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Published on: 01 Mar 2019 - Journal of The Optical Society of America A-optics Image Science and Vision (Optical Society of America)

Topics: Mueller calculus and Ellipsometry

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Complete Mueller matrix from a partial polarimetry experiment: the twelve-element case

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Abstract: Conventional generalized ellipsometry instrumentation is capable of measuring twelve out of the sixteen elements of the Mueller matrix of the sample. The missing column (or row) of the experimental partial Mueller matrix can be analytically determined under additional assumptions. We identify the conditions necessary for completing the partial Mueller matrix to a full one. More specifically, such a completion is always possible if the sample is nondepolarizing; the fulfilment of additional conditions, such as the Mueller matrix exhibiting symmetries or being of special two-component structure, are necessary if the sample is depolarizing. We report both algebraic and numerical procedures for completing the partial twelve-element Mueller matrix in all tractable cases and validate them on experimental examples.

OCIS codes: (260.5430) Polarization; (120.2130) Ellipsometry and polarimetry; (120.5410) Polarimetry; (050.1950) Diffraction gratings; (160.1190) Anisotropic optical materials

1. Introduction

The ever-growing complexity and increasing variety of materials and structures, either natural or artificial, of both fundamental and applied interest, demands advanced optical characterization methods for understanding and modifying their properties. A medium (material or system) that interacts linearly with probing polarized light is generally described by a 2×2 complex matrix **J**, called Jones matrix, transforming the incident transverse electric field vector into an outgoing one following the interaction. Whereas semi-infinite, isotropic media feature diagonal Jones matrices whose polarization properties are fully described by only two parameters provided by an ellipsometric measurement, called ellipsometric angles, complex media usually exhibit full, non-diagonal Jones matrices that require performing the so-called generalized ellipsometry (GE) for their complete determination [1]. If, furthermore, the medium under characterization is varying in space or in time (within the spatial or temporal resolution of the measurement equipment) then it is not described by a single Jones matrix, but rather by a statistical ensemble of Jones matrices. This statistical ensemble is formally equivalent to a 4×4 real matrix **M**, called Mueller matrix, transforming incident polarized light, in the form of a (four-component real) Stokes vector containing polarized light intensities, into an outgoing one. The corresponding medium, as well as its Mueller matrix, is called depolarizing, since it generally produces partially polarized outgoing light from totally polarized incident one. Conversely, if the medium does not depolarize the incident light, then its Mueller matrix is termed nondepolarizing and is formally reducible to a single Jones matrix.

Ellipsometry (conventional, as well as generalized) is experimentally based on the measurement of polarized light intensities and, because of this, it does not determine, formally speaking, any Jones matrix elements, but rather only Mueller matrix ones [2]. In this sense, ellipsometry appears as partial Mueller polarimetry, since it determines only partially the Mueller matrix, unlike Mueller matrix polarimetry that determines it completely. Any Mueller polarimeter comprises a polarization state generator (PSG) preparing the polarization state, i.e. the Stokes vector, of the probing light and a polarization state analyzer (PSA) analyzing the Stokes vector of the outgoing light after its interaction with the sample. To perform conventional ellipsometry [1,3,4], it is sufficient to have a PSA (PSG) analyzing (generating) a single polarization state and an incomplete PSG (PSA), i.e. a PSG (PSA) that generates (analyzes) just three of the four Stokes vector components [5]. (Note that in some ellipsometer designs the PSG (PSA) is complete, i.e. generates (analyzes) the complete Stokes vector.) The upgrade of conventional ellipsometry to generalized one requires the replacement of the single-polarization-state PSA (PSG) by a complete one while keeping the incomplete PSG (PSA) [5]. A “generalized ellipsometer” measures twelve out of the sixteen Mueller matrix elements from which the complete Jones matrix of the sample is derived (provided there is no depolarization). Finally, in the Mueller polarimeter both the PSG and the PSA are complete and the complete, sixteen-element Mueller matrix is measured.

Therefore, the principal instrumental difference between a “generalized ellipsometer” (actually, a twelve-element partial polarimeter) and a complete Mueller polarimeter is that in a “generalized ellipsometer” either the PSG or the PSA is incomplete, resulting in a measured partial Mueller matrix with either a column or a row missing (i.e. a column or a row whose elements are undetermined). However, as already mentioned, a twelve-element partial Mueller matrix with either a row or a column missing is fully sufficient for the obtainment of the complete Jones matrix of a nondepolarizing sample. Since there is a one-to-one correspondence between a (complete) Jones matrix and a (complete) Mueller one, this means that it is always possible to recover the complete Mueller matrix of a nondepolarizing sample knowing the partial, twelve-element one.

The purpose of the present paper is twofold. First, it provides an explicit procedure, illustrated on an experimental example, on how to recover the complete Mueller matrix from a partial, twelve-element one in the absence of depolarization. Second, it studies the more general case of recovering the complete Mueller matrix if depolarization is present and reports two practically important cases where such recovery is feasible. Like with the first part, analytical procedures and experimental validations are provided for both cases. The benefit of recovering the complete Mueller matrix from a partially known experimental one may be substantial since it makes possible the phenomenological interpretation of the measured medium (material or system) through the application of a number of algebraic decompositions available, independent on whether depolarization is absent [6,7] or not [8-12].

2. Twelve-element partial Mueller polarimetry

As mentioned, twelve-element Mueller polarimetry (yielding a Mueller matrix with a row or a column missing) is performed when either the PSG or the PSA unit of the polarimeter is incomplete (i.e. handles only three components of a Stokes vector), the remaining block being complete (i.e. handling all four components of a Stokes vector). Classic designs, available in single wavelength, spectroscopic or imaging versions, are those of the rotating polarizer and compensator ellipsometer (RPCE) or its dual, the rotating compensator and analyzer ellipsometer (RCAE) [2,13]. (A dual design is the one whereby a missing row is replaced by a missing column or vice versa.) In an alternative, but formally equivalent, design the rotating compensator is replaced by a variable retarder taking discrete azimuth values [14]. There also exist designs free of mechanical rotation whereby the polarization modulation is performed either spatially [15] or spectrally; however, these are typically limited to a narrow spectral window (or to a single wavelength) operation.

A special class of twelve-element Mueller polarimeters is represented by the extended photoelastic modulator ellipsometer (PME) [2,13,16]. It comes in either RPPM (rotating polarizer – photoelastic modulator) or PMRA (photoelastic modulator – rotating analyzer) designs that are dual to one another. Each design measures a nine-element partial Mueller matrix with both a row and a column missing [2,17]. By changing the azimuth setting θ_m of the PM the index of the missing row (or column) changes, so that by combining two nine-element partial matrices obtained at two different PM azimuths one obtains a twelve-element partial Mueller matrix with a single row (or column) missing.

Table 1 lists the various instrument designs and the partial, twelve-element Mueller matrices they are capable of measuring.

Table 1. Instruments and twelve-element partial Mueller matrices they measure.
(Bullets denote missing matrix elements; θ_m denotes the PM azimuth)

Instrument	partial Mueller matrix
RPCE	$\begin{bmatrix} M_{11} & M_{12} & M_{13} & \bullet \\ M_{21} & M_{22} & M_{23} & \bullet \\ M_{31} & M_{32} & M_{33} & \bullet \\ M_{41} & M_{42} & M_{43} & \bullet \end{bmatrix}$
RCAE	$\begin{bmatrix} M_{11} & M_{12} & M_{13} & M_{14} \\ M_{21} & M_{22} & M_{23} & M_{24} \\ M_{31} & M_{32} & M_{33} & M_{34} \\ \bullet & \bullet & \bullet & \bullet \end{bmatrix}$
Extended PME: RPPM	$\theta_m = 0^\circ(90^\circ)$: $\begin{bmatrix} M_{11} & M_{12} & M_{13} & \bullet \\ \bullet & \bullet & \bullet & \bullet \\ M_{31} & M_{32} & M_{33} & \bullet \\ M_{41} & M_{42} & M_{43} & \bullet \end{bmatrix}$ & $\theta_m = \pm 45^\circ$: $\begin{bmatrix} M_{11} & M_{12} & M_{13} & \bullet \\ M_{21} & M_{22} & M_{23} & \bullet \\ \bullet & \bullet & \bullet & \bullet \\ M_{41} & M_{42} & M_{43} & \bullet \end{bmatrix} \Rightarrow \begin{bmatrix} M_{11} & M_{12} & M_{13} & \bullet \\ M_{21} & M_{22} & M_{23} & \bullet \\ M_{31} & M_{32} & M_{33} & \bullet \\ M_{41} & M_{42} & M_{43} & \bullet \end{bmatrix}$
Extended PME: PMRA	$\theta_m = 0^\circ(90^\circ)$: $\begin{bmatrix} M_{11} & \bullet & M_{13} & M_{14} \\ M_{21} & \bullet & M_{23} & M_{24} \\ M_{31} & \bullet & M_{33} & M_{34} \\ \bullet & \bullet & \bullet & \bullet \end{bmatrix}$ & $\theta_m = \pm 45^\circ$: $\begin{bmatrix} M_{11} & M_{12} & \bullet & M_{14} \\ M_{21} & M_{22} & \bullet & M_{24} \\ M_{31} & M_{32} & \bullet & M_{34} \\ \bullet & \bullet & \bullet & \bullet \end{bmatrix} \Rightarrow \begin{bmatrix} M_{11} & M_{12} & M_{13} & M_{14} \\ M_{21} & M_{22} & M_{23} & M_{24} \\ M_{31} & M_{32} & M_{33} & M_{34} \\ \bullet & \bullet & \bullet & \bullet \end{bmatrix}$

Before proceeding, it should be emphasized that our ultimate objective is the recovery of the complete Mueller matrix from a partial, twelve-element one provided by any of the standard partial polarimeter designs listed in Table 1. It contrasts with that of the so-called adaptive polarimetry [18] where the Mueller matrix is either partially [19,20] or completely [21,22] reconstructed by using sets of predefined input polarization states generated by non-standard, adaptive polarimeter designs.

3. Recovery of the complete Mueller matrix from a twelve-element partial one

Mueller polarimetry measures light intensities from which the sixteen second-order conjugate moments $\left\langle J_i^* J_j \right\rangle$ of the four elements J_i of the 2×2 complex Jones matrix **J** of the sample,

$$\mathbf{J} = \begin{bmatrix} J_1 & J_3 \\ J_4 & J_2 \end{bmatrix} \tag{1}$$

are obtained (the brackets ⟨…⟩ denote spatial or time averaging; the asterisk stands for complex conjugation). If we denote by

$$E_i = \left\langle \left| J_i \right|^2 \right\rangle \tag{2a}$$

$$F_{ij} = F_{ji} = \mathrm{Re}\left\langle J_i^* J_j \right\rangle \tag{2b}$$

$$G_{ij} = -G_{ji} = \mathrm{Im}\left\langle J_i^* J_j \right\rangle \tag{2c}$$

the real and imaginary parts of the second-order conjugate moments then these can be arranged into two 4×4 matrices, the real Mueller matrix **M** [1],

$$\mathbf{M} = \begin{bmatrix} \frac{1}{2}(E_1 + E_2 + E_3 + E_4) & \frac{1}{2}(E_1 - E_2 - E_3 + E_4) & F_{13} + F_{42} & -G_{13} - G_{42} \\ \frac{1}{2}(E_1 - E_2 + E_3 - E_4) & \frac{1}{2}(E_1 + E_2 - E_3 - E_4) & F_{13} - F_{42} & -G_{13} + G_{42} \\ F_{14} + F_{32} & F_{14} - F_{32} & F_{12} + F_{34} & -G_{12} + G_{34} \\ G_{14} + G_{32} & G_{14} - G_{32} & G_{12} + G_{34} & F_{12} - F_{34} \end{bmatrix} \tag{3}$$

and the Hermitian (complex conjugate transpose) covariance matrix **H** [23,24],

$$\mathbf{H} = \tfrac{1}{4}\sum_{i,j} M_{ij}\left(\sigma_i \otimes \sigma_j\right) = \tfrac{1}{2}\begin{bmatrix} E_1 & F_{13} - iG_{13} & F_{14} - iG_{14} & F_{12} - iG_{12} \\ F_{13} + iG_{13} & E_3 & F_{34} - iG_{34} & F_{32} - iG_{32} \\ F_{14} + iG_{14} & F_{34} + iG_{34} & E_4 & F_{42} - iG_{42} \\ F_{12} + iG_{12} & F_{32} + iG_{32} & F_{42} + iG_{42} & E_2 \end{bmatrix} \tag{4}$$

where σ_i are the Pauli spin matrices (the symbol "⊗" denotes the Kronecker product). The two matrices **M** and **H** obviously carry the same amount of information; **M** has a clear operational meaning since it transforms input Stokes vectors into outgoing ones and is, therefore (completely or partially) determined by the Mueller polarimeter whereas **H**, being Hermitian and semi-positive definite, features useful algebraic properties. For instance, the rank of **H**, written *rank*(**H**), is an indicator of the activity of the averaging process denoted by the brackets ⟨…⟩. In particular, if *rank*(**H**) = 1 (i.e. **H** has a single non-vanishing eigenvalue) then **H** is a projection matrix and consequently, the second-order conjugate moments of the Jones matrix elements simply equal their respective products, $\left\langle J_i^* J_j \right\rangle = J_i^* J_j$, as follows from Eq. (4) [23,24]. The averaging is therefore inactive and the brackets can be omitted. The

Mueller matrix **M** is then termed nondepolarizing since transforming totally polarized input light into totally polarized outgoing one; furthermore, **M** is fully equivalent to its associated Jones matrix **J**. If $2 \leq rank(\mathbf{H}) \leq 4$ then the averaging over the second-order conjugate moments of the Jones matrix elements is effective. The resulting Mueller matrix **M** is depolarizing (i.e. it generally transforms totally polarized input light into partially polarized outgoing one) and the one-to-one correspondence between **M** and **J** is no more valid: to a single **M** there generally correspond many realizations of **J** over which the averaging takes place (**J** is then called the Jones generator of **M** [25]). Note that the averaging process arises during the polarimetric measurement and is, therefore, dependent on certain parameters (such as spectral and spatial resolution, measurement time) specific to the Mueller polarimeter. Consequently, the resulting depolarizing Mueller matrix not only characterizes the sample itself, but is furthermore affected by the properties of the measurement equipment.

If a partial, twelve-element Mueller matrix is measured, then the four missing (i.e. experimentally undetermined) elements belonging to the last row or column, see Table 1, can be completed (i.e. derived from the known elements) if **M** is nondepolarizing. Indeed, being equivalent to its associated Jones matrix **J**, **M** consequently depends only on seven real parameters, so that there must exist $16 - 7 = 9$ relations between its elements which are, in principle, sufficient to determine the four unknowns. It can be furthermore shown that the solution is unique. Appendix A reports an algebraic procedure, based on the property $rank(\mathbf{H}) = 1$ rather than on the nine relations constraining a nondepolarizing **M**, for completing the last column of a partial Mueller matrix. Note that determining the complete nondepolarizing **M** from its partial counterpart is equivalent to performing generalized ellipsometry (GE) since the Jones matrix **J** underlying **M** can then be uniquely determined by using a well-known procedure [2,26].

Could a similar procedure be devised for a depolarizing partial **M** where $rank(\mathbf{H}) > 1$? If $rank(\mathbf{H}) = 2$ then all four principal 3×3 minors of **H**, as well as its determinant $det(\mathbf{H})$ (i.e. its only 4×4 principal minor) must vanish [23] providing a total of five constraints on the elements of **M**. These are, in principle, sufficient to determine the missing row or column of **M**. However, numerical simulations show that, despite the over-determination, the solution of the algebraic problem is generally not unique. Additional information on the sample and its Mueller matrix, such as the possible presence of symmetry properties, is needed to determine uniquely the missing elements. Finally, if $rank(\mathbf{H}) = 3$ then only $det(\mathbf{H})$ must vanish which is clearly an insufficient constraint for determining the four missing Mueller matrix elements.

To summarize, the missing row or column of a partial, twelve-element Mueller matrix can be successfully completed either if **M** is nondepolarizing or if **M** is depolarizing, but its associated covariance matrix **H** is of rank two and it further obeys certain additional constraints. In what follows we shall assume, without restraining the generality, that the partial **M** has its last column missing. If instead the last row of **M** is missing, then all recovery procedures should be applied to the transposed partial **M**, $\mathbf{M}^T$, and the complete recovered **M** should be re-transposed.

A fundamental question which one faces when one deals with the problem of completing a partial Mueller matrix **M** is how to know the rank of its associated covariance matrix **H** if not all the elements of **M** are known. Indeed, a necessary condition for the recovery of a partial depolarizing **M** is that $rank(\mathbf{H}) = 2$, as already discussed. Furthermore, the recovery approaches in each one of the two cases $rank(\mathbf{H}) = 2$ and $rank(\mathbf{H}) = 1$ are generally different. In either case, the knowledge of $rank(\mathbf{H})$ is a necessary preliminary piece of information that can be only obtained from the optical properties of the sample. Thus, an optically semi-infinite, "bulk" sample consisting of a homogeneous, but not necessarily isotropic, medium is fully described by a unique Jones matrix when measured in reflection [1]. Therefore, such a sample features a nondepolarizing Mueller matrix and consequently, $rank(\mathbf{H}) = 1$. More generally, if the spatial and spectral variations of the sample response occur on scales much larger than, respectively, the coherence area and the spectral resolution of the instrument, then the Mueller matrix **M** of the sample is nondepolarizing (and *rank*(**H**) = 1) . Conversely, the superposition of different, spatially or spectrally unresolved or only partially resolved, contributions from the sample results in a depolarizing **M** (with *rank*(**H**) ≥ 2). In particular, the incoherent or partially coherent addition of the polarimetric responses of two different media (or optical structures) or of two different parts of the same medium (or optical structure) produces *rank*(**H**) = 2. The former case commonly arises in finite spot size measurements in reflection configuration where the spot covers two optically different areas, e.g. an isotropic substrate and an anisotropic diffraction grating ruled in it [27-31]. The latter case is typical of optically thick slabs of transparent homogeneous material (e.g. crystals or optical components, such as retardance waveplates) in which the front side contribution adds incoherently (or partially coherently) to the backside one in either measurement configuration, reflection [32-34] or transmission [34,35]. Thus, by knowing the geometry and the structure of the sample, one can deduce the rank of its covariance matrix, even if its Mueller matrix is unknown.

Once the rank of the covariance matrix **H** is determined (and turns out to equal either one or two) one can proceed with the recovery of the complete Mueller matrix **M** from the partial, twelve-element one. If the rank of **H** is two, then additional information on **M** is needed in order to get a unique solution to the recovery problem. Two practically important cases belonging to this class are discussed below.

3.1 Mueller matrix exhibiting symmetries

By assumption, *rank*(**H**) = 2 and **M** exhibits symmetries, i.e. its off-diagonal elements are either equal or opposite to one another, $M_{ij} = \pm M_{ji}$. The latter relations result directly from the symmetry property $J_4 = \pm J_3$ involving the two off-diagonal elements of the Jones matrix **J** associated with **M**, as easily follows from Eqs. (1-3). Algebraic symmetries originate from physical ones, which in turn follow from the intrinsic symmetry properties of the sample. Thus, it can be shown [17,36] that if the mirror image of the sample with respect to the plane perpendicular to the incidence plane (the plane defined by the incident and the outgoing light beams) and containing the sample normal coincides with the sample itself, then $J_4 = J_3$, i.e. the Jones matrix of the sample is symmetric. The corresponding Mueller matrix is of the form

$$\begin{bmatrix} M_{11} & M_{12} & M_{13} & M_{14} \\ M_{12} & M_{22} & M_{23} & M_{24} \\ M_{13} & M_{23} & M_{33} & M_{34} \\ -M_{14} & -M_{24} & -M_{34} & M_{44} \end{bmatrix} \tag{5}$$

The second symmetry case whereby the Jones matrix is antisymmetric, $J_4 = -J_3$, occurs whenever a 180°-rotation of the sample about its normal brings the sample into itself [17,36]. The corresponding Mueller matrix is

$$\begin{bmatrix} M_{11} & M_{12} & M_{13} & M_{14} \\ M_{12} & M_{22} & M_{23} & M_{24} \\ -M_{13} & -M_{23} & M_{33} & M_{34} \\ M_{14} & M_{24} & -M_{34} & M_{44} \end{bmatrix} \tag{6}$$

Finally, a special symmetry case occurs when $J_4 = J_3 = 0$, i.e. the Jones matrix is diagonal. The previous two symmetry properties, $J_4 = J_3$ and $J_4 = -J_3$, are then simultaneously satisfied by the sample and can be combined into a single one stating that the sample is mirror-symmetric with respect to the incidence plane [17,36]. This is the most common case of an isotropic medium whose Mueller matrix is of the special block-diagonal form

$$\mathbf{M}_{bd} = \begin{bmatrix} M_{11} & M_{12} & 0 & 0 \\ M_{12} & M_{11} & 0 & 0 \\ 0 & 0 & M_{33} & M_{34} \\ 0 & 0 & -M_{34} & M_{33} \end{bmatrix} \quad (7)$$

It is easy to see that the presence of symmetries reduces the rank of the covariance matrix **H**. Thus, if $J_4 = \pm J_3$ then it follows from Eqs. (2) and (4) that the second and the third row and column of **H** are either the same or are opposite. Since this is a trivial case of linear dependence between two rows (or columns), then *rank*(**H**) ≤ 4 – 1 = 3. In the special case where $J_4 = J_3 = 0$ both the second and the third row and column of the covariance matrix are identically zero, so that *rank*(**H**) ≤ 4 – 2 = 2. Thus, the covariance matrices associated with the Mueller matrices from Eqs. (5) and (6) are of rank three at most, whereas the one associated with the special block-diagonal Mueller matrix from Eq. (7) has a rank that cannot exceed two. These observations are fully compatible with the initial assumption that *rank*(**H**) = 2.

The presence of symmetries likewise reduces significantly the number of unknowns in a twelve-element partial Mueller matrix with a missing last column (or row), from a total of four to just a single one. Indeed, it follows directly from Eqs. (5) and (6) that $M_{14} = \mp M_{41}$, $M_{24} = \mp M_{42}$ and $M_{34} = -M_{43}$ for the first three elements of the last column of **M** leaving unknown only the M_{44} element. This reduction of the number of unknowns furthermore ensures the unicity of the solution for the single unknown left. In practice, if one knows that the sample obeys symmetries leading to Eq. (5) or (6) and that, moreover, *rank*(**H**) = 2, one should “fill in” the first three missing elements of the last column of the experimental partial **M** and find the last one, M_{44}, by using the algebraic constraints resulting from the constraint *rank*(**H**) = 2. An algebraic procedure for determining M_{44} under the above conditions is given in Appendix B.

The situation becomes trivial if the twelve-element partial **M** is known to be of the special block-diagonal form $\mathbf{M}_{bd}$ given by Eq. (7) because the sample is mirror-symmetric with respect to the incidence plane. As we have already seen, then *rank*($\mathbf{H}_{bd}$) ≤ 2 and so, the missing last column can be uniquely determined. Indeed, Eq. (7) shows that the unique solution to the problem is $M_{14} = M_{24} = 0$, $M_{34} = -M_{43}$ and $M_{44} = M_{33}$, without any algebraic procedure. One may further deduce $\mathbf{H}_{bd}$ from the completed $\mathbf{M}_{bd}$ and check whether *rank*($\mathbf{H}_{bd}$) = 1 or 2, i.e. whether $\mathbf{M}_{bd}$ is nondepolarizing or not. In practice, the last case is the one (however, not the only one) of an optically thick slab made of isotropic material.

One may wonder incidentally if the only missing M_{44} element could not be determined under the more general assumption that *rank*(**H**) ≤ 3 instead of the current, more restrictive one, that *rank*(**H**) = 2. Indeed, *rank*(**H**) ≤ 3 means that *det*(**H**) = 0 which potentially yields a fourth-order algebraic equation for the single unknown. However, as we have already seen, one gets *rank*(**H**) ≤ 3 for the covariance matrix associated with a Mueller matrix obeying symmetries and therefore, the relation *det*(**H**) = 0 is actually not an equation, but rather an identity.

3.2 Mueller matrix one of whose two matrix components is of special block-diagonal form

A Mueller matrix **M** such that *rank*(**H**) = 2 where **H** is its associated covariance matrix can be decomposed into the sum of two nondepolarizing Mueller matrices $\mathbf{M}_1$ and $\mathbf{M}_2$ (i.e. such that *rank*($\mathbf{H}_1$) = *rank*($\mathbf{H}_2$) = 1) in an infinite number of ways. However, if $\mathbf{M}_2$ is of the special block-diagonal form $\mathbf{M}_{bd}$ given by Eq. (7) then it can be shown [29] that the decomposition $\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_{bd}$ is unique, provided $\mathbf{M}_1$ is not also of this special form. Although seemingly artificial, this is a relatively common situation in finite spot size Mueller polarimetry where the probing light simultaneously falls upon an anisotropic medium described by $\mathbf{M}_1$ and an isotropic one, given by $\mathbf{M}_{bd}$. For instance, this is the case with an (anisotropic) diffraction grating whose lateral dimensions are smaller than the spot size, ruled in an (isotropic) substrate. There exist both algebraic [29,30] and numerical [31] robust practical procedures for recovering the two components $\mathbf{M}_1$ and $\mathbf{M}_{bd}$ given the experimental **M**.

It turns also out that the practically important case $\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_{bd}$ is not only "separable", but is also "completable", i.e. it is possible to complete a twelve-element partial **M** with a missing last column (row) to a complete one. Appendix C describes an algebraic procedure solving this problem. Once **M** is completed, it can be subsequently resolved into its two matrix components by the known methods [29-31].

If, furthermore, the sample is known to exhibit one of the first two kinds of symmetries discussed in the previous subsection then one can partially complete the missing last column by using the appropriate set of relations $M_{14} = \mp M_{41}$, $M_{24} = \mp M_{42}$ and $M_{34} = -M_{43}$. (Notice that the kind of symmetry obeyed by **M** and by its component $\mathbf{M}_1$ are the same, since the special block-diagonal component $\mathbf{M}_{bd}$ obeys both kinds; therefore, the symmetry of **M** is fully determined by that of its first component $\mathbf{M}_1$.) The only undetermined element left, M_{44}, can then be found by using the procedure from Appendix B. Clearly, the symmetry-based approach disregards the preliminary knowledge on the structure of **M** as being decomposable into $\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_{bd}$, whereas the one from Appendix C takes it into account but does not take any advantage of the presence of symmetries. In practice, one may apply both approaches and select the best of the two either by resorting to continuity considerations on the recovered elements (in spectroscopic or in imaging polarimetry) or, alternatively, by checking the rank of the recovered **H**; since *rank*(**H**) = 2, two of its eigenvalues should vanish to the experimental accuracy.

4. Experimental validation

4.1 Experimental details

The measurement equipment, described in detail in Ref. 37, is a home-made UV-visible spectroscopic Mueller polarimeter based on four photoelastic modulators. The complete Mueller matrices of the validation samples, a cleaved mica sheet (thickness ~0.4 mm) and a symmetric profile diffraction grating with a 500-nm period ruled in a silicon substrate (see Ref. 28 for more detail), were measured in reflection configuration over the 240-nm – 650-nm spectral range at the respective incidence angles of 75° and 65°.

4.2 Completion of a twelve-element partial nondepolarizing Mueller matrix

Figure 1 presents the complete Mueller matrix (solid line) of the diffraction grating sample. The azimuth of the grating was set at 45° with the respect to the incidence plane, ensuring anisotropic response with non-zero off-diagonal-block Mueller matrix elements. Being spatially homogeneous and optically thick (i.e. there is no signal contribution from its backside), the grating sample is nondepolarizing, i.e. the rank of its covariance matrix **H** is one. This makes possible the recovery of its complete Mueller matrix **M** from a twelve-element partial one with the last column missing. Assuming this is the case, we have recovered the fourth column of **M** by applying the algebraic procedure from Appendix A. The result of the analytical recovery is reported in Fig. 1 (red crosses) where, for comparison, a numerical recovery is also presented (green circles). The latter was obtained by simultaneously minimizing the six 2×2 principal minors of the covariance matrix to get the four missing Mueller matrix elements, implemented as fitting parameters in the algorithm. The agreement between the measured last column elements and the recovered ones is excellent for both algebraic and numerical approaches. The rms deviations for both approaches are below one percent, to be compared to the experimental error evaluated at about 0.5 percent [37]. In practice, one can use either approach provided the experimental data are not too noisy. Indeed, being based on a minimization principle, the numerical approach can be expected to be more robust to noise, whereas the algebraic one is computationally much faster, since based on an explicit analytical solution of the problem.

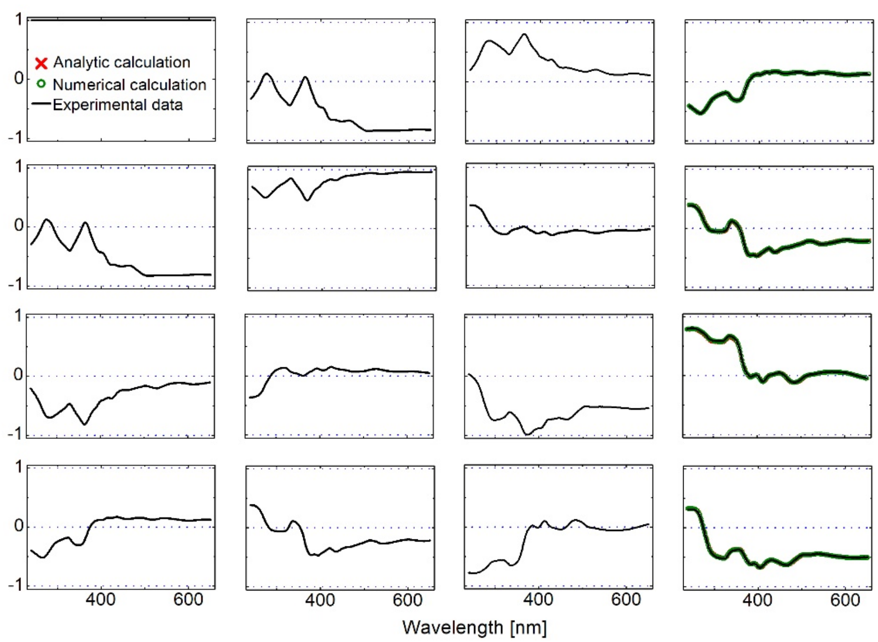

Fig. 1. Complete Mueller matrix of the diffraction grating (solid line) and the recovery of its last column from the twelve-element partial one by using the algebraic procedure from Appendix A (red crosses) and the numerical approach (green circles).

4.3 Completion of a twelve-element partial Mueller matrix obeying symmetries

Figure 2 reproduces the complete spectroscopic reflection Mueller matrix (solid line) of the mica sheet sample. Notice that the large M_{12} and M_{21} elements indicate the presence of significant diattenuation superimposing on the expected retardance; this is physically due to the Fresnel reflection at an incidence angle rather close to the Brewster angle of the material.

Also, the interference between ordinary and extraordinary beams produces spectral oscillations, as readily seen from the lower 2×2 diagonal block.

Optically, the mica sheet represents a thick (with respect to the coherence length of the probing light) parallel slab of uniaxially anisotropic material with in-plane optic axis. Because of the important thickness of the slab, front- and backside reflected partial beams add only partially coherently upon forming the outgoing beam. The rank of the covariance matrix associated with the experimental Mueller matrix is two, since two, front and back, contributions superimpose with loss of coherence. At a non-trivial azimuth value of the optic axis with respect to the incidence plane (i.e. different from 0° or 90°), the Mueller matrix of the anisotropic slab is full, i.e. its two 2×2 off-diagonal blocks are non-zero. Furthermore, the response of the slab being invariant with respect to a 180°-rotation about its normal (because of the in-plane orientation of the optic axis of the uniaxial material), its Mueller matrix is of the form given by Eq. (6), i.e. it obeys specific symmetries.

The two conditions for the recovery of the last column of the Mueller matrix, assumed to be missing, are therefore, met. First, one "fills in" the upper three missing elements of the fourth column by using the relations $M_{14} = M_{41}$, $M_{24} = M_{42}$ and $M_{34} = -M_{43}$ directly following from Eq. (6). Next, one applies the procedure from Appendix B to find the only missing element left, M_{44}. The result of the recovery is shown in Fig. 2 (red crosses). The numerical recovery of the M_{44} element, based on the simultaneous minimization of the four 3×3 principal minors of the covariance matrix, is likewise shown (green circles). The two, algebraic and numerical, approaches produce M_{44} element values virtually coinciding with the effectively measured one within the experimental accuracy (except for three isolated wavelengths in the algebraically recovered spectrum that can be readily interpolated on a continuity basis). Like in subsection 4.2, the practical performance of the two approaches is similar and one should choose the best suited one depending on the data noise level or execution time requirements.

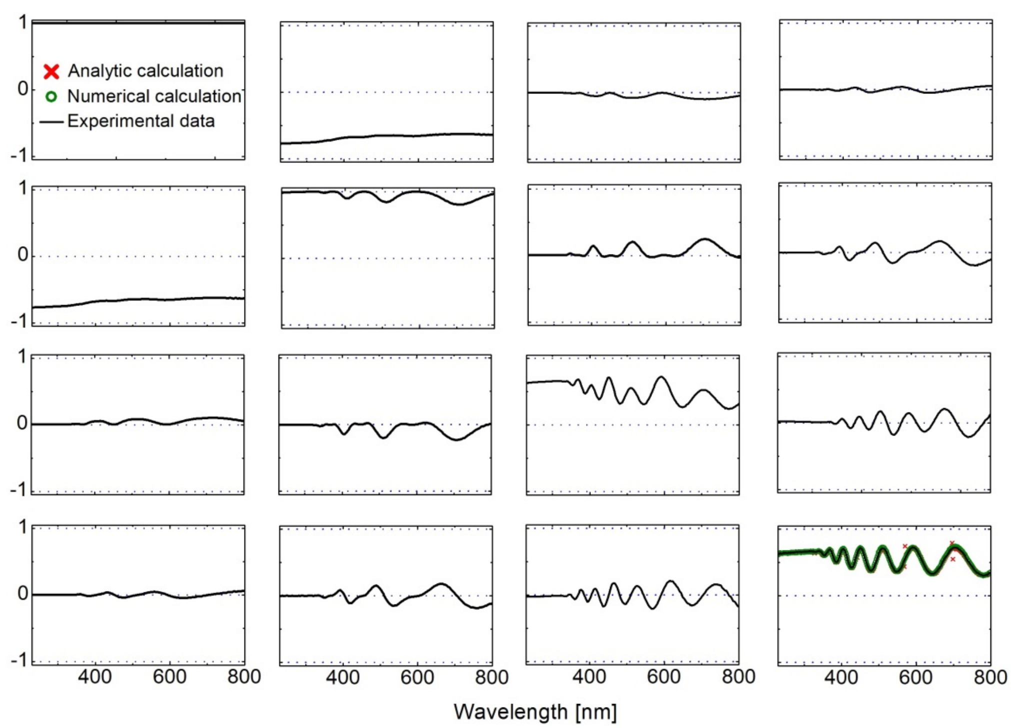

Fig. 2. Complete Mueller matrix of the mica slab (solid line) and the recovery of its M_{44} element from the twelve-element partial one by using the algebraic procedure from Appendix B (red crosses) and the numerical approach (green circles). The elements M_{14}, M_{24} and M_{34} have been recovered by exploiting the symmetries of the sample.

4.4 Completion of a twelve-element partial Mueller matrix one of whose two matrix components is of special block-diagonal form

Figure 3 shows the complete spectroscopic Mueller matrix (solid line) of the diffraction grating sample, with the probing light spot impinging not only on the grating itself but also partially on substrate surrounding it. As discussed in subsection 3.2, the experimental Mueller matrix is the weighted sum of the Mueller matrix of the grating shown in Fig. 1 (see subsection 4.2) and the special block-diagonal Mueller matrix from Eq. (7) of the isotropic substrate, and $rank(\mathbf{H}) = 2$.

In this specific case the algebraic procedure from Appendix C for the recovery of the last column of the Mueller matrix, assumed to be missing, is applicable. The recovered values of the four last column elements are shown in Fig. 2 in red crosses. For comparison, the numerically obtained ones (green circles) are also reported. The numerical procedure used was similar to that from previous subsection, with the only essential difference being that the number of fitting parameters was four instead of just one. In general, the agreement between the effectively measured last column and the recovered one, be it by using the algebraic or the numerical procedure, is very good. At certain spectral points there is, however, a noticeable disagreement for the algebraic recovery, as seen in the M_{14}-element spectrum, for instance. The outlier points are essentially caused by divisions by small numbers during the analytic calculations that amplify the measurement noise. This problem is alleviated by the numerical minimization procedure. Indeed, the numerical approach is based on the minimization of certain vanishing minors, evaluated from the experimentally determined matrix elements, whereas the algebraic one assumes these minors to be strictly zero in order to deduce the missing matrix elements and is consequently, more sensitive to the experimental noise. Notice also that the outliers from the algebraic procedure can be readily interpolated from their neighbors by exploiting the continuity of the spectra.

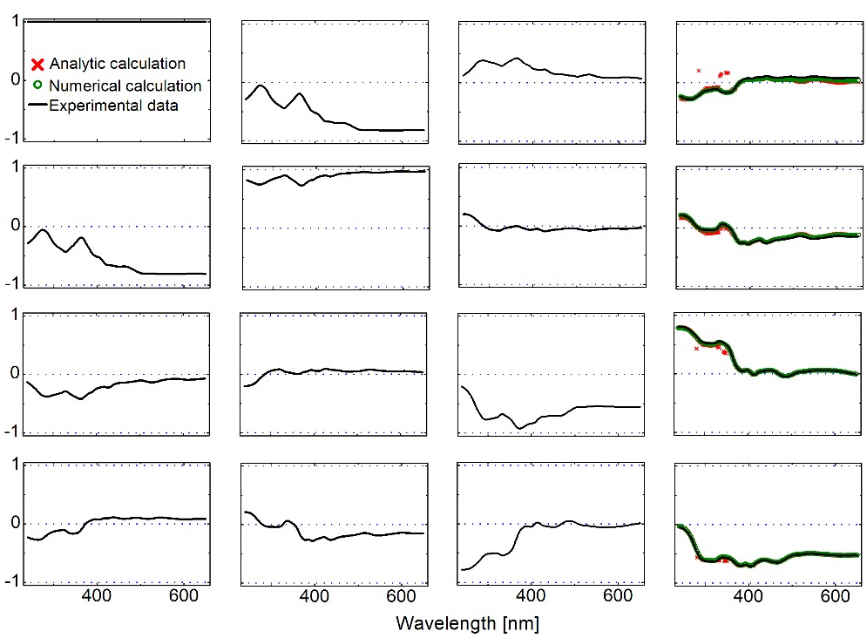

Fig. 3. Complete Mueller matrix of the grating-substrate mixture (solid line) and the recovery of its last column from the twelve-element partial one by using the algebraic procedure from Appendix C (red crosses) and the numerical approach (green circles).

Because of its structure, the diffraction grating qualitatively behaves like a uniaxially anisotropic medium whose optic axis is directed along the grating direction. Therefore, its polarimetric response is invariant with respect to 180°-rotation about the sample normal and its Mueller matrix exhibits the same symmetries as the mica sheet one from the previous subsection, see Fig. 2, i.e. both Mueller matrices have the form of Eq. (6). As already discussed in subsection 3.2, the addition of the contribution of the special-block diagonal matrix $\mathbf{M}_{bd}$ from (7), physically due to the isotropic substrate, into the overall polarimetric response does not break the symmetries: the global symmetry is determined from the lowest symmetry present, given by Eq. (6). Consequently, one is in a position to apply the algebraic procedure from Appendix B for the only missing M_{44} element, after having "filled in" the other three from the symmetry relations $M_{14} = M_{41}$, $M_{24} = M_{42}$ and $M_{34} = -M_{43}$. The result (red crosses) is reported in Fig. 4. One observes a recovery of excellent quality, practically coinciding with both the experimental values, as well as with the numerical recovery (green circles), reproduced for comparison from Fig. 3. Therefore, in the special case where the sample is decomposable into the sum of two matrix components one of which is of special block-diagonal form while the other exhibits symmetries, one may apply indifferently the procedures from Appendices B and C.

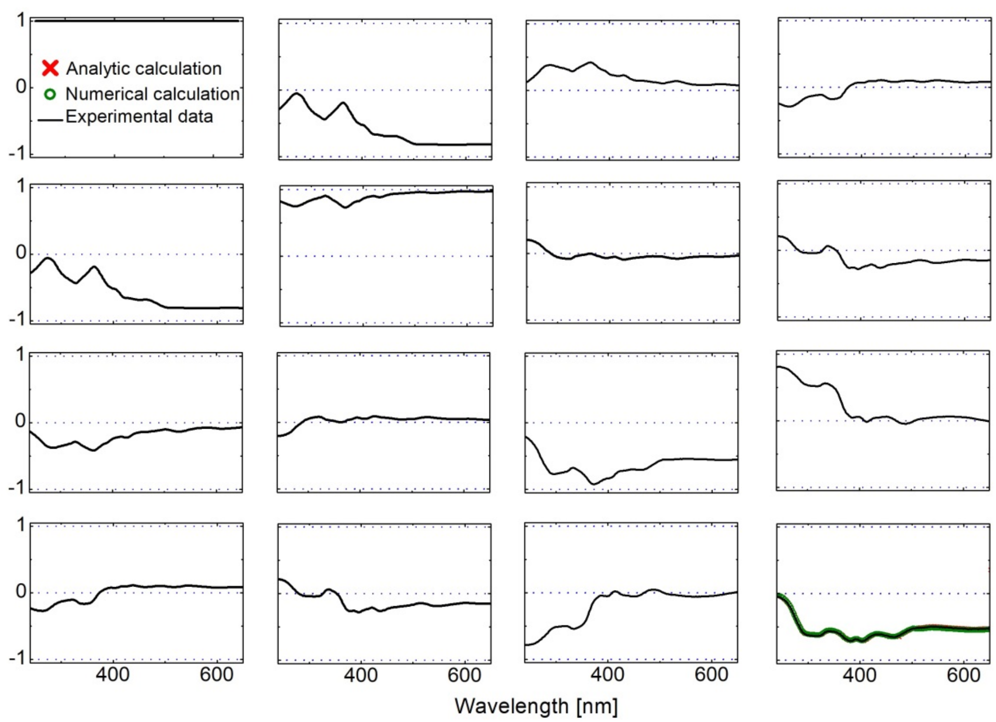

Fig. 4. Complete Mueller matrix of the grating-substrate mixture (solid line) and the recovery of its M_{44} element from the twelve-element partial one by using the algebraic procedure from Appendix B (red crosses) and the numerical approach (green circles). The elements M_{14}, M_{24} and M_{34} have been recovered by exploiting the symmetries of the sample.

5. Summary

We have shown that the partial, twelve element Mueller matrix with the last row or column missing obtained in a generalized ellipsometry experiment can be completed to a full one under certain conditions. In particular, this is always possible if the sample is nondepolarizing. If depolarization is present, the recovery is still possible in the practically important case where the rank of the covariance matrix associated with the Mueller matrix equals two. To obtain a unique solution, one further needs to either employ symmetry considerations, if present, or to assume a contribution of special block-diagonal form, most often due to an isotropic medium, in the overall response. We have reported both algebraic and numerical recovery procedures and have demonstrated their performance on experimental data in all three of the above cases. We believe these results to be interest to experimentalists performing generalized ellipsometry experiments on both nondepolarizing and depolarizing samples and willing to recover the complete Mueller matrices of the latter from the partial, twelve-element ones.

Appendix A: Algebraic procedure for completing the last column of a partial nondepolarizing Mueller matrix

The algebraic problem is that of determining the four unknown elements of the last column of the Mueller matrix **M** given its remaining twelve elements and using the fact that **M** is nondepolarizing. Inspection of Eq. (3) for **M** and of Eq. (4) for its covariance matrix **H** shows that the unknown column elements can be related to those of **H** through the real and imaginary parts of the Jones matrix second-order conjugate moments, G_{13}, G_{42}, G_{12}, G_{34}, F_{12} and F_{34},

$$M_{14} = -G_{13} - G_{42} = 2\,\mathrm{Im}(H_{12} + H_{34}) \qquad (\mathrm{A1}a)$$

$$M_{24} = -G_{13} + G_{42} = 2\,\mathrm{Im}(H_{12} - H_{34}) \qquad (\mathrm{A1}b)$$

$$M_{34} = -G_{12} + G_{34} = 2\,\mathrm{Im}(H_{14} - H_{23}) \qquad (\mathrm{A1}c)$$

$$M_{44} = F_{12} - F_{34} = 2\,\mathrm{Re}(H_{14} - H_{23}) \qquad (\mathrm{A1}d)$$

Clearly, one has to determine the four covariance matrix elements H_{12}, H_{34}, H_{14} and H_{23}. The last two, H_{14} and H_{23}, are not independent, but are rather interrelated through the condition,

$$\begin{aligned} H_{14} + H_{23} &= \tfrac{1}{2}(F_{12} - iG_{12}) + \tfrac{1}{2}(F_{34} - iG_{34}) \\ &= \tfrac{1}{2}(F_{12} + F_{34}) - i\tfrac{1}{2}(G_{12} + iG_{34}) = \tfrac{1}{2}(M_{33} - iM_{43}) \end{aligned} \qquad (\mathrm{A2})$$

All 2×2 minors of **H** being zero (since **H** is of rank one which is equivalent to **M** being nondepolarizing), one can write, in particular, for the two principal minors $H^{(34)}_{(34)}$ and $H^{(12)}_{(12)}$,

$$\begin{aligned} H^{(34)}_{(34)} = \begin{vmatrix} H_{11} & H_{12} \\ H_{21} & H_{22} \end{vmatrix} &= H_{11}H_{22} - H_{12}H_{21} = H_{11}H_{22} - |H_{12}|^2 \\ &= H_{11}H_{22} - (\mathrm{Re}\,H_{12})^2 - (\mathrm{Im}\,H_{12})^2 = 0 \end{aligned} \qquad (\mathrm{A3}a)$$

and

$$H_{(12)}^{(12)} = \begin{vmatrix} H_{33} & H_{34} \\ H_{43} & H_{44} \end{vmatrix} = H_{33}H_{44} - H_{34}H_{43} = H_{33}H_{44} - |H_{34}|^2$$
$$= H_{33}H_{44} - (\mathrm{Re}\, H_{34})^2 - (\mathrm{Im}\, H_{34})^2 = 0 \tag{A3b}$$

where the notation $H_{(ij)}^{(kl)}$ designates the 2×2 minor obtained by striking out the ith and jth rows, together with the kth and lth columns of $\mathbf{H}$. Since both

$$\mathrm{Re}\, H_{12} = \tfrac{1}{2} F_{13} = \tfrac{1}{4}(M_{13} + M_{23}) \tag{A4a}$$

and

$$\mathrm{Re}\, H_{34} = \tfrac{1}{2} F_{42} = \tfrac{1}{4}(M_{13} - M_{23}) \tag{A4b}$$

are known, one can readily evaluate the absolute values, $|\mathrm{Im}\, H_{12}|$ and $|\mathrm{Im}\, H_{34}|$, of $\mathrm{Im}\, H_{12}$ and $\mathrm{Im}\, H_{34}$ from Eqs. (A3). To determine the signs of $\mathrm{Im}\, H_{12}$ and $\mathrm{Im}\, H_{34}$, use can be made of the vanishing minor $H_{(23)}^{(14)}$,

$$H_{(23)}^{(14)} = \begin{vmatrix} H_{12} & H_{13} \\ H_{42} & H_{43} \end{vmatrix} = H_{12}H_{43} - H_{13}H_{42} = H_{12}H_{34}^{*} - H_{13}H_{24}^{*} = 0 \tag{A5}$$

(the asterisk denotes complex conjugation) in which both

$$H_{13} = \tfrac{1}{2}(F_{14} - iG_{14}) = \tfrac{1}{4}[M_{31} + M_{32} - i(M_{41} + M_{42})] \tag{A6a}$$

and

$$H_{24} = \tfrac{1}{2}(F_{32} - iG_{32}) = \tfrac{1}{4}[M_{31} - M_{32} - i(M_{41} - M_{42})] \tag{A6b}$$

are known. The signs of $\mathrm{Im}\, H_{12}$ and $\mathrm{Im}\, H_{34}$ are chosen in such a way that $H_{12} = \mathrm{Re}\, H_{12} + i\, \mathrm{Im}\, H_{12}$ and $H_{34} = \mathrm{Re}\, H_{34} + i\, \mathrm{Im}\, H_{34}$ satisfy simultaneously both the real and the imaginary parts of Eq. (A5); a total of four sign combinations has to be checked. The two elements H_{12} and H_{34} are thus fully determined.

To determine the remaining pair H_{14} and H_{23}, use is made of the vanishing minors $H_{(13)}^{(12)}$ and $H_{(34)}^{(24)}$,

$$H_{(13)}^{(12)} = \begin{vmatrix} H_{23} & H_{24} \\ H_{43} & H_{44} \end{vmatrix} = H_{23}H_{44} - H_{24}H_{43} = H_{23}H_{44} - H_{24}H_{34}^{*} = 0 \tag{A7a}$$

and

$$H_{(34)}^{(24)} = \begin{vmatrix} H_{11} & H_{13} \\ H_{21} & H_{23} \end{vmatrix} = H_{11}H_{23} - H_{13}H_{21} = H_{11}H_{23} - H_{13}H_{12}^{*} = 0 \qquad \text{(A7}b\text{)}$$

from which one gets the unknown H_{23},

$$H_{23} = \frac{H_{24}H_{34}^{*}}{H_{44}} = \frac{H_{13}H_{12}^{*}}{H_{11}} = \frac{H_{24}H_{34}^{*} + H_{13}H_{12}^{*}}{H_{44} + H_{11}} \qquad \text{(A8)}$$

after having determined the two diagonal elements H_{44} and H_{11} from

$$H_{44} = \tfrac{1}{2}E_2 = \tfrac{1}{4}\left(M_{11} + M_{22} - M_{12} - M_{21}\right) \qquad \text{(A9}a\text{)}$$

and

$$H_{11} = \tfrac{1}{2}E_1 = \tfrac{1}{4}\left(M_{11} + M_{22} + M_{12} + M_{21}\right) \qquad \text{(A9}b\text{)}$$

Knowing H_{23}, the only remaining unknown H_{14} is determined from Eq. (A2) and finally, the missing fourth column elements of **M** are obtained from Eqs. (A1). The above algebraic procedure is not the only possible one, but it demonstrates excellent noise resilience on experimental data.

Appendix B: Algebraic procedure for completing the M_{44} element of a partial Mueller matrix having a rank-two associated covariance matrix

The only unknown element M_{44} of **M** is expressed as

$$M_{44} = F_{12} - F_{34} = 2\left(\operatorname{Re} H_{14} - \operatorname{Re} H_{23}\right) \qquad \text{(B1)}$$

in terms of the elements of the covariance matrix **H**, as follows by direct identification from Eqs. (3) and (4). (Note that Eq. (B1) is identical to Eq. (A1*d*) from Appendix A.) One therefore needs to find the real parts, $\operatorname{Re} H_{14}$ and $\operatorname{Re} H_{23}$, of the two elements H_{14} and H_{23}. The two unknowns $\operatorname{Re} H_{14}$ and $\operatorname{Re} H_{23}$ are not independent since their sum is

$$\operatorname{Re} H_{14} + \operatorname{Re} H_{23} = \tfrac{1}{2}\left(F_{12} + F_{34}\right) = \tfrac{1}{2}M_{33} \qquad \text{(B2)}$$

The covariance matrix **H** being of rank two by assumption, all its 3×3 minors must vanish. Thus, if one denotes by $H^{(i)}$ the 3×3 principal minor obtained by striking out the *i*th row and column of **H**, one can write

$$H^{(1)} = \begin{vmatrix} H_{22} & H_{23} & H_{24} \\ H_{32} & H_{33} & H_{34} \\ H_{42} & H_{43} & H_{44} \end{vmatrix} = H_{22}H_{33}H_{44} + H_{24}^*H_{34}H_{23} + H_{24}H_{34}^*H_{23}^* - H_{22}|H_{34}|^2 - H_{33}|H_{24}|^2 - H_{44}|H_{23}|^2 = 0 \quad \text{(B3}a\text{)}$$

and

$$H^{(4)} = \begin{vmatrix} H_{11} & H_{12} & H_{13} \\ H_{21} & H_{22} & H_{23} \\ H_{31} & H_{32} & H_{33} \end{vmatrix} = H_{11}H_{22}H_{33} + H_{13}^*H_{12}H_{23} + H_{13}H_{12}^*H_{23}^* - H_{33}|H_{12}|^2 - H_{22}|H_{13}|^2 - H_{11}|H_{23}|^2 = 0 \quad \text{(B3}b\text{)}$$

so that

$$H_{11}H^{(1)} - H_{44}H^{(4)} = \left(H_{11}H_{24}^*H_{34} - H_{44}H_{13}^*H_{12}\right)H_{23} + \left(H_{11}H_{24}H_{34}^* - H_{44}H_{13}H_{12}^*\right)H_{23}^* - H_{11}\left(H_{22}|H_{34}|^2 + H_{33}|H_{24}|^2\right) + H_{44}\left(H_{11}|H_{23}|^2 + H_{22}|H_{13}|^2\right) = 0 \quad \text{(B4)}$$

From Eq. (B4) one gets for $\operatorname{Re} H_{23}$,

$$\operatorname{Re} H_{23} = \frac{A + 2\operatorname{Im}\left(H_{11}H_{24}^*H_{34} - H_{44}H_{13}^*H_{12}\right)\operatorname{Im} H_{23}}{2\operatorname{Re}\left(H_{11}H_{24}^*H_{34} - H_{44}H_{13}^*H_{12}\right)} \quad \text{(B5}a\text{)}$$

where

$$A = H_{11}\left(H_{22}|H_{34}|^2 + H_{33}|H_{24}|^2\right) - H_{44}\left(H_{11}|H_{23}|^2 + H_{22}|H_{13}|^2\right) \quad \text{(B5}b\text{)}$$

All elements of **H** appearing in Eqs. (B5) are known. Indeed, it follows from Eqs. (4) and (3) that

$$H_{11} = \tfrac{1}{2}E_1 = \tfrac{1}{4}\left(M_{11} + M_{22} + M_{12} + M_{21}\right) \quad \text{(B6}a\text{)}$$

$$H_{22} = \tfrac{1}{2}E_3 = \tfrac{1}{4}\left(M_{11} - M_{22} - M_{12} + M_{21}\right) \quad \text{(B6}b\text{)}$$

$$H_{33} = \tfrac{1}{2}E_4 = \tfrac{1}{4}\left(M_{11} - M_{22} + M_{12} - M_{21}\right) \quad \text{(B6}c\text{)}$$

$$H_{44} = \tfrac{1}{2}E_2 = \tfrac{1}{4}\left(M_{11} + M_{22} - M_{12} - M_{21}\right) \quad \text{(B6}d\text{)}$$

$$H_{13} = \tfrac{1}{2}\left(F_{14} - iG_{14}\right) = \tfrac{1}{4}\left[M_{31} + M_{32} - i\left(M_{41} + M_{42}\right)\right] \quad \text{(B6}e\text{)}$$

$$H_{24} = \tfrac{1}{2}(F_{32} - iG_{32}) = \tfrac{1}{4}[M_{31} - M_{32} - i(M_{41} - M_{42})] \tag{B6f}$$

$$H_{12} = \tfrac{1}{2}(F_{13} - iG_{13}) = \tfrac{1}{4}[M_{13} + M_{23} + i(M_{14} + M_{24})] \tag{B6g}$$

$$H_{34} = \tfrac{1}{2}(F_{42} - iG_{42}) = \tfrac{1}{4}[M_{13} - M_{23} + i(M_{14} - M_{24})] \tag{B6h}$$

and

$$\operatorname{Im} H_{23} = -\tfrac{1}{2} G_{34} = -\tfrac{1}{4}(M_{34} + M_{43}) \tag{B7}$$

Having obtained $\operatorname{Re} H_{23}$ from Eq. (B5*a*) one determines $\operatorname{Re} H_{14}$ with the help of Eq. (B2). An alternative way of finding $\operatorname{Re} H_{23}$ and $\operatorname{Re} H_{14}$ consists in solving the sum of the two quadratic equations (B3),

$$\begin{aligned} H^{(1)} + H^{(4)} = {} & H_{22}H_{33}(H_{11} + H_{44}) + \left(H_{13}^{*}H_{12} + H_{24}^{*}H_{34}\right)H_{23} + \left(H_{13}H_{12}^{*} + H_{24}H_{34}^{*}\right)H_{23}^{*} \\ & - H_{33}\left(\left|H_{12}\right|^2 + \left|H_{24}\right|^2\right) - H_{22}\left(\left|H_{13}\right|^2 + \left|H_{34}\right|^2\right) - (H_{11} + H_{44})\left|H_{23}\right|^2 = 0 \end{aligned} \tag{B8a}$$

providing two solutions for $\operatorname{Re} H_{23}$, together with

$$\begin{aligned} H^{(2)} + H^{(3)} = {} & H_{11}H_{44}(H_{22} + H_{33}) + \left(H_{13}^{*}H_{34}^{*} + H_{12}^{*}H_{24}^{*}\right)H_{14} + \left(H_{13}H_{34} + H_{12}H_{24}\right)H_{14}^{*} \\ & - H_{11}\left(\left|H_{34}\right|^2 + \left|H_{24}\right|^2\right) - H_{44}\left(\left|H_{12}\right|^2 + \left|H_{13}\right|^2\right) - (H_{22} + H_{33})\left|H_{14}\right|^2 = 0 \end{aligned} \tag{B8b}$$

yielding two possible values for $\operatorname{Re} H_{14}$. To solve Eq. (B8*b*) for $\operatorname{Re} H_{14}$ one needs first to determine $\operatorname{Im} H_{14}$ from

$$\operatorname{Im} H_{14} = -\tfrac{1}{2} G_{12} = \tfrac{1}{4}(M_{34} - M_{43}) \tag{B9}$$

The correct pair of solutions $(\operatorname{Re} H_{23}, \operatorname{Re} H_{14})$ is identified by matching the constraint (B2) on the sum $\operatorname{Re} H_{23} + \operatorname{Re} H_{14}$; four cases have to be checked. Although less direct, the second approach may turn out to be less prone to noise when applied on experimental data.

Finally, once $\operatorname{Re} H_{23}$ and $\operatorname{Re} H_{14}$ determined either way, the missing M_{44} element is obtained from Eq. (B1).

Appendix C: Algebraic procedure for completing the last column of a partial Mueller matrix decomposable into the sum of two nondepolarizing Mueller matrices one of which has a special block-diagonal form

Like in Appendix A, the elements of the missing fourth column of **M** are expressible through Eqs. (A1) in terms of the four covariance matrix elements H_{12}, H_{34}, H_{14} and H_{23}. The sum $H_{14} + H_{23}$ of the last two is likewise constrained by Eq. (A2). Since by assumption

$\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_{bd}$ where $\mathbf{M}_1$ is full (i.e. all of its elements are non-zero) whereas $\mathbf{M}_{bd}$ is of the special block-diagonal form given by Eq. (7), the covariance matrix **H** associated with **M** also decomposes into the sum,

$$\mathbf{H} = \mathbf{H}_1 + \mathbf{H}_{bd} = \begin{bmatrix} H'_{11} & H_{12} & H_{13} & H'_{14} \\ H_{21} & H_{22} & H_{23} & H_{24} \\ H_{31} & H_{32} & H_{33} & H_{34} \\ H'_{41} & H_{42} & H_{43} & H'_{44} \end{bmatrix} + \begin{bmatrix} H''_{11} & 0 & 0 & H''_{14} \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ H''_{41} & 0 & 0 & H''_{44} \end{bmatrix} \tag{C1}$$

in which each one of the two matrix summands is of rank one (since both $\mathbf{M}_1$ and $\mathbf{M}_{bd}$ are nondepolarizing). Consequently, all 2×2 minors $H_{1(ij)}^{(kl)}$ of $\mathbf{H}_1$, obtained by striking out the ith and jth rows, together with the kth and lth columns of **H**, must vanish. The resulting procedure is similar (but not identical) to the one from Appendix A. Thus, from the two vanishing principal minors $H_{1(24)}^{(24)}$ and $H_{1(13)}^{(13)}$,

$$H_{1(24)}^{(24)} = \begin{vmatrix} H'_{11} & H_{13} \\ H_{31} & H_{33} \end{vmatrix} = H'_{11}H_{33} - H_{13}H_{31} = H'_{11}H_{33} - \left|H_{13}\right|^2 = 0 \tag{C2a}$$

and

$$H_{1(13)}^{(13)} = \begin{vmatrix} H_{22} & H_{24} \\ H_{42} & H'_{44} \end{vmatrix} = H_{22}H'_{44} - H_{24}H_{42} = H_{22}H'_{44} - \left|H_{24}\right|^2 = 0 \tag{C2b}$$

one determines H'_{11} and H'_{44} after having evaluated H_{13} and H_{24} from Eqs. (A6) from Appendix A, together with H_{22} and H_{33} from Eqs. (B6*b*, *c*) from Appendix B. Knowing H'_{11} and H'_{44}, one finds next the absolute values, $\left|\operatorname{Im} H_{12}\right|$ and $\left|\operatorname{Im} H_{34}\right|$, of $\operatorname{Im} H_{12}$ and $\operatorname{Im} H_{34}$ from the two vanishing principal minors $H_{1(34)}^{(34)}$ and $H_{1(12)}^{(12)}$,

$$\begin{aligned} H_{1(34)}^{(34)} = \begin{vmatrix} H'_{11} & H_{12} \\ H_{21} & H_{22} \end{vmatrix} &= H'_{11}H_{22} - H_{12}H_{21} = H'_{11}H_{22} - \left|H_{12}\right|^2 \\ &= H'_{11}H_{22} - \left(\operatorname{Re} H_{12}\right)^2 - \left(\operatorname{Im} H_{12}\right)^2 = 0 \end{aligned} \tag{C3a}$$

and

$$\begin{aligned} H_{1(12)}^{(12)} = \begin{vmatrix} H_{33} & H_{34} \\ H_{43} & H'_{44} \end{vmatrix} &= H_{33}H'_{44} - H_{34}H_{43} = H_{33}H'_{44} - \left|H_{34}\right|^2 \\ &= H_{33}H'_{44} - \left(\operatorname{Re} H_{34}\right)^2 - \left(\operatorname{Im} H_{34}\right)^2 = 0 \end{aligned} \tag{C3b}$$

by using Eqs. (A4) from Appendix A for $\mathrm{Re}\,H_{12}$ and $\mathrm{Re}\,H_{34}$. The determination of the signs of $\mathrm{Im}\,H_{12}$ and $\mathrm{Im}\,H_{34}$ is identical to that of Appendix A. Use is made of the vanishing minor $H_{(23)}^{(14)}$, see Eq. (A5) from Appendix A, as well as of Eqs. (A6) yielding the two elements H_{13} and H_{24} that enter Eqs. (A5). The signs of $\mathrm{Im}\,H_{12}$ and $\mathrm{Im}\,H_{34}$ are chosen in such a way that $H_{12} = \mathrm{Re}\,H_{12} + i\,\mathrm{Im}\,H_{12}$ and $H_{34} = \mathrm{Re}\,H_{34} + i\,\mathrm{Im}\,H_{34}$ satisfy simultaneously both the real and the imaginary parts of Eq. (A5); a total of four sign combinations has to be checked. The two elements H_{12} and H_{34} are thus fully determined.

Next, one finds H_{23} from the vanishing minors $H_{1(13)}^{(12)}$ and $H_{1(34)}^{(24)}$,

$$H_{1(13)}^{(12)} = \begin{vmatrix} H_{23} & H_{24} \\ H_{43} & H_{44}^{'} \end{vmatrix} = H_{23}H_{44}^{'} - H_{24}H_{43} = H_{23}H_{44}^{'} - H_{24}H_{34}^{*} = 0 \qquad \text{(C4}a\text{)}$$

and

$$H_{1(34)}^{(24)} = \begin{vmatrix} H_{11}^{'} & H_{13} \\ H_{21} & H_{23} \end{vmatrix} = H_{11}^{'}H_{23} - H_{13}H_{21} = H_{11}^{'}H_{23} - H_{13}H_{12}^{*} = 0 \qquad \text{(C4}b\text{)}$$

from which one gets,

$$H_{23} = \frac{H_{24}H_{34}^{*}}{H_{44}^{'}} = \frac{H_{13}H_{12}^{*}}{H_{11}^{'}} = \frac{H_{24}H_{34}^{*} + H_{13}H_{12}^{*}}{H_{44}^{'} + H_{11}^{'}} \qquad \text{(C5)}$$

The last unknown H_{14} is determined from the Eq. (A2) from Appendix A knowing H_{23}. The missing last column elements of **M** are finally obtained from Eqs. (A1) from Appendix A.

Funding

This work was partially funded by Ministerio de Economía y Competitividad (EUIN2017-88598) and the European Commission (Polarsense, MSCA-IF-2017-793774).

Acknowledgments

The authors are grateful to their colleague Dr. E. Garcia-Caurel for having provided them with the diffraction grating sample.

Disclosures

The authors declare that there are no conflicts of interest related to this article.

References

1. R. M. A. Azzam and N. M. Bashara, *Ellipsometry and Polarized Light* (North Holland, Amsterdam, 1987).
2. R. A. Chipman, “Polarimetry”, in *Handbook of Opics II* (M. Bass Ed., McGraw Hill, New York, 1995).
3. H. Fujiwara, *Spectroscopic Ellipsometry: Principles and Applications* (Wiley, New York, 2007).
4. H. G. Tomkins and J. N. Hilfiker, *Spectroscopic Ellipsometry. Practical Application to Thin Film Characterization* (Momentum, New York, 2016).
5. P. S. Hauge, “Recent developments in instrumentation in ellipsometry”, *Surf. Sci.* **96**, 108-140 (1980).
6. J. J. Gil and E. Bernabeu, “Obtainment of the polarizing and retardation parameters of a non-depolarizing optical system from the polar decomposition of its Mueller matrix”, *Optik* **76**, 67-71 (1987).

7. R. Ossikovski, “Interpretation of nondepolarizing Mueller matrices based on singular value decomposition”, *J. Opt. Soc. Am.* A **25**, 473-482 (2008).
8. S.-Y. Lu and R. A. Chipman, “Interpretation of Mueller matrices based on polar decomposition”, *J. Opt. Soc. Am. A* **13**, 1106-1113 (1996).
9. R. Ossikovski, “Analysis of depolarizing Mueller matrices through a symmetric decomposition”, *J. Opt. Soc. Am. A* **26**, 1109-1118 (2009).
10. J. J. Gil and I. San José, “Polarimetric subtraction of Mueller matrices”, *J. Opt. Soc. Am. A* **30**, 1078-1088 (2013).
11. J. J. Gil, I. San José, and R. Ossikovski, “Serial-parallel decompositions of Mueller matrices”, *J. Opt. Soc. Am. A* **30**, 32-50 (2013).
12. R. Ossikovski, “Differential matrix formalism for depolarizing anisotropic media”, *Opt. Lett.* **36**, 2330-2332 (2011).
13. D. Goldstein, *Polarized Light* (Marcel Dekker, New York, 2003).
14. M. Schubert, “Generalized ellipsometry and complex optical systems”, *Thin Solid Films* **313-314**, 323-332 (1998).
15. T. Sato, T. Araki, Y. Sasaki, T. Tsuru, T. Tadokoro, and S. Kawakami, “Compact ellipsometer employing a static polarimeter module with arrayed polarizer and wave-plate elements”, *Appl. Opt.* **46**, 4963-4967 (2007).
16. G. E. Jellison, Jr. and F. A. Modine, “Polarization modulation ellipsometry” in *Handbook of Ellipsometry* (H. G. Tomkins and E. A. Irene Eds., W. Andrew, New York, 2005).
17. R. Ossikovski and O. Arteaga, “Complete Mueller matrix from a partial polarimetry experiment: the nine-element case”, submitted to *J. Opt. Soc. Am. A*
18. S. N. Savenkov, “Optimization and structuring of the instrument matrix for polarimetric measurements”, *Opt. Eng.* **41**, 965-972 (2002).
19. E. A. Oberemok and S. N. Savenkov, “Determination of the polarization characteristics of objects by the method of three probing polarizations”, *J. Appl. Spectrosc.* **69**, 72-77 (2002).
20. E. A. Oberemok and S. N. Savenkov, “Structure of deterministic Mueller matrices and their reconstruction in the method of three input polarizations”, *J. Appl. Spectrosc.* **70**, 224-229 (2003).
21. E. A. Oberemok and S. N. Savenkov, “Recovery of the complete Mueller matrix of an arbitrary object in the method of three input polarizations”, *J. Appl. Spectrosc.* **71**, 128-132 (2004).
22. S. Savenkov, R. Muttiah, E. Oberemok, and A. Klimov, “Incomplete Mueller polarimetry: Measurement of the block-diagonal scattering matrix”, *J. Quant. Spectrosc. Radiat. Transfer* **112**, 1796-1802 (2011).
23. S. R. Cloude, “Conditions for the physical realizability of matrix operators in polarimetry”, *SPIE Proc. Polarization Considerations for Optical Systems II*, vol. 1166, 177-185 (1989).
24. J. J. Gil and R. Ossikovski, *Polarized Light. The Mueller Matrix Approach* (CRC Press, Boca Raton, 2016).
25. R. Ossikovski and J. J. Gil, “Basic properties and classification of Mueller matrices derived from their statistical definition”, *J. Opt. Soc. Am. A* **34**, 1727-1737 (2017).
26. O. Arteaga and A. Canillas, “Analytic inversion of the Mueller–Jones polarization matrices for homogeneous media”, *Opt. Lett.* **35**, 559-561; (erratum) 3525-3525 (2010).
27. M. Foldyna, A. De Martino, R. Ossikovski, E. Garcia-Caurel, and C. Licitra, “Characterization of grating structures by Mueller polarimetry in presence of strong depolarization due to finite spot size”, *Opt. Commun.* **282**, 735-741 (2009).
28. M. Foldyna, E. Garcia-Caurel, R. Ossikovski, A. De Martino, and J. J. Gil, “Retrieval of a non-depolarizing component of experimentally determined depolarizing Mueller matrices”, *Opt. Express* **17**, 12794-12806 (2009).
29. R. Ossikovski, E. Garcia-Caurel, M. Foldyna, and J. J. Gil, “Application of the arbitrary decomposition to finite spot size Mueller matrix measurements”, *Appl. Opt.* **53**, 6030-6036 (2014).
30. E. Kuntman and O. Arteaga, “Decomposition of a depolarizing Mueller matrix into its nondepolarizing components by using symmetry conditions”, *Appl. Opt.* **55**, 2543-2550 (2016).
31. E. Kuntman, A. Canillas, and O. Arteaga, “Retrieval of nondepolarizing components of depolarizing Mueller matrices by using symmetry conditions and least squares minimization”, *Appl. Surf. Sci.* **421**, 697-701 (2017).
32. M. Kildemo, R. Ossikovski, and M. Stchakovsky, “Measurement of the absorption edge of thick transparent substrates using the incoherent reflection model and spectroscopic UV-visible-near-IR ellipsometry”, *Thin Solid Films* **313-314**, 108-113 (1998).
33. R. Ossikovski, M. Stchakovsky, M. Kildemo, and M. Mooney, “Incoherent reflection model for spectroscopic ellipsometry of a thick semi-transparent anisotropic substrate”, *Appl. Opt.* **39**, 2071-2077 (2000).
34. S. Nichols, O. Arteaga, A. Martin, and B. Kahr, “Measurement of transmission and reflection from a thick anisotropic crystal modeled by a sum of incoherent partial waves”, *J. Opt. Soc. Am. A* **32**, 2049-2057 (2015).
35. O. Arteaga, J. Freudenthal, S. Nichols, A. Canillas, and Bart Kahr, “Transmission ellipsometry of anisotropic substrates and thin films at oblique incidence. Handling multiple reflections”, *Thin Solid Films* **571**, 701-705 (2014).
36. H. C. van de Hulst, *Light Scattering by Small Particles* (Dover, New York, 1981).
37. O. Arteaga, J. Freudenthal, B. Wang, and B. Kahr, “Mueller matrix polarimetry with four photoelastic modulators: theory and calibration”, *Appl. Opt.* **51**, 6805-6817 (2012).