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Sum regression decomposition of spectral and angle-resolved Mueller matrices from biological reflectors

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We show spectroscopic Mueller-matrix data measured at multiple incidence angles of the scarab beetle *C. aurata*. The Mueller matrix is decomposed by method of regression decomposition and it is shown that this Mueller matrix can be decomposed into a set of two matrices representing one polarizer and one dielectric reflector. We also report on a tentative decomposition of the beetle *C. argenteola* using the same method. © 2016 Optical Society of America

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1. INTRODUCTION

A Mueller matrix is a complete description of the linear optical response of a sample when light is reflected specularly off it or transmitted through it [1]. With Mueller-matrix spectroscopic ellipsometry (MMSE), the spectral and angle of incidence dependence of a Mueller matrix for a sample can be measured accurately [2] and can be used to extract optical and structural properties of samples composed of complex nanostructured materials. Mueller matrices are based on a Stokes vector description of light and thus have the benefit of being able to include not only interaction with polarized light but also with unpolarized and partially polarized light. MMSE is frequently used in analysis of bulk and thin film structures and during the last decade it has also been employed to study natural photonic structures [3, 4]. These results may be helpful in biomimicry as nature has long been a source of inspiration for developing and refining technologies in materials science. Structural coloring may serve as one example and mimicking of structures of *morpho* butterflies could lead to environmentally friendly long-lasting colors [5]. In this report we present studies on beetles in the Scarabaeidae family. The selected beetles show brilliant colors and in addition interesting polarization features. Mueller matrices measured on such beetles are of large interest to explore for biomimetics and for the understanding of the biological relevance of the observed polarization phenomena. Several species in the Scarabaeidae family have been studied by Hodgkinson [6], Goldstein [3] and our group [4, 7] to mention a few. Ellipticity, degree of polar-

ization and other derived parameters have been reported [4] and Arwin et al. [7] also did optical modeling to determine structural parameters of the scutellum part of the exoskeleton of *Cetonia aurata*.

Mueller matrices are very rich in information about the sample properties and can also be analyzed by addressing depolarization. Cloude [8] showed that a depolarizing Mueller matrix can be represented by a sum of up to four non-depolarizing Mueller matrices weighted by the eigenvalues of the covariance matrix of the Mueller matrix. These eigenvalues are all positive for a physically realizable Mueller matrix and this, so called sum decomposition can be used to filter matrices and obtain a measure of experimental fidelity [9]. The result of the decomposition can also be used to describe a Mueller matrix as a set of basic optical elements having direct physical meaning, such as polarizers and retarders. Pioneering work on decomposition of Mueller-matrix images, including studies of beetles, was performed by Ossikovski et al. [10]. We have also previously [11] demonstrated this with Cloude as well as regression decomposition of Mueller-matrix spectra and images measured at near-normal incidence on *C. aurata*. Using Cloude decomposition we found that the experimentally determined Mueller matrix of *C. aurata* at near normal incidence decomposes into a set of a mirror and a circular polarizer. Those results were then the basis for a more stable regression decomposition where the result was confirmed.

Our objective in this work is to further develop the parameterization of Mueller matrices of beetles in terms of basic optical

elements using regression decomposition. We generalize the previous decomposition by including angle-of-incidence depending Mueller matrices which leads to more complicated decompositions.

2. EXPERIMENTAL DETAILS

We have measured Mueller matrices of two species of scarab beetles: *Cetonia aurata* (Linnaeus, 1758) collected on the island Öland in Sweden and *Chrysina argenteola* (Bates, 1888) which is on loan from the Natural Museum of History in Stockholm, Sweden, but originates from Colombia. MMSE measurements were performed on the scutellum, which is a small triangular and relatively flat area on the dorsal side of the beetle. The incidence angles were in the range 15° to 55° . The ellipsometer used is a dual rotating compensator ellipsometer (RC2[®]) from J.A. Woolam Co., Inc. equipped with focusing optics to reduce the spot size to $< 100 \mu\text{m}$. The spectral range of the instrument is 245–1690 nm but only data in the range 245–1000 nm are presented here. All calculations regarding decomposition was made using the lsqcurvefit function in Matlab.

3. THEORY

When the polarization state of light is described with a Stokes vector, the incoming and outgoing waves are related by a 4×4 Mueller matrix $\mathbf{M}$ as [1]

$$\mathbf{S}_o = \mathbf{M}\mathbf{S}_i \quad (1)$$

where $\mathbf{S}_i$ and $\mathbf{S}_o$ are Stokes vectors for the incoming and the outgoing beams of light, respectively. If we expand the equation we get

$$\begin{bmatrix} I_o \\ Q_o \\ U_o \\ V_o \end{bmatrix} = \begin{bmatrix} 1 & m_{12} & m_{13} & m_{14} \\ m_{21} & m_{22} & m_{23} & m_{24} \\ m_{31} & m_{32} & m_{33} & m_{34} \\ m_{41} & m_{42} & m_{43} & m_{44} \end{bmatrix} \begin{bmatrix} I_i \\ Q_i \\ U_i \\ V_i \end{bmatrix} \quad (2)$$

where I , Q , U and V denote the Stokes parameters [1] and m_{ij} ($i, j = 1, 2, 3, 4$) are the elements of $\mathbf{M}$. $\mathbf{M}$ and $\mathbf{S}_i$ are normalized to m_{11} and I_i , respectively. Based on this formalism we briefly describe sum decomposition and present some basic Mueller matrices used in this work.

A. Regression sum decomposition

Cloude has shown that a depolarizing Mueller matrix can be represented as a sum of up to four non-depolarizing matrices [8]. As an alternative to Cloude's method, regression decomposition can be used as recently shown for spectral data and Mueller-matrix images on *C. aurata* [11]. An experimentally determined normalized Mueller matrix $\mathbf{M}$ is then decomposed into a sum of Mueller matrices $\mathbf{M}_i^{\text{reg}}$ representing optical devices such as polarizers, retarders and rotators.

$$\mathbf{M}^{\text{reg}} = a\mathbf{M}_1^{\text{reg}} + b\mathbf{M}_2^{\text{reg}} + c\mathbf{M}_3^{\text{reg}} + d\mathbf{M}_4^{\text{reg}} \quad (3)$$

The coefficients a , b , c and d are fit parameters and the matrices $\mathbf{M}_1^{\text{reg}}$, $\mathbf{M}_2^{\text{reg}}$, $\mathbf{M}_3^{\text{reg}}$, $\mathbf{M}_4^{\text{reg}}$ are defined beforehand. The decomposition can be made using matrices with fixed values on all elements or with generalized matrices whereby some matrix parameters can also be fitted. Later we will describe examples

of such matrices that have been used in this paper. Any Mueller-matrix decomposition based on Cloude's method will result in up to four trace-orthogonal matrices. The matrices possible to use in our regression-based decomposition are not necessarily trace-orthogonal and thus an infinite number of sum decompositions are possible. In fact, Eq. 3 is a special form of the arbitrary decomposition of a Mueller matrix [12] where four terms are included but they can be fewer or more depending on the complexity of $\mathbf{M}$. However, the number of fit parameters including those within the matrices can not exceed the degrees of freedom of $\mathbf{M}$. The number of non-zero matrix components in Eq. 2 can be determined from Cloude's decomposition. In the regression procedure, the Frobenius norm [13]

$$F = \|\mathbf{M} - \mathbf{M}^{\text{reg}}\|_F \quad (4)$$

is minimized and the coefficients a , b , c , d are all positive and $a + b + c + d = 1$.

B. Mueller matrices for rotated polarizing components

If an optical device is rotated an angle α counterclockwise with respect to the x -axis in a reference Cartesian coordinate system when looking into the source, the Mueller matrix $\mathbf{M}'$ of the rotated device will be

$$\mathbf{M}' = \mathbf{R}(-\alpha)\mathbf{M}\mathbf{R}(\alpha) \quad (5)$$

where $\mathbf{M}$ is the Mueller matrix of the unrotated device and $\mathbf{R}(\alpha)$ is given by [14]

$$\mathbf{R}(\alpha) = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos 2\alpha & \sin 2\alpha & 0 \\ 0 & -\sin 2\alpha & \cos 2\alpha & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix} \quad (6)$$

C. Polarizers

A polarizer will attenuate two orthogonal components of the electric field in a plane wave by different amounts. An ideal linear polarizer with its transmission axis in the x -direction is given by [1]

$$\mathbf{M}_p^{\text{linear}} = \frac{1}{2} \begin{bmatrix} 1 & 1 & 0 & 0 \\ 1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix} \quad (7)$$

If such an ideal polarizer is rotated so that the transmission axis becomes oriented at an angle α from the x -axis, its Mueller matrix as obtained from Eq. 5 will be

$$\mathbf{M}_p^{\text{linear}}(\alpha) = \frac{1}{2} \begin{bmatrix} 1 & \cos 2\alpha & \sin 2\alpha & 0 \\ \cos 2\alpha & \cos^2 2\alpha & \sin 2\alpha \cos 2\alpha & 0 \\ \sin 2\alpha & \sin 2\alpha \cos 2\alpha & \sin^2 2\alpha & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix} \quad (8)$$

A circular polarizer will attenuate left-handed and right-handed polarized light by different amounts and its Mueller matrix is

given by

$$\mathbf{M}_P^{\text{circular}} = \frac{1}{2} \begin{bmatrix} 1 & 0 & 0 & \pm 1 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ \pm 1 & 0 & 0 & 1 \end{bmatrix} \quad (9)$$

where $m_{14} = m_{41} = 1$ is for right-handed and -1 for left-handed polarization capability. Linear and circular polarizers are special cases of the more general elliptical polarizer and a general elliptical polarizer with azimuth angle equal to 0° or 90° has a Mueller matrix given by

$$\mathbf{M}_P^{\text{elliptic}}(\varepsilon) = \frac{1}{2} \begin{bmatrix} 1 & \cos 2\varepsilon & 0 & \sin 2\varepsilon \\ \cos 2\varepsilon & \cos^2 2\varepsilon & 0 & \cos 2\varepsilon \sin 2\varepsilon \\ 0 & 0 & 0 & 0 \\ \sin 2\varepsilon & \cos 2\varepsilon \sin 2\varepsilon & 0 & \sin^2 2\varepsilon \end{bmatrix} \quad (10)$$

where ε is the ellipticity angle of the major eigenpolarization [15]. For incident unpolarized light, the Stokes vector of emerging light moves along the meridian of the Poincaré sphere, passing through horizontal linear, right-handed circular, vertical linear and left-handed circular polarization states as 2ε goes through values 0° , 90° , 180° and 270° , respectively. With an arbitrary azimuth angle α , Eqs. 5 and 10 lead to

$$\mathbf{M}_P^{\text{elliptic}}(\varepsilon, \alpha) = \frac{1}{2} \begin{bmatrix} 1 & ab & ac & d \\ ab & a^2b^2 & a^2bc & abd \\ ac & a^2bc & a^2c^2 & acd \\ d & abd & acd & d^2 \end{bmatrix} \quad (11)$$

where $a = \cos 2\varepsilon$, $b = \cos 2\alpha$, $c = \sin 2\alpha$ and $d = \sin 2\varepsilon$. Eq. 11 corresponds to transmission mode but in reflection off a symmetric, reciprocal material it has been shown [16, 17] that the off-diagonal elements of the Jones matrix are anti symmetrical and therefore the Mueller matrix will satisfy the following symmetries: $m_{31} = -m_{13}$, $m_{32} = -m_{23}$ and $m_{34} = -m_{43}$ (see e.g. Garcia-Caurel et al. [18]) and we obtain

$$\mathbf{M}_{P,R}^{\text{elliptic}}(\varepsilon, \alpha) = \frac{1}{2} \begin{bmatrix} 1 & ab & ac & d \\ ab & a^2b^2 & a^2bc & abd \\ -ac & -a^2bc & a^2c^2 & -acd \\ d & abd & acd & d^2 \end{bmatrix} \quad (12)$$

D. Isotropic reflector

An isotropic reflector used at oblique incidence introduces retardation δ and diattenuation $\cos 2\psi$ between the electric field components in two orthogonal directions parallel and perpendicular to the plane of incidence [2].

$$\mathbf{M}_R^{\text{iso}} = \begin{bmatrix} 1 & -\cos 2\psi & 0 & 0 \\ -\cos 2\psi & 1 & 0 & 0 \\ 0 & 0 & \sin 2\psi \cos \delta & \sin 2\psi \sin \delta \\ 0 & 0 & -\sin 2\psi \sin \delta & \sin 2\psi \cos \delta \end{bmatrix} \quad (13)$$

Two special cases are noted. One occurs when δ is 180° and ψ is 45° . Eq. 13 then becomes

$$\mathbf{M}_M = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & -1 \end{bmatrix} \quad (14)$$

which is an ideal mirror. The second case occurs when δ is 180° and ψ is 0° and then Eq. 13 becomes

$$\mathbf{M}_{LP} = \begin{bmatrix} 1 & -1 & 0 & 0 \\ -1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix} \quad (15)$$

which is a linear polarizer with its absorption axis parallel to the plane of incidence.

4. RESULTS AND DISCUSSION

A. Regression sum decomposition of Mueller matrices of *C. aurata*

Mueller matrices measured on *C. aurata* at incidence angles in the range 15° to 55° with a step size of 1° in the spectral range 245 nm to 1000 nm are summarized in a contour plot in Fig. 1. Below 500 nm and above 600 nm all off-diagonal block elements

Fig. 1. A contour plot of experimentally determined 4×4 Mueller matrices of *C. aurata*.

as well as m_{34} and m_{43} are almost constant and close to zero and the rest of the elements have monotonously changing values with increasing incidence angle. Between 500 nm and 600 nm, however, many elements show dramatic changes. In this region the Mueller matrix is depolarizing and can therefore be sum decomposed according to Cloude [8] as shown in our previous report [11]. We call this region the circularly polarizing regime and will be referred to as the P_C -regime. Within the P_C -regime the cuticle will reflect incident unpolarized light as near-circular polarized at small angles of incidence θ with an ellipticity decreasing with θ . In our previous work based on near-normal incidence data, we used an ansatz with a decomposition into a circular polarizer and a dielectric mirror. A circular polarizer is a special case of an elliptical polarizer and a dielectric mirror can be represented by a retarder with $\delta = 180^\circ$ with the conventions used. Here we expand the circular polarizer to an elliptical polarizer and the mirror to a isotropic reflector according to Eqs. 10 and 13, respectively.

We apply a regression analysis according to Eqs. 3 and 4 with the ansatz

$$\mathbf{M}^{reg} = a(\lambda, \theta) \mathbf{M}_{P,R}^{elliptic}(\alpha, \varepsilon) + b(\lambda, \theta) \mathbf{M}_R^{Iso}(\psi, \delta) \quad (16)$$

where λ and θ are wavelength and incidence angle, respectively; $\mathbf{M}_{P,R}^{elliptic}$ is the Mueller matrix of a general normalized elliptical polarizer with variable ellipticity angle ε and azimuth angle α as in Eq. 12 and $\mathbf{M}_R^{Iso}$ is the Mueller matrix of an isotropic reflector as in Eq. 13 with variable diattenuation ψ and retardation δ . The coefficients a and b are limited to vary between 0 and 1 with the constraint $a + b = 1$. The result of the regression can be seen in Fig. 2. The coefficients a and b in Figs 2(a) and 2(d) both show quite featureless variations except in the P_C -regime. Coefficient a shows values close to zero outside the P_C -regime whereas inside the P_C -regime values reach 0.9. Since the sum of a and b is unity, coefficient b is close to one outside the P_C -regime and 0.1 inside. This behavior in combination with the choice of matrices $\mathbf{M}_{P,R}^{elliptic}$ and $\mathbf{M}_R^{Iso}$ in Eq. 16 is consistent with previous results [11].

Fig. 2. Result of regression analysis of data from *C. aurata* using the ansatz in Eq. 16.

A.1. Outside the P_C -regime

Outside the P_C -regime the contribution from the elliptical polarizer to the optical properties is approximately zero and the values of ε and α in Figs. 2(b) and 2(c), respectively, are therefore of little relevance.

The values of ψ and δ of the isotropic reflector can be seen in Figs. 2(e) and 2(f), respectively. The value of ψ is a function of

wavelength and varies from 43° at near normal incidence to 0° at larger incidence angles and δ is constant at 180° .

When the values at near-normal incidence are put into Eq. 13 the isotropic reflector becomes a mirror as in Eq. 14, and for large incidence angles the same isotropic reflector is a linear polarizer described in Eq. 15.

A.2. Inside the P_C -regime

The optical properties inside the P_C -regime can be described by a sum of the elliptical polarizer and the retarder. Here the values of ψ and δ of the retarder show some small variations as seen in Figs. 2(e) and 2(f). Compared to the value outside P_C -regime, there is a small increase in ψ of approximately 10° for incidence angles above 35° in a very narrow band in the center of the P_C -regime. δ varies from 180° down to 160° for incidence angles below approximately 40° and it drops to 110° in the same narrow band in the center of the P_C -regime close to 55° .

The angle ε and the azimuthal rotation α of the polarizer both have a strong dependence on wavelength and oscillate rapidly if viewed at a fixed angle of incidence. ε (Fig. 2(b)) varies from approximately 130° to 150° with a tendency to lower values at smaller incidence angles. α (Fig. 2(c)) shows small variations between 30° and 60° throughout the P_C -regime. When these values are put into Eq. 12 and multiplied by its weight factor a , the matrix (Fig. 3) will be similar to that of a left-handed circular polarizer. However, all elements will have non-zero values inside the P_C -regime, particularly in rows 1 and 4 and in columns 1 and 4. This deviation from a pure circular polarizer is likely due to in-plane anisotropy in the sample under inspection.

Fig. 3. The first term of Eq. 16, $a(\lambda, \theta) \mathbf{M}_{P,R}^{elliptic}(\alpha, \varepsilon)$.

The values from Figs. 2(e) and 2(f) are put into Eq. 13 and multiplied by its weight factor b and the resulting matrix can be seen in Fig. 4. Here we see very small values inside the P_C -regime and outside it has the characteristics of a reflection from a dielectric material, i.e. Eq. 13 with $\delta = 180^\circ$.

When the matrices in Figs. 3 and 4 are added according to Eq. 16 the result is very close to the experimental matrix. The differences between the experimental data and $\mathbf{M}^{reg}$ after decomposition can be seen in Fig. 5. Given the complexity of the biological reflector the agreement is generally very good and the differences between the experimental data and the result of the regression is within ± 0.08 for all θ and λ . From Figs. 2(a) and 2(d) the conclusion can be drawn that outside the P_C -regime the polarizing properties for all λ and θ of *C. aurata* are completely described by an isotropic reflector with ψ according to Fig. 2(e) and $\delta = 180^\circ$. Inside the P_C -regime the situation is slightly more complex and the polarizing properties cannot be completely described by a single, easily identifiable Mueller matrix. Here the properties are described by a sum of an elliptical

Fig. 4. The second term of Eq. 16, $b(\lambda, \theta)M_R^{Iso}(\psi, \delta)$.

Fig. 5. The difference between the experimental Mueller matrix and M^{reg} as calculated by Eq. 16 for *C. aurata*.

polarizer and an isotropic reflector where the contribution from the reflector is only in the diagonal of the Mueller matrix and by a small amount. The main contribution comes from the polarizer where the values are such that it is close to a circular polarizer. However, the elements m_{12} , m_{13} , m_{21} , m_{24} , m_{31} , m_{34} , m_{42} and m_{43} are non-zero which indicates that some anisotropy in the surface-plane is involved. The sensitivity to such anisotropy depends on the spot size which will increase along the plane of incidence with larger incidence angles. This has been discussed in previous work [11] and the effect is seen in Fig. 5 as an increasing value in the matrix elements with increasing incidence angle.

B. Regression sum decomposition of Mueller matrices of *C. argenteola*

Mueller matrices of *C. argenteola* were recorded at incidence angles in the range 15° to 55° with a step size of 1° in the spectral range 245 nm to 1000 nm and are summarized in a contour plot in Fig. 6. The broadband reflector *C. argenteola* exhibits left-handed polarization features for small θ seen as the large blue regions in Mueller matrix elements m_{14} and m_{41} , and right-handed polarization features for large θ , seen as the small red regions close to 55° in the same elements. A notable feature in Fig. 6 is also the interference oscillations which are seen in most elements as rapid variations in wavelength dependence. We here present only rudimentary trials on regression decomposition of *C. argenteola* using the same ansatz as was used for *C. aurata* (i.e. Eq. 16). The resulting parameters are presented in Fig. 7. However, the fit is not satisfactory as the Frobenius norm using Eq. 4 has values up to 0.3 in some regions for several of the elements, and up to 0.5 in element m_{44} . We also find that a Cloude decomposition results in three (or more depending on the incidence angle; see our previous work [11]) non-zero eigen-

Fig. 6. A contour plot of the experimentally determined Mueller matrix of *C. argenteola*.

Fig. 7. Result of regression analysis of data from *C. argenteola* using the ansatz in Eq. 16.

values for *C. argenteola* compared to only two for *C. aurata* as seen in Fig. 8. This means that more than two matrices should be included in a regression analysis and not only two as was used to derive the parameters in Fig. 7. One reason for the larger complexity in the analysis of *C. argenteola* data is that the pronounced interference patterns seen both in primary data in Fig. 6 and in eigenvalues in Fig. 8 shows that its cuticle is more transparent compared to that of *C. aurata*. The probe depth is therefore larger and structures deeper inside the cuticle are probed. In a broadband reflector like *C. argenteola* the Mueller matrix then becomes more complex as structures have different pitches at different depths. Similar effects has been observed and analyzed in the beetle *Cotinis mutabilis* [19]. Work is in progress to determine

Fig. 8. The eigenvalues from a Cloude decomposition of (a) *C. argenteola* and (b) *C. aurata*, both at 45° incidence angle.

additional matrices suitable for the regression decomposition of *C. argenteola* data.

C. Final remarks

We have presented a regression-based decomposition analysis of Mueller matrices measured as a function of wavelength and incidence angle. A sum of an elliptical polarizer and an isotropic reflector has been fitted to these experimental Mueller matrices. In the case of *C. aurata* this works well. In the case of *C. argenteola*, however, the ansatz presented in Eq. 16 does not work which is explained by the fact that a Cloude decomposition reveals that additional matrices are needed in the ansatz and further investigations will be performed.

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