Mueller polarimetry involves a variety of instruments and technologies whose importance and scope of applications are rapidly increasing. The exploitation of these powerful resources depends strongly on the mathematical models that underlie the analysis and interpretation of the measured Mueller matrices and, very particularly, on the theorems for their serial and parallel decompositions. In this letter, the most general formulation for the parallel decomposition of a Mueller matrix is presented, which overcomes certain critical limitations of the previous approaches. In addition, the results obtained lead to a generalization of the polarimetric subtraction procedure and allow for a formulation of the arbitrary decomposition that integrates, in a natural way, the passivity criterion.

A complementary criterion refers to passivity and implies that the action of the medium does not amplify the intensity of the electromagnetic wave interacting with it. More specifically, the assumption of the ensemble criterion entails the necessity that a physically realizable Mueller matrix is susceptible to be expressed as a convex combination of pure and passive Mueller matrices [6]. The main aim of this work is the formulation, in the most general form, of the arbitrary decomposition of a Mueller matrix into a convex sum of a minimum number of pure Mueller matrices, in such a manner that the new formulation overcomes the limitation of the previous approaches [7-13] where the Mueller matrices of all parallel components have to be normalized to have equal values for their mean intensity coefficients (defined below), and therefore opens strongly the scope of its applications. This result also provides the way to express the arbitrary decomposition in terms of passive Mueller matrices in accordance with the passivity criterion and, furthermore, leads to a generalization of the polarimetric subtraction procedure [12].

To simplify further expressions, the partitioned block expression of a Mueller matrix [14] will be used when appropriate

\[
\mathbf{M} = m_{00} \mathbf{M} \quad \mathbf{M} = \begin{pmatrix} 1 & \mathbf{D}^T \\ \mathbf{P} & \mathbf{m} \end{pmatrix},
\]

\[
\mathbf{m} = \begin{pmatrix} m_{11} \\ m_{21} \\ m_{31} \\ m_{12} \\ m_{22} \\ m_{32} \end{pmatrix},
\]

\[
\mathbf{D} = \begin{pmatrix} m_{41}, m_{51}, m_{61} \\ m_{42}, m_{52}, m_{62} \end{pmatrix}^T \quad \mathbf{P} = \begin{pmatrix} m_{00}, m_{20}, m_{30} \\ m_{00} \end{pmatrix}^T,
\]

where the superscript \( T \) indicates transpose, \( m_{00} \) is the mean intensity coefficient (MIC) (i.e., the transmittance or gain [15-19] of \( \mathbf{M} \) for input unpolarized light), while \( \mathbf{D} \) and \( \mathbf{P} \) are the respective diattenuation and polarization vectors of \( \mathbf{M} \) [20]. The absolute values of these vectors are the diattenuation \( D = |\mathbf{D}| \) and the polarization \( P = |\mathbf{P}| \) [20,21]. \( \mathbf{M} \) denotes the normalized form of \( \mathbf{M} \) with MIC \( m_{00} = 1 \). It is also worth to recall that, given the peculiar mathematical structure of a Mueller matrix, its transposed matrix \( \mathbf{M}^T \) is also a Mueller matrix [22,23].

Let us consider a light beam with a given state of polarization determined by the corresponding Stokes vector \( \mathbf{s} \), whose spot size on a material sample covers \( n \) areas with different deterministic nondepolarizing polarimetric behavior and that the exiting light pencils are incoherently recombined, so that the state of polarization of the whole outgoing beam is represented by a Stokes vector \( \mathbf{s}' \). Thus, the total intensity \( I \) of the incoming light is shared among \( n \) portions \( I_i \) falling on respective elements \( r \) contained in the illuminated area, which are represented by their respective pure Mueller matrices \( \mathbf{M}_{s_i} \) (Fig. 1). The polarimetric
transformation of the input Stokes vector \( \mathbf{s} \) into the output \( \mathbf{s}' \) is given by

\[
\mathbf{s}' = \sum_{i=1}^{k} \left( \frac{l_i}{k} \mathbf{M}_i \right) \mathbf{s} = \sum_{i=1}^{k} k_i \mathbf{M}_i \mathbf{s},
\]

(2)

where \( k = \sum_{i=1}^{k} k_i = 1 \).

The explicit expressions for \( \mathbf{H} (\mathbf{M}) \) and \( \mathbf{M} (\mathbf{H}) \) can be found in [1,25].

Since \( \mathbf{H} \) is a positive semidefinite Hermitian matrix [4], it can be diagonalized as

\[
\mathbf{H} = \mathbf{U} \text{diag} (\lambda_1, \lambda_2, \lambda_3, \lambda_4) \mathbf{U}^\dagger,
\]

(6)

where \( \lambda_i \) are the four non-negative eigenvalues of \( \mathbf{H} \), taken in decreasing order \((0 \leq \lambda_1 \leq \lambda_2 \leq \lambda_3 \leq \lambda_4)\). The columns \( \mathbf{u}_i \) \((i = 0, 1, 2, 3)\) of the \( 4 \times 4 \) unitary matrix \( \mathbf{U} \) are the respective unit, mutually orthogonal, eigenvectors of \( \mathbf{H} \).

Therefore, \( \mathbf{H} \) can be expressed as the following convex linear combination of four rank-1 covariance matrices that represent respective pure systems

\[
\mathbf{H} = \sum_{i=0}^{3} \lambda_i \mathbf{H}_{i}, \quad \mathbf{H}_{i} = m_{00} \left( \mathbf{u}_i \otimes \mathbf{u}_i^\dagger \right), \quad m_{00} = \text{tr} \mathbf{H}. \tag{7}
\]

This (Claude decomposition [4], or spectral decomposition) can be written in terms of the corresponding Mueller matrices by means of the following convex sum

\[
\mathbf{M} = \sum_{i=0}^{3} \lambda_i \mathbf{M}_{i}, \quad \left( \mathbf{M}_{i} \right)_{00} = m_{00} = \text{tr} \mathbf{H}, \tag{8}
\]

where all pure Mueller matrices \( \mathbf{M}_{i} \) have equal MIC, equal to \( m_{00} \).

Prior to establish the expressions for the general decomposition of a depolarizing \( \mathbf{M} \) in terms of a minimum number of pure incoherent components of \( \mathbf{M} \), let us recall that it has been shown that such number is given by \( r = \text{rank} (\mathbf{H}) \) [8,12].

While the components of the spectral decomposition are defined from the respective eigenvectors \( \mathbf{u}_i \) of \( \mathbf{H} \) with nonzero eigenvalue, any Mueller matrix also admit the so-called arbitrary decomposition [8,12] (hereafter homogeneous arbitrary decomposition)

\[
\mathbf{M} = \sum_{i=0}^{r} p_i \mathbf{M}_{i} = \sum_{i=0}^{r} p_i m_{00} \mathbf{M}_{i},
\]

(9)

\[
\left( \mathbf{M}_{i} \right)_{00} = \text{tr} \left[ \left( \sigma_i \otimes \sigma_i^\dagger \right) \left( \mathbf{w}_i \otimes \mathbf{w}_i^\dagger \right) \right],
\]

\[
P_i = \frac{1}{m_{00}} \sum_{i=0}^{3} \lambda_i \left( \mathbf{w}_i \otimes \mathbf{w}_i^\dagger \right)^\dagger, \quad \sum_{i=0}^{r} P_i = 1,
\]

where the subscripts \( i \) run the elements of \( \mathbf{M}_{i} \), \( i = 0, \ldots, r \) is a set of \( r \) independent unit vectors belonging to the image subspace of \( \mathbf{H} \) (denoted as \( \text{im} (\mathbf{H}) \)) [12]. Note that when \( \mathbf{w}_i = \mathbf{u}_i \) (\( \mathbf{u}_i \) being the unit eigenvectors of \( \mathbf{H} \) with nonzero eigenvalue), then the arbitrary decomposition adopts the particular form of the spectral decomposition.

The detailed demonstration of the homogeneous arbitrary decomposition can be found in [12]. Note that the denominator of the expression of \( p_i \) in (9) can also be expressed as

\[
m_{00} \sum_{i=0}^{r} \lambda_i \left( \mathbf{w}_i \otimes \mathbf{w}_i^\dagger \right)^\dagger = m_{00} \mathbf{H} \mathbf{w}_i \mathbf{w}_i^\dagger.
\]

(10)

where \( \mathbf{H}^{-1} \) is the pseudoinverse of \( \mathbf{H} \) defined as \( \mathbf{H}^{-1} = \mathbf{U} \text{diag} (\lambda_1, \lambda_2, \lambda_3, \lambda_4) \mathbf{U}^\dagger \), \( \mathbf{D} \) being the diagonal matrix whose \( r \) first diagonal elements are \( 1/\lambda_1, 1/\lambda_2, \ldots, 1/\lambda_r \) and the last \( 4 - r \) elements are zero.

Decompositions (8) and (9) have been formulated for the case where all pure components have equal MICs, equal to \( m_{00} \). This
exigency should be avoided because the MICs of the pure components can take specific independent values. In fact, as it will be shown below by means of an example, the homogeneous arbitrary decomposition (9) is not always physically realizable in terms of passive Mueller matrices.

Prior to introduce the generalization of the arbitrary decomposition that contains pure components with arbitrary respective MICs, let us note that by writing a given parallel decomposition (2) in the form (9) and comparing the "f" elements appearing in the respective summations in (2) and (9), it follows that

\[ k_j m_{00j} \hat{M}_k = p_j m_{00j} \hat{M}_k \Rightarrow k_j m_{00j} = p_j m_{00j}, \]

and therefore the arbitrary decomposition can be expressed in the following generalized form where the components have different MICs denoted as \( m_{00j} \) [note that the synthesized expression in (10) is applied]

\[ \hat{M} = \sum_{i=1}^{n} k_i \hat{M}_k = \sum_{i=1}^{n} k_i m_{00j} \hat{M}_k, \]

\[ k_j = \frac{1}{m_{00j}} \hat{w}_i \hat{H} \hat{w}_j, \quad \sum_{i=1}^{n} k_i = 1. \]

Since passivity is a natural feature of experimental samples, the arbitrary decomposition should be performed in terms of passive Mueller matrices. To do so, let us first recall that the necessary and sufficient conditions for a Mueller matrix \( \hat{M} \) to be passive are the following [6,26]

\[ m_{00}(1+D) \leq 1, \quad m_{00}(1+P) \leq 1. \]  

(13)

(Note that, in the case of a pure Mueller matrix, the equality \( P = D \) is satisfied [22] and both conditions become a single one). Therefore, the passive formulation of the arbitrary decomposition adopts the form

\[ q \hat{M} = \sum_{i=1}^{n} k_i \hat{M}_k = \sum_{i=1}^{n} \frac{q_j}{1+X_j} \hat{M}_k, \]

\[ k_j = \frac{1+X_j}{q_j \sum_{i=1}^{n} \left( U^* \hat{w}_i \right)^2}, \quad \sum_{i=1}^{n} k_i = 1, \]

where

\[ X = \max(D,P), \quad X_j = \max(D_j,P_j), \]

\[ q = m_{00}(1+X) \leq 1, \quad q_j = m_{00j}(1+X_j) \leq 1. \]

(14)

(15)

To illustrate the above results, let us consider a parallel composition of two elements, namely, a quarter-wave plate oriented at \( 0^\circ \) with Mueller matrix \( \hat{M}_q \), and a linear polarizer oriented at \( 0^\circ \) with Mueller matrix \( \hat{M}_p \), such that the spot size of the uniform light beam that illuminates this system is shared in such a manner that \( 1/3 \) of the intensity falls on the retarder and the remaining \( 2/3 \) fall on the polarizer. The composed Mueller matrix \( \hat{M} \) is obtained as follows

\[ \hat{M} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \]

\[ \hat{M}_q = \begin{pmatrix} 1 & 1 & 0 & 0 \\ 1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}, \]

\[ \hat{M}_p = \begin{pmatrix} 0 & 1/2 & 0 & 0 \\ 1/2 & 1 & 0 & 0 \\ 0 & 0 & 0 & -1/3 \\ 0 & 0 & 1/3 & 0 \end{pmatrix}. \]

(16)

so that \( m_{00} = 1, \ m_{02} = 1/2, \ m_{00} = 2/3, \ k_1 = 1/3 \) and \( k_2 = 2/3 \). The corresponding homogenous decomposition of \( \hat{M} \) takes the form

\[ \hat{M} = \frac{1}{2} \hat{M}_q + \frac{2}{3} \hat{M}_p. \]

(17)

where the respective coefficients are \( p_1 = k_1 m_{00} / m_{00} = 1/2 \) and \( p_2 = k_2 m_{00} / m_{00} = 1/2 \). Note that, since \( m_{00} = 2/3 > 1/2 \), the polarizer \( (2/3) \hat{M}_p \) in the homogeneous decomposition does not satisfy the passivity conditions (13) and, therefore, it is not physically realizable. Obviously, given a measured Mueller matrix \( \hat{M} \), the arbitrary decomposition provides infinite specific parallel decompositions of it, and it is the experimentalist, with his experience and knowledge of the problem and its constraints, who can decide which decomposition is more appropriate or plausible for each situation.

As a numerical additional example, let us now consider the application of the arbitrary decomposition (12) to the following experimentally determined Mueller matrix, which corresponds to the reflection (angle of incidence 50°) on a steel specimen with surface roughness of 0.256-μm, and with a MgF2 film thickness of 89 nm [27].

\[ \hat{M} = \begin{pmatrix} 1.0000 & 0.1631 & -0.0322 & 0.0802 \\ 0.0083 & 0.4038 & 0.2555 & -0.2158 \\ -0.0026 & 0.4297 & -0.1376 & 0.2016 \\ -0.0116 & 0.0597 & -0.3175 & -0.3690 \end{pmatrix}. \]

(18)

The eigenvalues of the covariance matrix \( \hat{H} \) associated with \( \hat{M} \) have the nonnegative values

\[ \lambda_1 = 0.6270, \quad \lambda_2 = 0.1888, \quad \lambda_3 = 0.1292, \quad \lambda_4 = 0.0550. \]

(19)

showing that \( \hat{M} \) satisfies the covariance conditions and that \( r = 4 \) (within the assumed experimental accuracy limits), that is, any arbitrary decomposition should include four parallel components. Other relevant quantities of \( \hat{M} \) that are invariant with respect to dual-retarder transformations [28] are

\[ D = 0.1846, \quad P = 0.0145, \quad P_5 = 0.5032 \] (\( P_5 \) being the degree of spherical purity [29]),

\[ P_6 = 0.5144 \] (\( P_6 \) being the depolarization index, [21], also called degree of polarimetric purity [8]) and

\[ \det \hat{M} = 0.1176. \]

Note that, in this case \( D > P \), so that the passive representative [6] \( \hat{M} = (\hat{M} / \det \hat{M}) \), (i.e. the Mueller matrix \( \hat{M} \) proportional to \( \hat{M} \) having the maximal MIC compatible with passivity) is given by

\[ \hat{M} = \frac{1}{1+D} \hat{M} = \begin{pmatrix} 0.8442 & 0.1377 & -0.0272 & 0.0677 \\ 0.0070 & 0.3409 & 0.2157 & -0.1822 \\ -0.0022 & 0.3627 & -0.1162 & 0.1702 \\ -0.0098 & 0.0504 & -0.2680 & -0.3115 \end{pmatrix}. \]
which admit (among other) an arbitrary decomposition of the form (12), where

\[
\mathbf{M}_{J1} = \begin{pmatrix}
1.0000 & 0.0000 & 0.0000 & 0.0000 \\
0.0000 & 0.3203 & 0.6300 & -0.7074 \\
0.0000 & 0.8373 & 0.1610 & 0.5225 \\
0.0000 & 0.4431 & -0.7597 & -0.4760
\end{pmatrix},
\]

\[
\mathbf{M}_{J2} = \begin{pmatrix}
1.0000 & 0.0000 & 0.0000 & 0.0000 \\
0.0000 & 0.9794 & -0.1341 & -0.1803 \\
0.0000 & -0.0825 & -0.9598 & 0.2682 \\
0.0000 & -0.2091 & -0.2465 & -0.9463
\end{pmatrix},
\]

\[
\mathbf{M}_{J3} = \begin{pmatrix}
0.6532 & 0.3064 & -0.0605 & 0.1507 \\
0.2774 & 0.3255 & -0.0377 & 0.5254 \\
0.1360 & 0.4399 & 0.3158 & -0.1781 \\
-0.1576 & -0.3176 & 0.4571 & 0.1464
\end{pmatrix},
\]

\[
\mathbf{M}_{J4} = \begin{pmatrix}
0.5527 & 0.3953 & -0.0780 & 0.1944 \\
-0.3577 & -0.4652 & 0.1285 & -0.0195 \\
-0.2096 & -0.2058 & -0.0206 & -0.2583 \\
0.1679 & 0.0529 & -0.2338 & 0.2758
\end{pmatrix},
\]

with

\[
k_1 = 0.3713,
\]

\[
k_2 = 0.2270,
\]

\[
k_3 = 0.2373,
\]

\[
k_4 = 0.1644,
\]

\[
(k_1 + k_2 + k_3 + k_4 = 1).
\]

Note that, unlike what would occur with homogeneous arbitrary decompositions, in this particular parallel decomposition the MICs of the components are different, allowing, in addition, that all of them are represented by passive Mueller matrices simultaneously.

Once the arbitrary decomposition has been generalized and even expressed in terms of passive elements, let us revisit the procedure for the polarimetric subtraction and formulate it in the light of this new framework. A given pure component \( \mathbf{M}_{Ji} \), with associated \( \mathbf{H}_{Ji} = m_{00i} \left( \mathbf{w}^\dagger \otimes \mathbf{w}_i \right) \), can be considered an arbitrary component of \( \mathbf{M} \) if and only if \( \text{rank} (\mathbf{H} + \mathbf{H}_{Ji}) = \text{rank} \mathbf{H} \) [12]. If this inclusion (or subtractability) criterion is satisfied, then, in accordance with (12), the coefficient \( k_i \) corresponding to \( \mathbf{H}_{Ji} \) in the arbitrary decomposition is given by

\[
k_i = \frac{1}{m_{00i}} \sum_{j=1}^{4} \frac{1}{\lambda_j} \left( \mathbf{U}^\dagger \mathbf{H} \mathbf{w}_j \right)^2 = \frac{1}{m_{00i}} \left( \mathbf{w}_i^\dagger \mathbf{H} \mathbf{w}_i \right),
\]

(22)

The polarimetric subtraction of \( \mathbf{M}_{Ji} \) from \( \mathbf{M} \) is then performed in the following manner [12]

\[
\mathbf{M} = \frac{1}{1 - k_i} \left( \mathbf{M} - k_i \mathbf{M}_{Ji} \right),
\]

(23)

where the rank of the covariance matrix \( \mathbf{H} \), associated with the resulting matrix \( \mathbf{M} \), is \( r-1 \).

If other pure elements are wanted to be consecutively subtracted, the subtraction procedure can be iterated until the difference matrix obtained has rank equal to 1. In each step, the inclusion criterion \( \text{rank} (\mathbf{H} + \mathbf{H}_{Ji}) = \text{rank} \mathbf{H} \) should be checked. In addition, as shown in [12], the subtraction of nonpure elements can also be performed and its formulation from the generalized form of the arbitrary decomposition (12) is straightforward.

In summary, unlike the previous approaches, the arbitrary decomposition has been formulated in its most general form, thus allowing one to apply it to any physical or experimental situation and providing the appropriate procedure for the calculation of the coefficients of the parallel components. The new approach has been also expressed in terms of Mueller matrices satisfying the passivity criterion required by natural and man-made samples (except for certain artificial situations [30]). Furthermore, the polarimetric subtraction procedure has been revisited and reformulated in the light of the new generalized arbitrary decomposition framework presented.

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