doi.org/10.6084/m9.figshare.15164154

Parent Article DOI: https://doi.org/10.1364/OL.437542

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1. Model-based calibration method (MCM) for DRC-MMP

The MCM relies on establishing a system model for the DRC-MMP first. The calibration is then done by exploring the explicit relationship between the calibration parameters and the Fourier coefficients of the modulated light intensity [1], or fitting the measured quantities to their counterpart obtained from the system model [2-4]. To be more specific, Eq. (1) can be generally rewritten in terms of Fourier series as

$$I(t) = I_0 + \sum_{n=1}^{N_{\max}} \left(\alpha_{2n} \cos 2n\omega t + \beta_{2n} \sin 2n\omega t \right), \tag{S1}$$

where $\{I_0, (\alpha_{2n}, \beta_{2n})$, $n = 1, 2, ..., N_{\max}\}$ are the Fourier coefficients of the modulated light intensity, and $N_{\max}$ denotes the maximum Fourier components that is determined by the rotary speed ratio of the two compensators. For the common ratios of 5:1 and 5:3, $N_{\max} = 12$ and 16, respectively [1]. For brevity, we denote the Fourier coefficients of the modulated light intensity as a column vector $\mathbf{\Gamma} = [\alpha_{2n}, \beta_{2n}]^{\mathrm{T}}$. The MCM based on the nonlinear regression can then be formulated as

$$\arg\min_{\boldsymbol{x}} \left\| \mathbf{\Gamma}_{\mathrm{calc}}(\boldsymbol{x}) - \mathbf{\Gamma}_{\mathrm{meas}} \right\|_2 , \tag{S2}$$

where $\|\cdot\|_2$ represents L_2-norm, $\boldsymbol{x}$ is the vector that consists of the calibration parameters, and $\mathbf{\Gamma}_{\mathrm{meas}}$ and $\mathbf{\Gamma}_{\mathrm{calc}}$ denote the Fourier coefficients that are directly obtained from the collected light intensity and those obtained according to the established system model, respectively. In the implementation of the MCM, the calibration samples can be air or SiO_2/Si thin films.

Note that to achieve accurate calibration the established system model in MCM should take into account of all the optical components that may change polarization states. For example, for DRC-MMP that contains not only polarizers and compensators, but also optical components that change polarization states, such as beam splitters and high numerical aperture objective lenses [4, 5], the implementation of MCM requires parameterizing all these components first and then solving Eq. (S2).

2. The relationship between the eigenvalues of matrices $\mathbf{C}_{\mathrm{f}i}$ ($\mathbf{C}_{\mathrm{b}i}$) and $\mathbf{M}_{\mathrm{f}i}$ ($\mathbf{M}_{\mathrm{b}i}$)

For the single-pass system, it is very clear that the eigenvalues of $\mathbf{C}_{\mathrm{f}i}$ ($\mathbf{C}_{\mathrm{b}i}$) are equal to the eigenvalues of $\mathbf{M}_{\mathrm{f}i}$ ($\mathbf{M}_{\mathrm{b}i}$), but in the double-pass system, the relationship between the eigenvalues are more complicated. According to Eq. (8), $\mathbf{M}_{\mathrm{f}i}$ equals to $\mathbf{M}_{\mathrm{b}i}$ in the double-pass system so they are uniformly denoted by $\mathbf{M}_i$ here, and $\mathbf{C}_{\mathrm{f}i}$ is denoted by $\mathbf{C}_i$. Figure 1(b) shows the double-pass setup of the dual rotating-compensator MMP. It's very easy to know the intensity projection matrix is given by

$$\mathbf{D}_i = \mathbf{A}\mathbf{M}_i\mathbf{M}_0\mathbf{M}_i\mathbf{W}. \tag{S3}$$

Since $\mathbf{A}$ is a column full-rank matrix, $\mathbf{W}$ is a row full-rank matrix, and the inverse of a non-square matrix is regarded as its Moore-Penrose pseudo-inverse matrix, there is

$$\mathbf{C}_i = \mathbf{D}_0^{+}\mathbf{D}_i = \mathbf{W}^{+}\mathbf{M}_0^{+}\mathbf{A}^{+}\mathbf{A}\mathbf{M}_i\mathbf{M}_0\mathbf{M}_i\mathbf{W} = \mathbf{W}^{+}\mathbf{M}_0^{+}\mathbf{M}_i\mathbf{M}_0\mathbf{M}_i\mathbf{W} , \tag{S4}$$

where $\mathbf{M}_i$ represents the Mueller matrix of the calibration sample containing the azimuth angle, and it can also be represented by the rotation matrix $\mathbf{R}(\theta)$ and the sample Mueller matrix without the azimuth angle $\mathbf{M}_i'$ as

$$\mathbf{M}_i = \mathbf{R}(-\theta)\mathbf{M}_i'\mathbf{R}(\theta) . \tag{S5}$$

The Mueller matrix of the mirror $\mathbf{M}_0$ can be expressed as

$$\mathbf{M}_0 = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & -1 \end{bmatrix} . \tag{S6}$$

Substituting Eqs. (S5) -(S6) into Eq. (S4), Eq. (S4) can be expressed as

$$\mathbf{C}_i = \mathbf{W}^{+}\mathbf{R}(\theta)\mathbf{M}_i'\mathbf{M}_i'\mathbf{R}(-\theta)\mathbf{W} . \tag{S7}$$

Observing the characteristic equation of $\mathbf{M}_i'$ and the characteristic equation of $\mathbf{M}_i' \cdot \mathbf{M}_i'$ shown in Eq. (S8), we can know that the eigenvalue of $\mathbf{M}_i' \cdot \mathbf{M}_i'$ is the square of the eigenvalues of $\mathbf{M}_i'$.

$$\begin{aligned} \mathbf{M}_i' \cdot \mathbf{X} &= \lambda \mathbf{X} \\ \mathbf{M}_i' \cdot \mathbf{M}_i' \cdot \mathbf{X} &= \mathbf{M}_i' \cdot \lambda \mathbf{X} = \lambda^2 \mathbf{X} \end{aligned} \tag{S8}$$

Therefore, the eigenvalues of the matrix $\mathbf{C}_i$ are the square of the eigenvalues of the Mueller matrix of the calibration sample. The Mueller matrix of the calibration sample can be solved by the eigenvalues of the Mueller matrix. In order to facilitate the solution, a polarizer or waveplate is usually selected as the calibration sample.

As stated in the manuscript, matrix $\mathbf{C}_i$ has five eigenvalues, but the Mueller matrix of the calibration sample only has four eigenvalues. In order to accurately find the Mueller matrix of the calibration sample, the eigenvalues of matrix $\mathbf{C}_i$ must be studied.

A $4\times n$-dimensional matrix can be written as a combination of n column vectors, so we express a general form of the modulation matrix as

$$\begin{aligned} \mathbf{W}_{4\times n} &= [\boldsymbol{\eta}_1 \quad \boldsymbol{\eta}_2 \quad \boldsymbol{\eta}_3 \quad \cdots \quad \boldsymbol{\eta}_n] \\ \mathbf{W}_{4\times n}^{+} &= [\boldsymbol{\xi}_1 \quad \boldsymbol{\xi}_2 \quad \boldsymbol{\xi}_3 \quad \cdots \quad \boldsymbol{\xi}_n]^{\mathrm{T}} \end{aligned} , \tag{S9}$$

where $\boldsymbol{\xi}_i$ and $\boldsymbol{\eta}_i$ are 4×1 column vectors. And there are at most four linearly independent four-dimensional vectors that can form a set of basis vectors. Therefore, the following relationship holds:

$$\begin{aligned} \boldsymbol{\eta}_i &= q_1\boldsymbol{\eta}_1 + q_2\boldsymbol{\eta}_2 + q_3\boldsymbol{\eta}_3 + q_4\boldsymbol{\eta}_4 \quad (i = 1,2,3,...,n) \\ \boldsymbol{\xi}_i &= p_1\boldsymbol{\xi}_1 + p_2\boldsymbol{\xi}_2 + p_3\boldsymbol{\xi}_3 + p_4\boldsymbol{\xi}_4 \quad (i = 1,2,3,...,n) \end{aligned} . \tag{S10}$$

Then the equation of $\mathbf{C}_{n\times n} = \mathbf{W}_{4\times n}^{+}\mathbf{M}_{4\times 4}\mathbf{W}_{4\times n}$ can be expressed as

$$\mathbf{C}_{n\times n} = \begin{bmatrix} \boldsymbol{\xi}_1\mathbf{M}\boldsymbol{\eta}_1 & \boldsymbol{\xi}_1\mathbf{M}\boldsymbol{\eta}_2 & \boldsymbol{\xi}_1\mathbf{M}\boldsymbol{\eta}_3 & \cdots & \boldsymbol{\xi}_1\mathbf{M}\boldsymbol{\eta}_n \\ \boldsymbol{\xi}_2\mathbf{M}\boldsymbol{\eta}_1 & \boldsymbol{\xi}_2\mathbf{M}\boldsymbol{\eta}_2 & \boldsymbol{\xi}_2\mathbf{M}\boldsymbol{\eta}_3 & \cdots & \boldsymbol{\xi}_2\mathbf{M}\boldsymbol{\eta}_n \\ \boldsymbol{\xi}_3\mathbf{M}\boldsymbol{\eta}_1 & \boldsymbol{\xi}_3\mathbf{M}\boldsymbol{\eta}_2 & \boldsymbol{\xi}_3\mathbf{M}\boldsymbol{\eta}_3 & \cdots & \boldsymbol{\xi}_3\mathbf{M}\boldsymbol{\eta}_n \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ \boldsymbol{\xi}_n\mathbf{M}\boldsymbol{\eta}_1 & \boldsymbol{\xi}_n\mathbf{M}\boldsymbol{\eta}_2 & \boldsymbol{\xi}_n\mathbf{M}\boldsymbol{\eta}_3 & \cdots & \boldsymbol{\xi}_n\mathbf{M}\boldsymbol{\eta}_n \end{bmatrix} . \tag{S11}$$

According to Eq. (S10), each row from the 5th-row to the n^{th}-row of matrix **C** can be expressed as a linear combination of the first four rows of matrix **C**, that is, the value of any fifth-order sub-form determinant of **C** is 0. Therefore, the rank of matrix C is 4, which means that compared to the sample's Mueller matrix, the extra eigenvalues of matrix **C** must be 0.

To sum up, what is different from traditional ECM is that in the single-pass system, the non-zero eigenvalue of $\mathbf{C}_i$ is equal to the eigenvalue of $\mathbf{M}_i$, while in the double-pass system, the non-zero eigenvalues of $\mathbf{C}_i$ is the square of the eigenvalues of $\mathbf{M}_i$.

3. Calibration of the double-pass system by ECM

As shown in Fig. 1(b), the double-pass system adopts a BS to realize the purpose of sharing the optical path between the PSG and PSA. The ECM only needs to measure four calibration samples in sequence to complete the system calibration, with the polarization effect of the BS considered automatically. The calibration samples were placed on the calibration plane (CP1 or CP2) between the mirror and the BS shown in Fig. 1(b). Here, the calibration samples are air, a polarizer with an azimuth angle of 0° (P0), a polarizer with an azimuth angle of about 90° (P90), and a one-eighth waveplate (Customized wave plate, Union Optic) with an azimuth angle of about 30° (C30).

During the experiment, we roughly adjust the calibration samples to the azimuth angles mentioned above with respect to the plane of incidence according to the label associated with each calibration sample indicating its transmission axis or fast axis. It should be pointed out that here we only need to know the approximate orientation of these angles, because we will finally re-calculate their actual angles in the calibration by ECM.

We first place sample P0 on the CP1 to complete the measurement. And then the response of sample P90 or C30 can be obtained respectively by replacing P0 with P90 or C30. After removing the above calibration samples, the null response of the system is obtained by measuring a reflection mirror. The light intensity projection process is then performed according to the Eqs. (1)-(3) and Eq. (14), and we can obtain the light intensity projection matrix of different calibration samples. After that, we find the eigenvalues of Mueller matrices of calibration samples by performing eigenvalue analysis according to Section 2 of the Supplement 1. Finally, the azimuth angles of the calibration samples are determined by minimizing $\lambda_{20}/\lambda_{19}$ of matrix in Eq. (13). And the zero eigenvectors of $\mathbf{K}_{\mathrm{tot}}$ and $\mathbf{K}'_{\mathrm{tot}}$ can be rearranged to obtain the modulation matrix **W** and the analysis matrix **A** respectively.

The calibration steps are summarized as follows:

Step 1: Install the calibration samples, and roughly adjust the azimuth angle of each calibration sample.

Step 2: Each calibration sample (air, P0, P90, C30) should be measured individually for calibration.

Step 3: Perform the light intensity projection process according Eqs. (1)-(3) and (14).

Step 4: Mueller matrices of calibration samples are solved by Eqs. (5)-(9). And azimuth angles of calibration samples are determined by Eqs. (10)-(13) as well as Section 2 of the Supplement 1.

Step 5: Rearrange the zero eigenvectors of $\mathbf{K}_{\mathrm{tot}}$ and $\mathbf{K}'_{\mathrm{tot}}$ in Eq. (13) to obtain the modulation matrix **W** and the analysis matrix **A**, respectively.

4. Calibration of the double-pass system by MCM

As shown in Fig. 1(b), the double-pass system adopts a BS to realize the purpose of sharing the optical path between the PSG and PSA. The multi-layer films in the BS will induce a shift between s- and p-polarizations in both transmission and reflection setups and thus change the polarization state, even though the BS is declared to be non-polarizing [4, 5]. We use $\mathbf{M}^{\mathrm{t}}_{\mathrm{BS}}$ and

$\mathbf{M}_{\mathrm{BS}}^{\mathrm{r}}$ to denote Mueller matrices of the BS in transmission and reflection setups, respectively. The sample Mueller matrix $\mathbf{M}_{\mathrm{S}}$ in Eq. (1) for the double-pass system should be

$$\mathbf{M}_{\mathrm{s}} = \mathbf{M}_{\mathrm{BS}}^{\mathrm{t}} \cdot \mathbf{M}_{\mathrm{s}}^{\mathrm{real}} \cdot \mathbf{M}_{\mathrm{BS}}^{\mathrm{r}}, \tag{S12}$$

where $\mathbf{M}_{\mathrm{s}}^{\mathrm{real}}$ is the Mueller matrix of the sample without the polarization effect of the BS.

To achieve accurate calibration for the double-pass system by MCM, we should first parameterize the BS. Assuming that the multi-layer films in the BS is isotropic, the polarization effect of the BS can thereby be parameterized with four parameters: Ψ_{bt}, Ψ_{br}, Δ_{bt}, and Δ_{br}, where Ψ_{bt} and Δ_{bt} denote the amplitude ratio and phase retardance in transmission setup, respectively, and Ψ_{br} and Δ_{br} denote the amplitude ratio and phase retardance in reflection setup, respectively. Thus, $\mathbf{M}_{\mathrm{BS}}^{\mathrm{t}}$ and $\mathbf{M}_{\mathrm{BS}}^{\mathrm{r}}$ can be represented by

$$\mathbf{M}_{\mathrm{BS}}^{\mathrm{t}} = \begin{bmatrix} 1 & -\cos 2\Psi_{\mathrm{bt}} & 0 & 0 \\ -\cos 2\Psi_{\mathrm{bt}} & 1 & 0 & 0 \\ 0 & 0 & \sin 2\Psi_{\mathrm{bt}} \cos \Delta_{\mathrm{bt}} & \sin 2\Psi_{\mathrm{bt}} \sin \Delta_{\mathrm{bt}} \\ 0 & 0 & -\sin 2\Psi_{\mathrm{bt}} \sin \Delta_{\mathrm{bt}} & \sin 2\Psi_{\mathrm{bt}} \cos \Delta_{\mathrm{bt}} \end{bmatrix}, \tag{S13}$$

$$\mathbf{M}_{\mathrm{BS}}^{\mathrm{r}} = \begin{bmatrix} 1 & -\cos 2\Psi_{\mathrm{br}} & 0 & 0 \\ -\cos 2\Psi_{\mathrm{br}} & 1 & 0 & 0 \\ 0 & 0 & \sin 2\Psi_{\mathrm{br}} \cos \Delta_{\mathrm{br}} & \sin 2\Psi_{\mathrm{br}} \sin \Delta_{\mathrm{br}} \\ 0 & 0 & -\sin 2\Psi_{\mathrm{br}} \sin \Delta_{\mathrm{br}} & \sin 2\Psi_{\mathrm{br}} \cos \Delta_{\mathrm{br}} \end{bmatrix}, \tag{S14}$$

respectively. Taking into account of the calibration parameters associated with the polarizers and compensators as well as the four parameters of the BS and solving Eq. (S1), we can finally obtain the Mueller matrix of the mirror as follows

$$\mathbf{M}_{\mathrm{S}}^{\mathrm{real}} = \begin{bmatrix} 1.0000 & 0.0024 & 0.0023 & -0.0069 \\ 0.0024 & 0.9998 & 0.0008 & 0.0017 \\ -0.0015 & 0.0006 & -0.9947 & 0.0074 \\ 0.0060 & 0.0032 & -0.0070 & -0.9958 \end{bmatrix}. \tag{S15}$$

According to the analysis results of mirror, the Mueller matrix element error of the mirror calculated by the MCM does not exceed 0.01 after compensating for the polarization effect of the BS. Therefore, we can conclude that the polarization effect of the non-polarizing BS is not negligible, but the system is too complex for the MCM to model it. In contrast, the ECM, as a model-free calibration method, is very suitable for this complex ellipsometry system.

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