Tutorial sesion:

\[
M = \begin{bmatrix}
  m_{00} & m_{01} & m_{02} & m_{03} \\
  m_{10} & m_{11} & m_{12} & m_{13} \\
  m_{20} & m_{21} & m_{22} & m_{23} \\
  m_{30} & m_{31} & m_{32} & m_{33}
\end{bmatrix}
\]

Mueller Matrix Ellipsometry

Oriol Arteaga
Dep. Applied Physics and Optics
University of Barcelona
Outline

- Historical introduction
- Basic concepts about Mueller matrices
- Mueller matrix ellipsometry instrumentation
- Further insights. Measurements and simulations
- Symmetries and asymmetries of the Mueller matrix. Relation to anisotropy.
- Applications and examples
- Concluding remarks
Historical introduction
Historical introduction

G G. Stokes
in 1852

Stokes Parameters

Francis Perrin
in 1942

90 years, almost forgotten!

Translation from the french:
Historical introduction

1852
G G. Stokes (1819-1903)

1929
Paul Soleillet (1902-1992)

1942
Francis Perrin (1901-1992)

1943
Hans Mueller (1900-1965)

Stokes Parameters

P. Soleillet, Ann. Phys. 12, 23 (1929)


H. Mueller, Report no. 2 of OSR project OEMsr-576 (1943)

K. Järrendahl and B. Kahr, Woollam newsletter, February 2011, pp. 8–9

SUR LES PARAMÈTRES CARACTÉRISANT LA POLARISATION PARTIELLE DE LA LUMIÈRE DANS LES PHÉNOMÈNES DE FLUORESCENCE

On en conclut que, $\gamma', \psi, \gamma, \sigma$ sont des fonctions linéaires et homogènes de $\psi, \alpha, \gamma, \sigma$ et que de même $I', M', C', S'$ sont des fonctions linéaires et homogènes de $I, M, C, S$.

\[ \begin{align*}
I' &= a_{11}I_1 + a_{12}M_1 + a_{13}C_1 + a_{14}S_1, \\
M' &= a_{21}I_1 + a_{22}M_1 + a_{23}C_1 + a_{24}S_1, \\
C' &= a_{31}I_1 + a_{32}M_1 + a_{33}C_1 + a_{34}S_1, \\
S' &= a_{41}I_1 + a_{42}M_1 + a_{43}C_1 + a_{44}S_1. 
\end{align*} \] (33)

The writer knows of no textbook in which these mathematical methods are discussed. Certain aspects of the method discussed here are given by F. Perrin, Journal of Chemical Physics 10, 415 (1942) where older references may be found.

9TH WORKSHOP ELLIPSOOMETRY @ UTWENTE
**Historical introduction**


Instrumental papers about the dual rotating compensator technique

"Generalized ellipsometry"  
"Mueller matrix ellipsometry"  
"Mueller matrix spectroscopic ellipsometry"

Web of Science Citation Reports

9TH WORKSHOP ELLIPSOMETRY @ UTWENTE
Basic concepts about Mueller matrices
Basic concepts about Mueller matrices

\[
S = \begin{bmatrix}
    I \\
    Q \\
    U \\
    V \\
\end{bmatrix} = \begin{bmatrix}
    S_0 \\
    S_1 \\
    S_2 \\
    S_3 \\
\end{bmatrix} = \begin{bmatrix}
    I \\
    I_x - I_y \\
    I_{45} - I_{135} \\
    I_+ - I_- \\
\end{bmatrix} = \begin{bmatrix}
    I \\
    Ip \cos(2\varphi) \cos(2\chi) \\
    Ip \cos(2\varphi) \sin(2\chi) \\
    Ip \sin(2\varphi) \\
\end{bmatrix}
\]

No depolarization: \( I = \sqrt{Q^2 + U^2 + V^2} \)

\[
S_{out} = MS_{in} \quad M = \begin{bmatrix}
    m_{00} & m_{01} & m_{02} & m_{03} \\
    m_{10} & m_{11} & m_{12} & m_{13} \\
    m_{20} & m_{21} & m_{22} & m_{23} \\
    m_{30} & m_{31} & m_{32} & m_{33} \\
\end{bmatrix}
\]

Phenomenological description of any scattering experiment

\( I \) Intensity  
\( p \) Degree of polarization  
\( \chi \) Azimuth  
\( \varphi \) Ellipticity
Basic concepts about Mueller matrices. No depolarization

A nondepolarizing Mueller matrix is called a **Mueller-Jones** matrix

Equivalence

\[ S_{out} = MS_{in} \quad \leftrightarrow \quad E_{out} = JE_{in} \]

\( M \) is 4x4 real  \( \leftrightarrow \)  \( J = \begin{bmatrix} r_{pp} & r_{ps} \\ r_{sp} & r_{ss} \end{bmatrix} \) is 2x2 complex matrix

A Jones or Mueller-Jones Jones depends on 6-7 parameters.

But note that the 16 elements of a Mueller-Jones matrix can be still all different!

Transformation

\[ M = T(J \otimes J^*)T^{-1} \]

\[ T = \begin{bmatrix} 1 & 0 & 0 & 1 \\ 1 & 0 & 0 & -1 \\ 0 & 1 & 1 & 0 \\ 0 & i & -i & 0 \end{bmatrix} \]
Basic concepts about Mueller matrices. No depolarization and isotropy

All modern ellipsometers measure elements of the Mueller matrix.

This is a common representation for isotropic media:

\[
\mathbf{J}_{\text{sample}} = \begin{bmatrix} r_p & 0 \\ 0 & r_s \end{bmatrix}
\]

\[
\mathbf{M}_{\text{sample}} = \begin{bmatrix} 1 & -N & 0 & 0 \\ -N & 1 & 0 & 0 \\ 0 & 0 & C & S \\ 0 & 0 & -S & C \end{bmatrix}
\]

\[
N = \cos(2\psi)
\]

\[
S = \sin(2\psi) \sin(\Delta)
\]

\[
C = \sin(2\psi) \cos(\Delta)
\]

\[
N^2 + S^2 + C^2 = 1
\]

\[
\rho = (\rho_{\text{real}} + i\rho_{\text{imag}}) = \frac{r_p}{r_s} = \tan(\psi) e^{i\Delta} = \frac{C + iS}{1 + N}
\]

Standard ellipsometry:
- Thickness measurements of thin films
- Optical functions of isotropic materials

This Mueller matrix depends only on 2 parameters.
Basic concepts about Mueller matrices. Depolarization

Depolarization is the reduction of the degree of polarization of light. Typically occurs when the emerging light is composed of several incoherent contributions.

![Diagram](image)

Quantification of the depolarization: Depolarization index (DI)

\[
DI = \sqrt{\sum_{ij} m_{ij}^2 - m_{00}^2} \quad 0 \leq DI \leq 1
\]

Reasons:
Sample exhibits spatial, temporal or frequency heterogeneity over the illuminated area


The DI of a Mueller-Jones matrix is 1

9TH WORKSHOP ELLIPSOLOGY @ UTWENTE
Mueller matrix ellipsometry instrumentation
Mueller matrix ellipsometry instrumentation

- Polarization state generator: PSG
- Polarization state analyzer: PSA

In a MM ellipsometer the PSG and PSA typically contain:
- A polarizer (P)
- A compensating or retarding element (C)

One exception: division-of-amplitude ellipsometers
**Mueller matrix ellipsometry instrumentation**

The compensating element is the main difference between different types of Mueller matrix ellipsometers.

<table>
<thead>
<tr>
<th>Rotating Retarders</th>
<th>Fixed Retardation</th>
<th>Waveplates are not very acromatic</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Changing azimuth</td>
<td>Fresnel rhoms are hard to rotate</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Mechanical rotation</td>
</tr>
</tbody>
</table>

| Liquid crystal cells | Variable Retardation (nematic LC) | Not transparent in the UV |
|                     | Changing azimuth (ferroelectric LC) | Temperature dependence |
|                     |                                 | No frequency domain analysis |

| Piezo-optic modulators (photoelastic modulators) | Variable Retardation | Two PEMs for each PSG or PSA |
|                                                | Fixed azimuth        | Too fast for imaging         |
| O. Arteaga et al. Appl. Optics 51, 28 6805-6817 (2012). | Fixed azimuth        | Two cells for each PSG or PSA |

| Electro-optic modulators (Pockels cells) | Variable Retardation | Two cells for each PSG or PSA |
|                                         | Fixed azimuth        | Small acceptance angle       |
|                                         | Fixed azimuth        | Too fast for imaging          |

**9TH WORKSHOP ELLIPSOMETRY @ UTWENTE**
**Mueller matrix ellipsometry instrumentation**

The PSA and PSG of Mueller matrix ellipsometers are no different from other Mueller matrix polarimetric approaches.

Mueller matrix microscope with two rotating compensators


Normal-incidence reflection imaging based on liquid crystals

Spectroscopic polarimeter based on four photoelastic modulators

Instrumentally wise no different from a MM ellipsometer. Lots of imaging applications in chemistry, medicine, biology, geology, etc.
Further insights
Measurement and simulations
Further insights. Measurement and simulations

A spectroscopic Mueller matrix ellipsometer produces this type of data:

Is this MM depolarizing?
If the depolarization is not significative we can find a proper non-depolarizing estimate.
Further insights. Measurement and simulations

Measurement: Mueller matrix  
Simulation usually generates a Jones/Mueller-Jones matrix (coherent model)

Objective: Finding a good nondepolarizing estimate (a Mueller-Jones matrix) for a experimental Mueller matrix

One option,

\[ \delta^2 = \sum_{i,j} (M_{ij} - M_{ij})^2 \rightarrow \min \]

Cloude estimate using the Cloude sum decomposition

\[ M = \lambda_0 M_{J0} + \lambda_1 M_{J1} + \lambda_2 M_{J2} + \lambda_3 M_{J3} \]

\[ M \approx \lambda_0 M_{J0} \]

S. R. Cloude, Optik 75, 26 (1986).  
Further insights. Measurement and simulations. Example

Experimental Mueller matrix

<p>| | | | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
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</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.6125</td>
<td>0.3377</td>
<td>-0.2466</td>
<td></td>
</tr>
<tr>
<td>-0.5766</td>
<td>0.7387</td>
<td>0.0096</td>
<td>0.4076</td>
<td></td>
</tr>
<tr>
<td>0.3778</td>
<td>-0.1007</td>
<td>0.6254</td>
<td>-0.3359</td>
<td></td>
</tr>
<tr>
<td>0.2536</td>
<td>-0.4551</td>
<td>0.3205</td>
<td>0.3956</td>
<td></td>
</tr>
</tbody>
</table>

\[
DI = \sqrt{\frac{\sum m_{ij}^2 - m_{00}^2}{3m_{00}}} = 0.963
\]

1. Calculate the Coherency matrix

\[
h_{00} = (m_{00} + m_{11} + m_{22} + m_{33})/4, \quad h_{01} = (m_{01} + m_{10} - im_{23} + im_{32})/4,
\]
\[
h_{02} = (m_{02} + m_{20} + im_{13} - im_{31})/4, \quad h_{03} = (m_{03} - im_{12} + im_{21} + m_{30})/4,
\]
\[
h_{10} = (m_{10} + m_{01} - im_{13} - im_{31})/4, \quad h_{11} = (m_{11} + m_{00} - im_{23} + im_{32})/4,
\]
\[
h_{12} = (m_{12} + m_{21} + im_{13} + im_{31})/4, \quad h_{13} = (m_{13} - im_{12} + im_{21} + m_{30})/4,
\]
\[
h_{20} = (m_{20} + m_{02} + im_{13} - im_{31})/4, \quad h_{21} = (m_{21} - im_{12} + im_{21} + m_{30})/4,
\]
\[
h_{22} = (m_{22} - im_{13} - im_{31} + m_{33})/4, \quad h_{23} = (m_{23} + m_{02} - im_{23} + im_{32})/4,
\]
\[
h_{30} = (m_{30} + m_{02} - im_{23} - im_{31})/4, \quad h_{31} = (m_{31} + m_{00} + im_{13} + im_{31})/4,
\]
\[
h_{32} = (m_{32} - m_{12} - im_{13} + im_{31})/4, \quad h_{33} = (m_{00} - m_{11} - m_{22} + m_{33})/4.
\]

Coherency matrix, H

\[
\begin{bmatrix}
0.7056 + 0i & -0.3318 - 0.2896i & 0.0468 + 0.0713i & 0.0032 - 0.0254i \\
-0.3318 + 0.2896i & 0.1601 + 0i & 0.038 - 0.015i & 0.0097 + 0.0043i \\
0.0468 - 0.0713i & 0.038 + 0.015i & 0.1288 + 0i & -0.022 + 0.0192i \\
0.0032 + 0.0254i & 0.0097 - 0.0043i & -0.022 - 0.0192i & 0.0055 + 0i
\end{bmatrix}
\]

2. Calculate the eigenvectors of H (is a hermitian matrix, so eigenvectors are real)

\[
\lambda_0 = 0.972 \quad \lambda_2 = 0.009
\]
\[
\lambda_1 = 0.022 \quad \lambda_3 = -0.003
\]

\[
M = \lambda_0 M_{J0} + \lambda_1 M_{J1} + \lambda_2 M_{J2} + \lambda_3 M_{J3}
\]

\[
M \approx \lambda_0 M_{J0}
\]
Further insights. Measurement and simulations. Example

3. The eigenvector corresponding to $\lambda_0$ defines the Jones matrix corresponding to $\mathbf{M}_{\text{J}_0}$:

\[
\begin{align*}
\mathbf{r}_{pp} &= \mathbf{\Psi}_0 + \mathbf{\Psi}_1 \\
\mathbf{r}_{rs} &= \mathbf{\Psi}_2 - i\mathbf{\Psi}_3 \\
\mathbf{r}_{sp} &= \mathbf{\Psi}_2 + i\mathbf{\Psi}_3 \\
\mathbf{r}_{ss} &= \mathbf{\Psi}_0 - \mathbf{\Psi}_1
\end{align*}
\]

The eigenvector corresponding to $\lambda_0$ defines the Jones matrix corresponding to $\mathbf{M}_{\text{J}_0}$:

\[
\begin{pmatrix}
0.465 & -0.1959i \\
0.1786 & -0.2497i
\end{pmatrix}
\begin{pmatrix}
0.2436 & -0.2603i \\
1.173 & +0.1959i
\end{pmatrix}
\]

Initial Experimental Mueller matrix

Best nondepolarizing estimate

Suitable to compare with coherent models
Further insights. Measurement and simulations. Expressing non-depolarizing data

\[ J = \begin{bmatrix} r_{pp} & r_{ps} \\ r_{sp} & r_{ss} \end{bmatrix} \]

\[
\begin{align*}
\rho &= \frac{r_{pp}}{r_{ss}} = \tan(\psi)e^{i\Delta} \\
\rho_{ps} &= \frac{r_{ps}}{r_{ss}} = \tan(\psi_{ps})e^{i\Delta_{ps}} \\
\rho_{sp} &= \frac{r_{sp}}{r_{ss}} = \tan(\psi_{sp})e^{i\Delta_{sp}}
\end{align*}
\]

1. \( \rho = 0.359 - 0.227i \)
   \( \rho_{ps} = 0.166 - 0.250i \)
   \( \rho_{sp} = 0.114 - 0.231i \)

2. \( \psi = 23.0^\circ \quad \Delta = -32.3^\circ \)
   \( \psi_{ps} = 16.7^\circ \quad \Delta_{ps} = -56.4^\circ \)
   \( \psi_{sp} = 14.5^\circ \quad \Delta_{sp} = -63.9^\circ \)

3. \( CD = 0.012 \quad CB = 0.082 \)
   \( LD = 0.885 \quad LB = 0.510 \)
   \( LD' = -0.544 \quad LB' = 0.635 \)

This notation is very suitable for normal-incidence transmission and reflection data:
CD: circular dichroism/diatt.
CB: circular birefringence/retard.
LD: horiz. linear dichroism/diatt.
... etc

Mueller matrix symmetries and anisotropy
**Mueller matrix symmetries and anisotropy**

In the **isotropic** case

\[
\mathbf{M} = \begin{bmatrix}
1 & m_{01} & 0 & 0 \\
0 & 1 & 0 & 0 \\
0 & m_{01} & m_{22} & m_{23} \star \\
0 & 0 & -m_{23} \star & m_{22}
\end{bmatrix}
\]

But this symmetry also applies to some situations with anisotropy!

The MM elements with an asteriks vanish in absence of absorption and J is real (assuming semi-infinite substrate as a sample).

---

**Uniaxial**

**Biaxial (orthorombic)**

Arrows are O. A.

**Biaxial (monoclinic)**

Arrow is P. A.
Mueller matrix symmetries and anisotropy

\[
M = \begin{bmatrix}
1 & m_{01} & m_{02} & m_{03}^* \\
 m_{01} & m_{11} & m_{12} & m_{13}^* \\
- m_{02} & - m_{12} & m_{22} & m_{23}^* \\
 m_{03}^* & m_{13}^* & - m_{23}^* & m_{33}
\end{bmatrix}
\]

\[
J = \begin{bmatrix}
 r_{pp} & r_{ps} \\
- r_{ps} & r_{ss}
\end{bmatrix}
\]

The MM elements with an asterisk vanish in absence of absorption and \( J \) is real (assuming semi-infinite substrate as a sample)

Uniaxial

Arrows are O. A.

Biaxial (orthorombic)

Biaxial (monoclinic)

Arrow is P. A.
Mueller matrix symmetries and anisotropy

\[
M = \begin{bmatrix}
1 & m_{01} & m_{02} & m_{03}^* \\
m_{01} & m_{11} & m_{12} & m_{13}^* \\
m_{02} & m_{12} & m_{22} & m_{23}^* \\
-m_{03}^* & -m_{13}^* & -m_{23}^* & m_{33}
\end{bmatrix}
\]

\[
J = \begin{bmatrix}
r_{pp} & r_{ps} \\
r_{ps} & r_{ss}
\end{bmatrix}
\]

The MM elements with an asterisks vanish in absence of absorption and J is real (assuming semi-infinite substrate as a sample)

Uniaxial
Arrow is O. A.

Biaxial (orthorombic)
Arrows are O. A.

Biaxial (monoclinic)
Arrow is P. A.
Mueller matrix symmetries and anisotropy

\[ M = \begin{bmatrix} 1 & m_{01} & m_{02}^* & m_{03} \\ m_{01} & m_{11} & m_{12}^* & m_{13} \\ -m_{02}^* & -m_{12}^* & m_{22} & m_{23} \\ m_{03} & m_{13} & -m_{23}^* & m_{33} \end{bmatrix} \]

\[ J = \begin{bmatrix} r_{pp} & r_{ps} \\ -r_{ps} & r_{ss} \end{bmatrix} \]

The MM elements with an asteriks vanish in absence of absorption and \( J \) is imaginary (assuming semi-infinite substrate as a sample).

Bi-isotropic media
Applications and examples
Applications and examples. A general idea about anisotropy

Intrinsic anisotropy vs structural/form anisotropy

\[ \mathbf{M} = \begin{pmatrix} m_{00} & m_{01} & m_{02} & m_{03} \\ m_{10} & m_{11} & m_{12} & m_{13} \\ m_{20} & m_{21} & m_{22} & m_{23} \\ m_{30} & m_{31} & m_{32} & m_{33} \end{pmatrix} \]

Expect small values of these elements for intrinsic anisotropy

E.g. Reflection on a calcite substrate

\[ \begin{align*}
\text{AOI} 65^\circ \\
\varepsilon_o &= 2.749 \\
\varepsilon_e &= 2.208
\end{align*} \]

\[
\begin{array}{cccc}
1 & -0.8864 & -0.08172 & 0 \\
-0.8864 & 0.9877 & 0.1331 & 0 \\
0.08172 & -0.1331 & 0.4434 & 0 \\
0 & 0 & 0 & 0.4557
\end{array} \]

\[
\begin{array}{cccc}
1 & -0.9504 & 0.1038 & 0 \\
-0.9504 & 0.9917 & -0.07624 & 0 \\
0.1038 & -0.07624 & 0.3016 & 0 \\
0 & 0 & 0 & 0.2933
\end{array} \]
Applications. Dielectric tensor of crystals

Measure the complex dielectric function (DF) tensor above and below the band edge

\[ D_i = \varepsilon_{ij}(\lambda)E_j \]

The dielectric tensor is symmetric

\[ \varepsilon = \begin{bmatrix} \varepsilon_{11} & \varepsilon_{12} & \varepsilon_{13} \\ \varepsilon_{12} & \varepsilon_{22} & \varepsilon_{23} \\ \varepsilon_{13} & \varepsilon_{23} & \varepsilon_{33} \end{bmatrix} \]

\[ \varepsilon' = A\varepsilon A^T \]

A magnetic field breaks the symmetry. E.g. MOKE

The principal values of the tensor correspond to crystal symmetry directions for isotropic, uniaxial and orthorhombic materials

Berreman’s 4x4 complex formalism is used to calculate \( \rho, \rho_{sp} \) and \( \rho_{ps} \) from elements of \( \varepsilon \) and the angle of incidence (a fully analytical treatment is sometimes possible).
Applications. Dielectric tensor of crystals

General scheme of the approach:

EXPERIMENT
MUELLER MATRIX

THEORY
CONSTITUTIVE TENSORS $\mathbf{\varepsilon}$

JONES MATRIX

e.g. Cloude’s

MAXWELL EQUATIONS
(Berreman formulation)

MATRIX
MULTIPLICATION of complex 4x4 matrices
(forward and backward propagating waves)
Applications. Dielectric tensor of crystals

Rutile (Uniaxial)

Applications. Dielectric tensor of crystals

Rutile (Uniaxial)
Applications. Dielectric tensor of crystals

Monoclinic CdWO₄

\[ \varepsilon = \begin{bmatrix} \varepsilon_{11} & \varepsilon_{12} & 0 \\ \varepsilon_{12} & \varepsilon_{22} & 0 \\ 0 & 0 & \varepsilon_{33} \end{bmatrix} \]

Note that a non-diagonal dielectric tensor can lead to a block diagonal MM

Mueller matrix Scatterometry

Measurements in periodic grating-like structures
Analysis of the Zeroth-order diffracted light (specular reflection).

\[ n_2 \sin(\theta_m) = n_i \sin(\theta_i) - m \frac{\lambda}{P} \]

Qualitative understanding of the measurements is possible attending to MM symmetries, and Rayleigh anomalies of higher orders. Energy distribution to higher orders

Expect the same symmetries as for a sample with optic axis lying in the plane of the sample

Rigorous-coupled wave analysis (RCWA). Field components expanded into Fourier series
Mueller matrix Scatterometry

S. Liu, et al., Development of a broadband Mueller matrix ellipsometer as a powerful tool for nanostructure metrology, Thin Solid Films, in press
Mueller matrix Scatterometry

S. Liu, et al., Development of a broadband Mueller matrix ellipsometer as a powerful tool for nanostructure metrology, Thin Solid Films, in press
Helicoidal Bragg reflectors

Cholesteric liquid crystal

\[ \lambda_0 = n_{\text{Average}} \cdot p \]
\[ \lambda_{\text{peak}} = \lambda_0 \cos \theta_i \]
\[ \Delta \lambda = p \Delta n \]
\[ \Delta n = (n_{\text{slow}} - n_{\text{fast}}) \]

Classical approximate formulas for Bragg reflection from cholesteric liquid crystals

Structural chirality, no real magneto-electric origin.

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Helicoidal Bragg reflectors

\[
M = \begin{bmatrix}
1 & m_{01} & m_{02}^* & m_{03} \\
m_{01} & m_{11} & m_{12}^* & m_{13} \\
-m_{02}^* & -m_{12}^* & m_{22} & m_{23} \\
m_{03} & m_{13} & -m_{23}^* & m_{33}
\end{bmatrix}
\]

\[P \approx 282 \text{nm}\]
Helicoidal Bragg reflectors

Fig. 5. Mueller-matrix spectra (solid curves) at $\theta = 25^\circ$, $40^\circ$, $60^\circ$ and $75^\circ$ measured on a green-colored C. aurata. The dashed curves show model-generated spectra using the model in Fig. 3. Only data for $\theta = 25^\circ$, $40^\circ$ and $60^\circ$ are used in the regression analysis whereas the model data for $\theta = 75^\circ$ are predicted.


$$
M = \begin{bmatrix}
1 & m_{01} & m_{02} & m_{03} \\
m_{01} & m_{11} & m_{12} & m_{13} \\
-m_{02} & -m_{12} & m_{22} & m_{23} \\
m_{03} & m_{13} & -m_{23} & m_{33}
\end{bmatrix}
$$
**Plasmonic nanostructures**

Typically, measurements are made on 2D periodic nanostructures with characteristic dimensions comparable or smaller than the wavelength of light.

![Nanostructures Image]

\[ D_i = \varepsilon_{ij}(\omega,\mathbf{k})E_j \sim \frac{a}{\lambda} \]

Big spatial dispersion effects

The electric polarization at a certain position is determined not only by the electric field at that position, but also by the fields at its neighbors.

\[ d = 250 \text{ nm} \]
\[ a = 530 \text{ nm} \]

... And the neighbors change depending on how we orient the sample in the ellipsometer....
Plasmonic nanostructures

Projections of a square lattice

In transmission a square lattice is isotropic.

This what the photons of the ellipsometer will “see”.

RECTANGULAR

RHOMBIC

OBLIQUE
Plasmonic nanostructures

Even a highly symmetric plasmonic nanostructure must be described by a non-diagonal, asymmetric Jones matrix whenever the plane of incidence does not coincide with a mirror line.

Concluding remarks

I have a isotropic sample, should I study with Mueller matrix ellipsometry?
Yes, it never hurts. Having access to the whole MM also helps to verify the alignment of the sample.

I have an anisotropic sample, can I study it with standard ellipsometry?
Most likely yes, although Mueller matrix ellipsometry is arguably better suited. Reorientations are going to be necessary. Will fail if there is some significant depolarization.

I have an optically active sample, can I study it with standard ellipsometry? And with Mueller ellipsometry?
Not with standard ellipsometry. Possibly with Mueller ellipsometry. But be aware! In reflection you will be NOT measuring directly optical rotatory dispersion or circular dichroism.
Summary of ideas to take home

- When possible (small depo) convert a experimental MM in a Mueller-Jones matrix or a Jones matrix and work from that

- Symmetries or assymetries of a MM give information about the orientation the sample and/or the crystallographic system

- For intrinsic anisotropy the non-diagonal Jones elements are small and the Mueller matrix is close to a NSC matrix. If they are large suspect about structure-induced anisotropy or misalignment of the sample

- Mueller matrix ellipsometry has the same applications as standard ellipsometry, plus it handles accurately anisotropy and depolarization. Important for crystals, nanotechnology, scatterometry, etc
Some further references

MM symmetries

DF of low symmetry crystals

MMs at normal incidence transmission

MMs at normal-incidence reflection

MM scatterometry
- S. Liu, et al., Development of a broadband Mueller matrix ellipsometer as a powerful tool for nanostructure metrology, Thin Solid Films, in press

MM and metamaterials
Acknowledgments

R. Ossikovski (EP), A. Canillas (UB), S. Nichols (NYU), G. E. Jellison (ORNL)

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