Tutorial: Mueller matrix ellipsometry: a hands-on understanding of the matrix elements

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Preface

The goal of this tutorial is to explain ellipsometry is to get an intuitive understanding of **Mueller matrix ellipsometry without exiting the Stokes-Mueller formalism**

In this presentation:

NO JONES MATRICES

NO PSI DELTA

NO COMPLEX NUMBERS

Outline

Introduction. MM types

Mueller matrices

$$\mathbf{S} = \begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix} = \begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix} = \begin{bmatrix} I \\ I_x - I_y \\ I_{45} - I_{135} \\ I_+ - I_- \end{bmatrix}$$

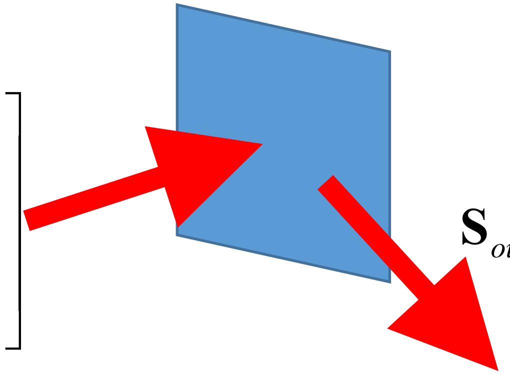
$$\mathbf{S}_{out} = \mathbf{M}\mathbf{S}_{in}$$

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

Phenomenological description of any linear scattering experiment

- Paul Soleillet (1902-1992)
- Francis Perrin (1901-1992)
- Hans Mueller (1900-1965)

$$\mathbf{S}_{in} = \begin{bmatrix} I_0 \\ Q_0 \\ U_0 \\ V_0 \end{bmatrix}$$



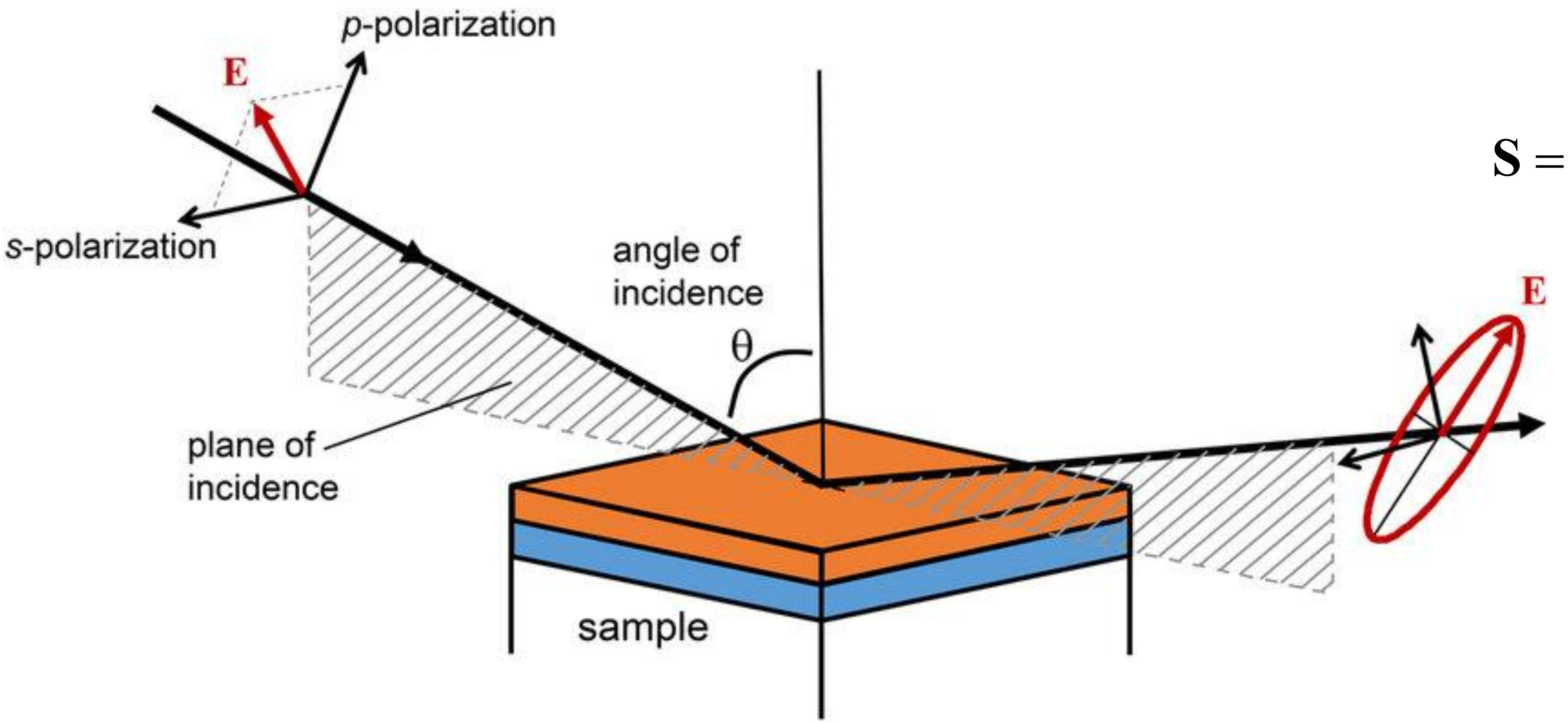
$$\mathbf{S}_{out} = \begin{bmatrix} I_1 \\ Q_1 \\ U_1 \\ V_1 \end{bmatrix}$$

$$\mathbf{S}_{out} = \mathbf{M}\mathbf{S}_{in}$$

Introduction. MM types

In Mueller matrix ellipsometry involving samples, we almost always choose our coordinate system such that:

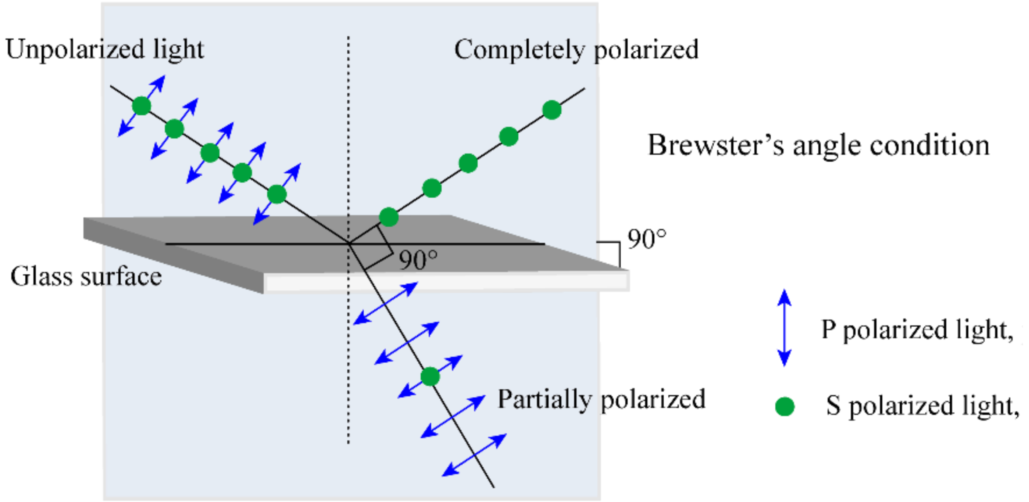
- ***x* axis → *p* direction**
- ***y* axis → *s* direction** **(for ellipsometers where the sample sits horizontally, this may seem counterintuitive!)**

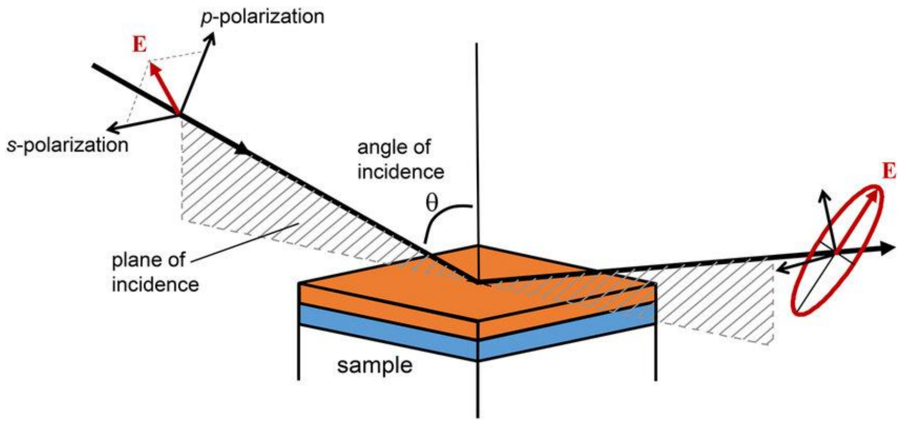


$$\mathbf{S} = \begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix} = \begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix} = \begin{bmatrix} I \\ I_p - I_s \\ I_{45} - I_{135} \\ I_+ - I_- \end{bmatrix}$$

Introduction. MM types

E.g. at Brewster angle





$$\mathbf{M} = \begin{bmatrix} 1 & -1 & 0 & 0 \\ -1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}$$

Introduction. MM types

3 types of elemental Mueller matrices

Anisotropy in s/p

$$\mathbf{M}_L = \begin{bmatrix} 1 & m_{01} & 0 & 0 \\ m_{01} & 1 & 0 & 0 \\ 0 & 0 & m_{22} & m_{23} \\ 0 & 0 & -m_{23} & m_{22} \end{bmatrix}$$

If there is no depolarization $m_{01}{}^2 + m_{22}{}^2 + m_{23}{}^2 = 1$

$$\mathbf{M}_L \begin{bmatrix} |S_1| \\ S_1 \\ 0 \\ 0 \end{bmatrix} = \begin{bmatrix} |S_1'| \\ S_1' \\ 0 \\ 0 \end{bmatrix}$$

s and *p* polarizations are eigenpolarizations of the Mueller matrix

Introduction. MM types

3 types of elemental Mueller matrices

Anisotropy in 45/-45

$$\mathbf{M}_{L'} = \begin{bmatrix} 1 & 0 & m_{02} & 0 \\ 0 & m_{11} & 0 & m_{13} \\ m_{02} & 0 & 1 & 0 \\ 0 & -m_{13} & 0 & m_{33} \end{bmatrix}$$

If there is no depolarization $m_{02}{}^2 + m_{11}{}^2 + m_{13}{}^2 = 1$

$$\mathbf{M}_{L'} \begin{bmatrix} |S_2| \\ 0 \\ S_2 \\ 0 \end{bmatrix} = \begin{bmatrix} |S_2'| \\ 0 \\ S_2' \\ 0 \end{bmatrix}$$

+45 and *-45* polarizations are eigenpolarizations of the Mueller matrix

Introduction. MM types

3 types of elemental Mueller matrices

Anisotropy in R/L Circular

$$\mathbf{M}_{L'} = \begin{bmatrix} 1 & 0 & 0 & m_{03} \\ 0 & m_{11} & m_{12} & 0 \\ 0 & -m_{12} & m_{11} & 0 \\ m_{03} & 0 & 0 & 1 \end{bmatrix}$$

If there is no depolarization $m_{03}{}^2 + m_{11}{}^2 + m_{12}{}^2 = 1$

$$\mathbf{M}_{L'} \begin{bmatrix} |S_3| \\ 0 \\ 0 \\ S_3 \end{bmatrix} = \begin{bmatrix} |S_3'| \\ 0 \\ 0 \\ S_3' \end{bmatrix}$$

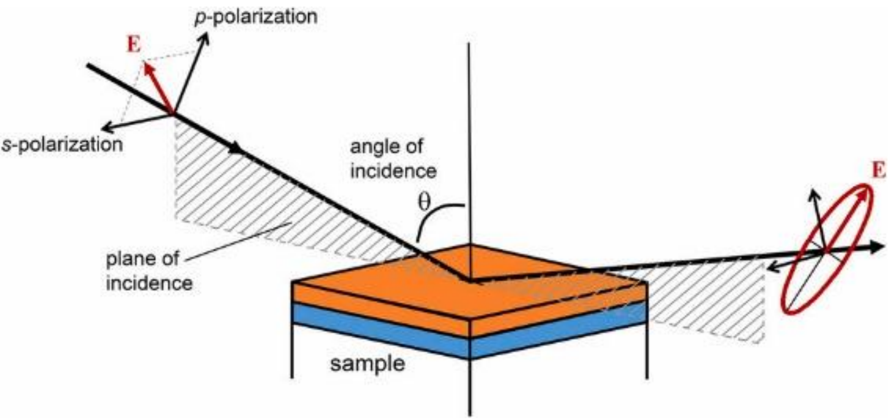
+ and - *circular* polarizations are eigenpolarizations of the Mueller matrix

Introduction. MM types

3 types of elemental Mueller matrices

The specular reflection ellipsometric geometry leads to MMs that usually have this form (or close to this)

$$\begin{bmatrix} 1 & m_{01} & 0 & 0 \\ m_{01} & 1 & 0 & 0 \\ 0 & 0 & m_{22} & m_{23} \\ 0 & 0 & m_{23} & m_{22} \end{bmatrix} \longleftrightarrow \begin{bmatrix} 1 & -N & 0 & 0 \\ -N & 1 & 0 & 0 \\ 0 & 0 & C & S \\ 0 & 0 & -S & C \end{bmatrix}$$

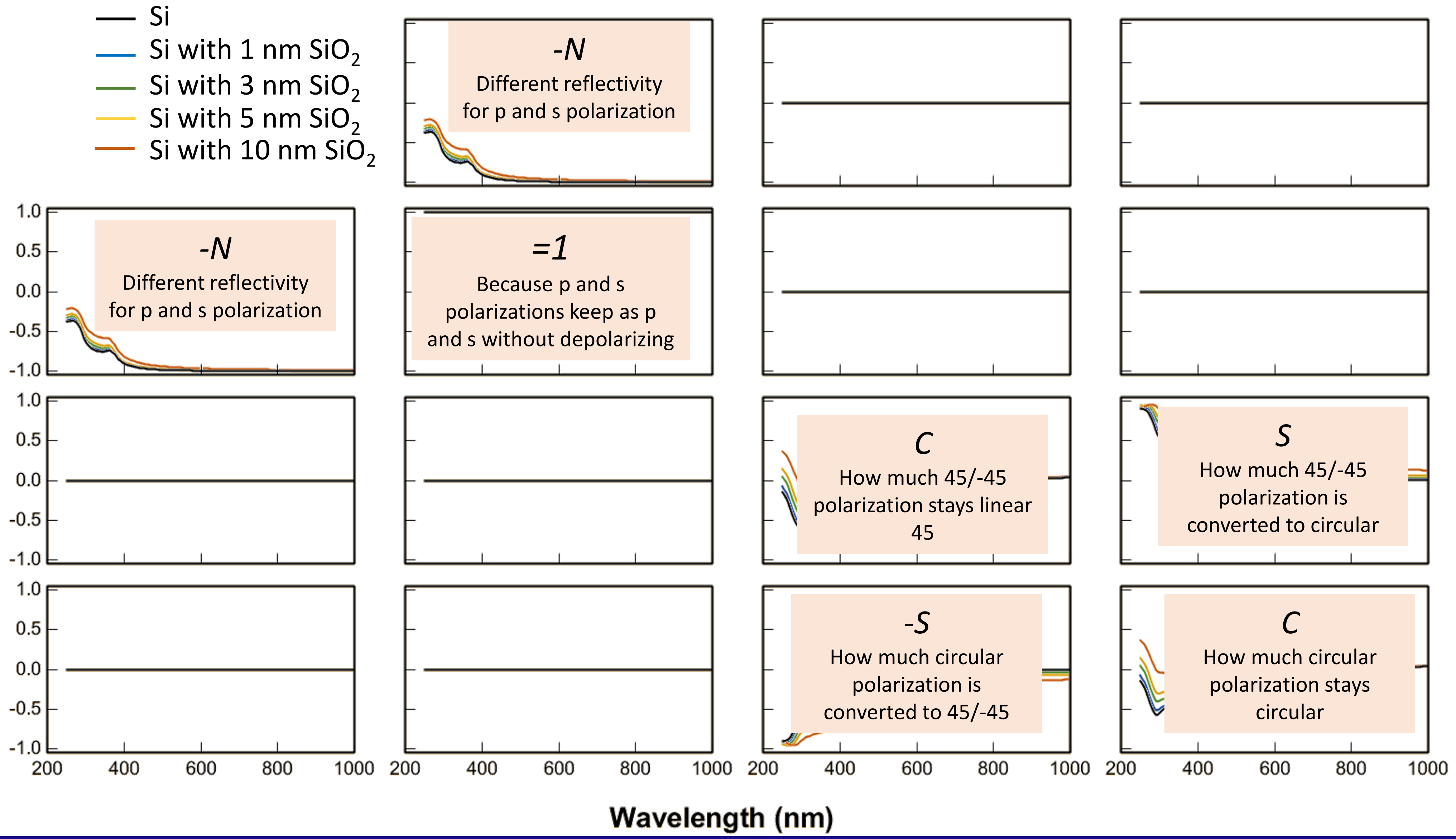


$$\begin{bmatrix} 1 & -N & 0 & 0 \\ -N & 1 & 0 & 0 \\ 0 & 0 & C & S \\ 0 & 0 & -S & C \end{bmatrix} \begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix} = \begin{bmatrix} S_0 - NS_1 \\ S_1 - NS_0 \\ CS_2 + SS_3 \\ CS_3 - SS_2 \end{bmatrix}$$

Diattenuation

Retardance

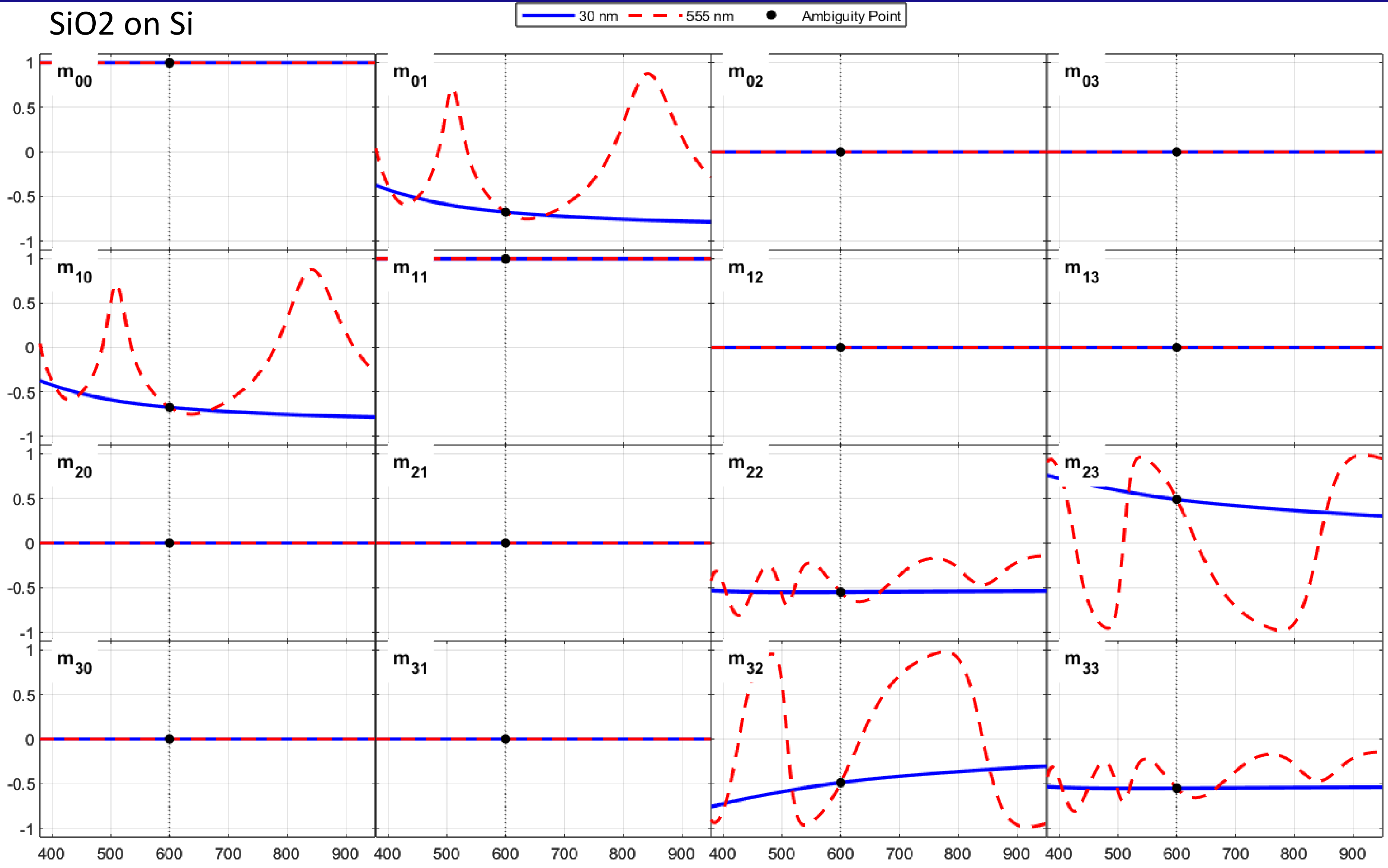
Introduction. MM types



Sensitivity to the overlayer thickness!!

Introduction. MM types

*One MM measurement is not enough => **spectroscopic***



Retardance information is experimentally accessed only through cyclic trigonometric functions, which map infinite possible phase delays onto a repeating range of -1 to +1

Outline

MM Symmetries

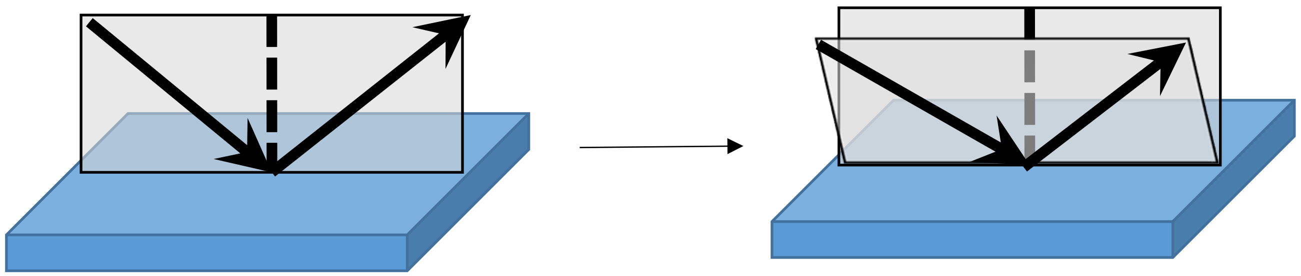
Ellipsometry MMs are not always block-diagonal

$$\begin{bmatrix} 1 & -N & 0 & 0 \\ -N & 1 & 0 & 0 \\ 0 & 0 & C & S \\ 0 & 0 & -S & C \end{bmatrix}$$

Block-diagonal=> ISOTROPIC (may be depolarizing or non-depolarizing)

Non-block-diagonal=> ANISOTROPIC (may be depolarizing or non-depolarizing)

Caution! Isotropic materials can lead to a substantially nonsymmetric Mueller matrix if the sample is **poorly aligned.**



Sample **misaligned** with respect to the plane of incidence

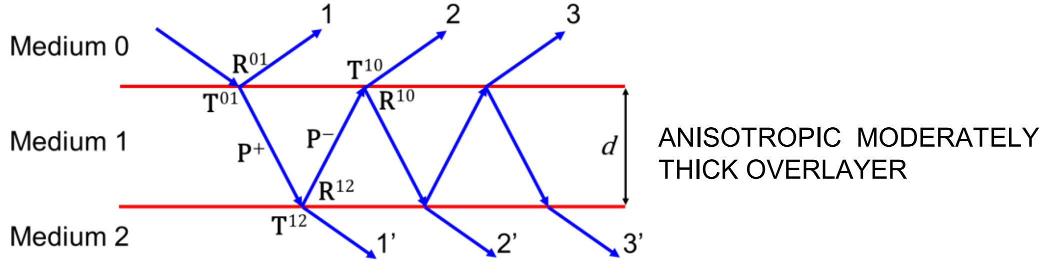
MM Symmetries

These elements are small (e.g.<0.2) even for materials with large ansisotropy

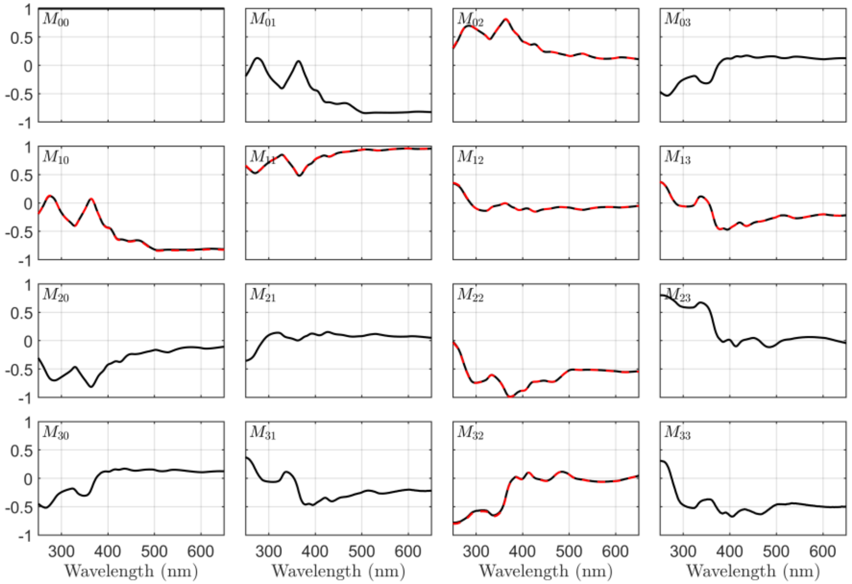
$$\mathbf{M} = \begin{bmatrix} m_{00} & m_{01} & m_{02} & m_{03} \\ m_{10} & m_{11} & m_{12} & m_{13} \\ m_{20} & m_{21} & m_{22} & m_{23} \\ m_{30} & m_{31} & m_{32} & m_{33} \end{bmatrix}$$

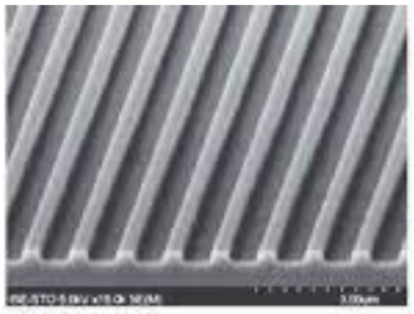
Exceptions

1. **Bulk** propagation contributes to the measurement



2. **Structural anisotropy (gratings, beetles)**





MM Symmetries

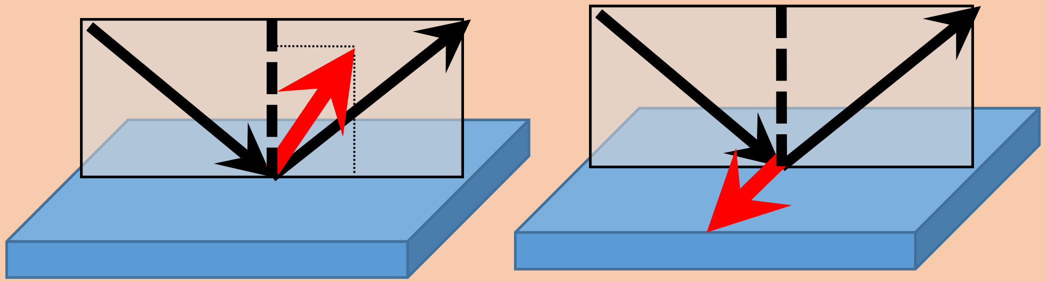
No crosspolarization (preservation of eigenpolarizations)

In the **isotropic** case and some anisotropy situations of high symmetry.

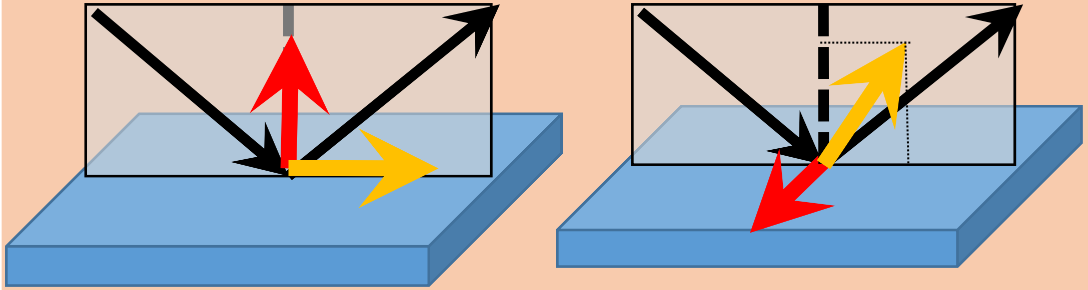
$$\mathbf{M}_L = \begin{bmatrix} 1 & m_{01} & 0 & 0 \\ m_{01} & 1 & 0 & 0 \\ 0 & 0 & m_{22} & m_{23} \\ 0 & 0 & -m_{23} & m_{22} \end{bmatrix}$$

This symmetry appears whenever the sample coincides with its own mirror image with respect to the incidence plane (i.e. the sample is mirror-symmetric with respect to this plane)

Uniaxial

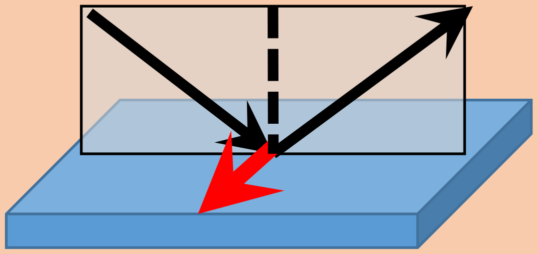


Biaxial (orthorombic)



Arrows are Principal axes (P.A)

Biaxial (monoclinic)



Arrow is P. A.

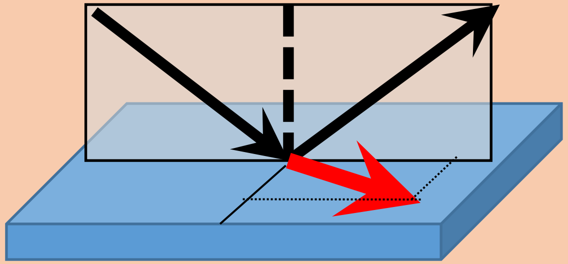
MM Symmetries

Anti-symmetric crosspolarization

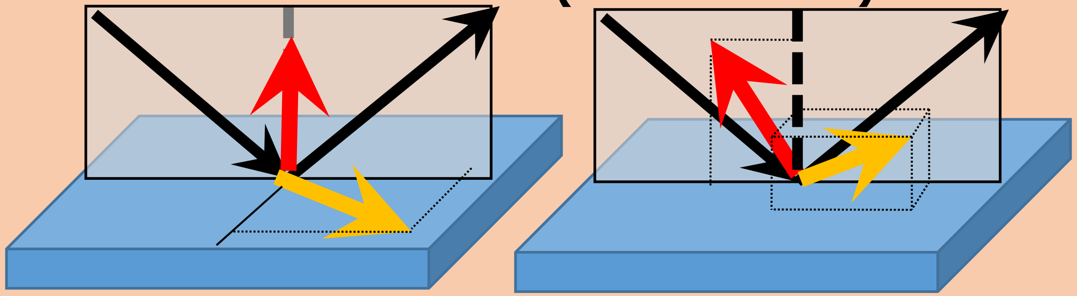
$$\mathbf{M} = \begin{bmatrix} 1 & m_{01} & m_{02} & m_{03} \\ m_{01} & m_{11} & m_{12} & m_{13} \\ -m_{02} & -m_{12} & m_{22} & m_{23} \\ m_{03} & m_{13} & -m_{23} & m_{33} \end{bmatrix}$$

A 180°-rotation of the sample about its normal brings it in its initial position

Uniaxial

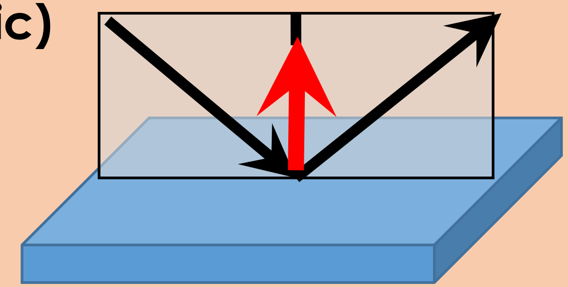


Biaxial (orthorombic)



Arrows are Principal axes (P.A)

Biaxial (monoclinic)



Arrow is P. A.

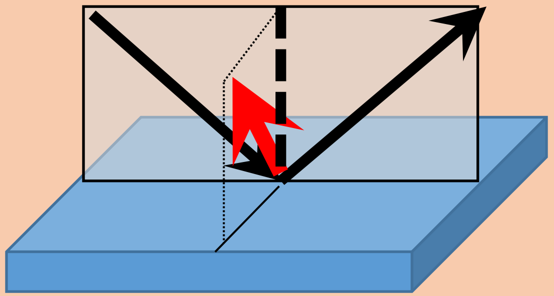
MM Symmetries

Symmetric crosspolarization

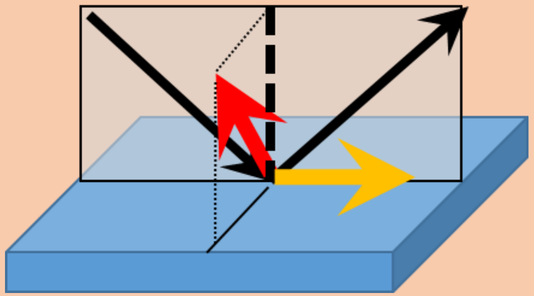
$$\mathbf{M} = \begin{bmatrix} 1 & m_{01} & m_{02} & m_{03} \\ m_{01} & m_{11} & m_{12} & m_{13} \\ m_{02} & m_{12} & m_{22} & m_{23} \\ -m_{03} & -m_{13} & -m_{23} & m_{33} \end{bmatrix}$$

The sample is mirror-symmetric with respect to the plane perpendicular to the incidence plane and containing the sample normal

Uniaxial

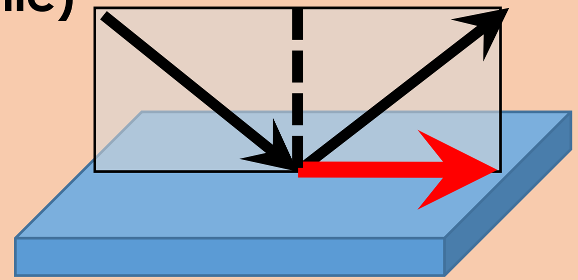


Biaxial (orthorombic)



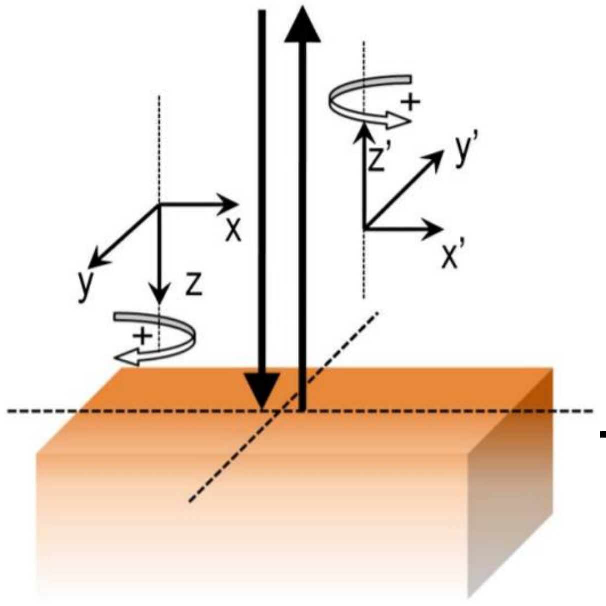
Arrows are Principal axes (P.A)

Biaxial (monoclinic)



Arrow is P. A.

MM Symmetries



When the **angle of incidence approaches zero** this is the most general possible symmetry

$$\begin{bmatrix} m_{00} & m_{01} & m_{02} & m_{03} \\ m_{01} & m_{11} & m_{12} & m_{13} \\ -m_{20} & -m_{12} & m_{22} & m_{23} \\ m_{03} & m_{13} & -m_{23} & m_{33} \end{bmatrix}$$

There is no contrast about the change of refractive index along the direction of propagation of light!

Measures

- Differential phase retardations
- Differential reflectivity

In this situation typically off-diagonal elements are very small and measurement is close to

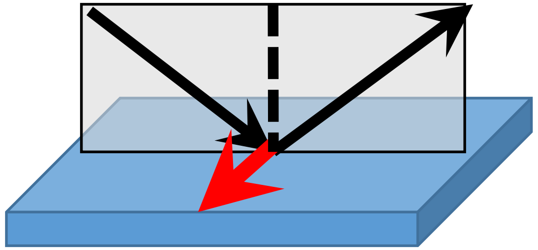
$$\sim \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & -1 \end{bmatrix}$$

(always reciprocal symmetry)

Similarly, at grazing incidence (AOI close to 90^{o})

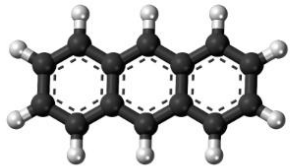
$$\sim \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix}$$

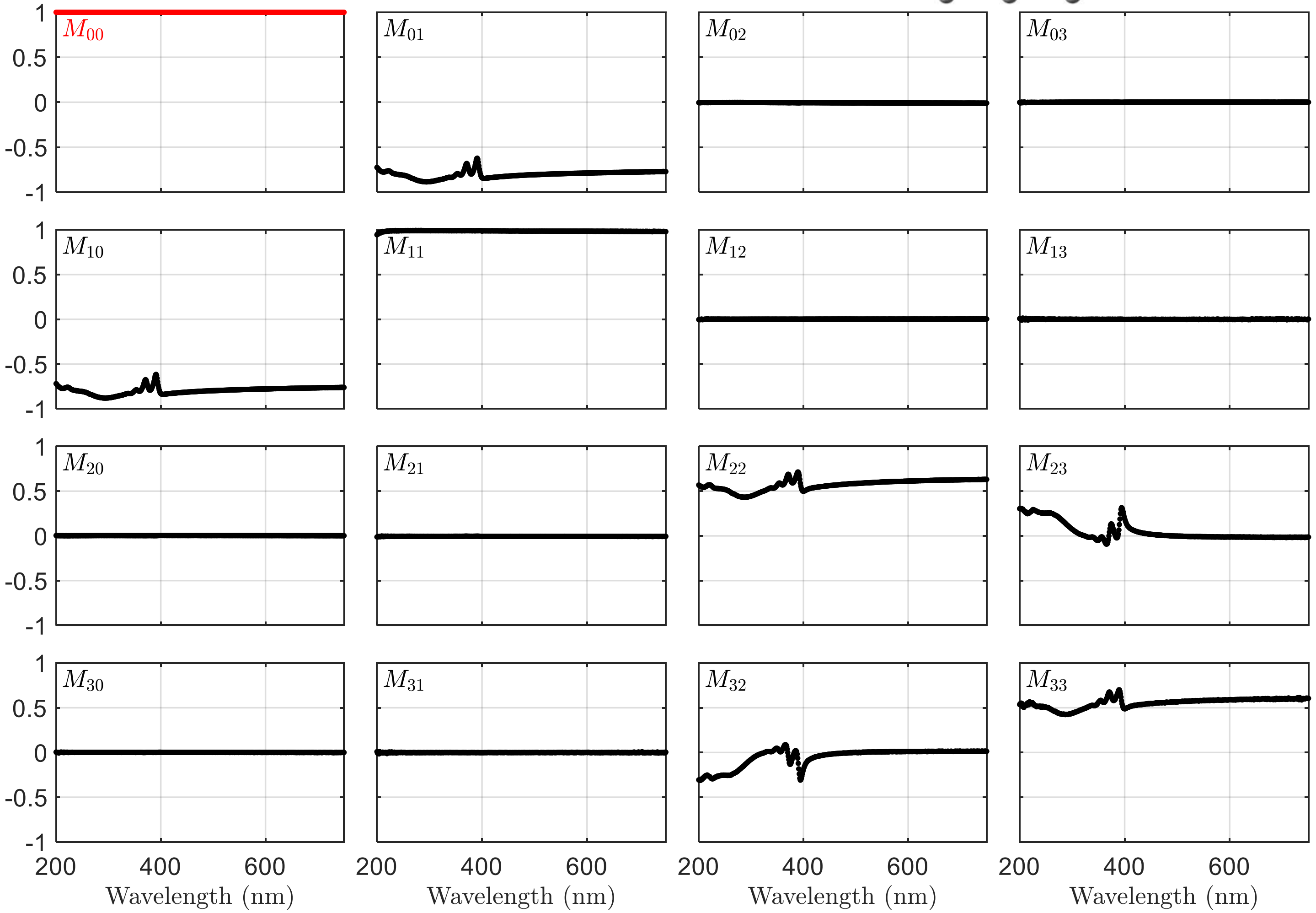
MM Symmetries



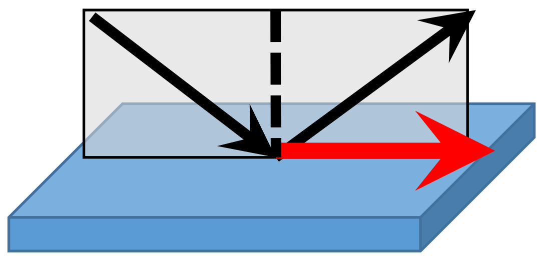
$$\mathbf{M}_L = \begin{bmatrix} 1 & m_{01} & 0 & 0 \\ m_{01} & 1 & 0 & 0 \\ 0 & 0 & m_{22} & m_{23} \\ 0 & 0 & -m_{23} & m_{22} \end{bmatrix}$$

Example: Anthracene crystal (monoclinic)



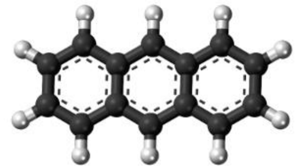


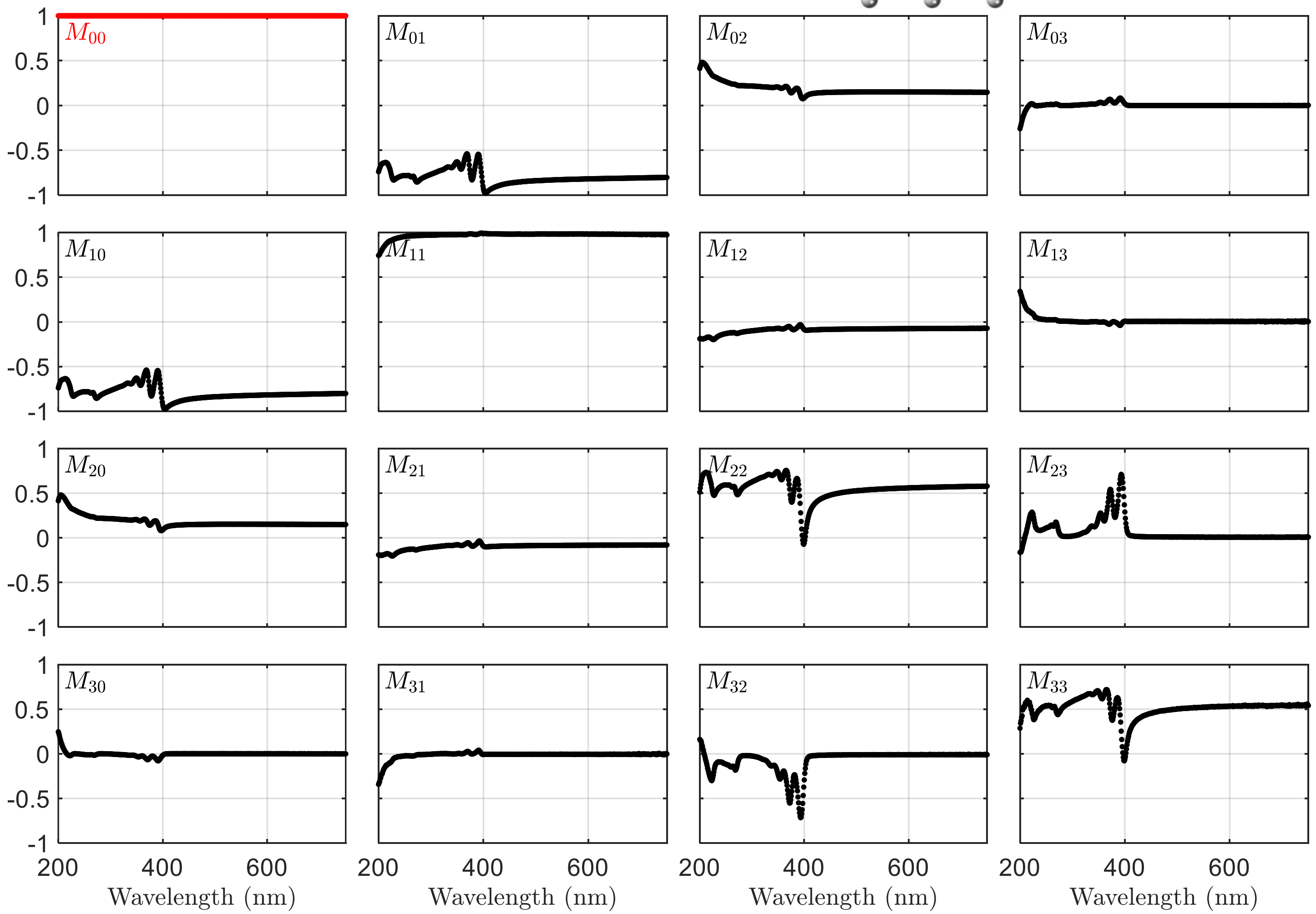
MM Symmetries



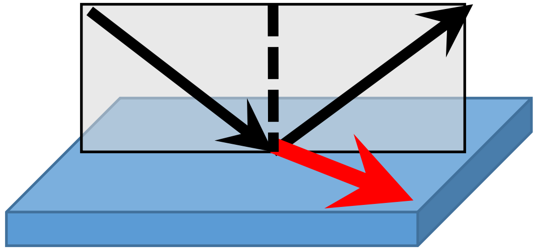
$$\mathbf{M} = \begin{bmatrix} 1 & m_{01} & m_{02} & m_{03} \\ m_{01} & m_{11} & m_{12} & m_{13} \\ m_{02} & m_{12} & m_{22} & m_{23} \\ -m_{03} & -m_{13} & -m_{23} & m_{33} \end{bmatrix}$$

Example Anthracene (monoclinic)



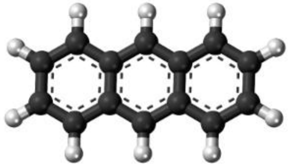


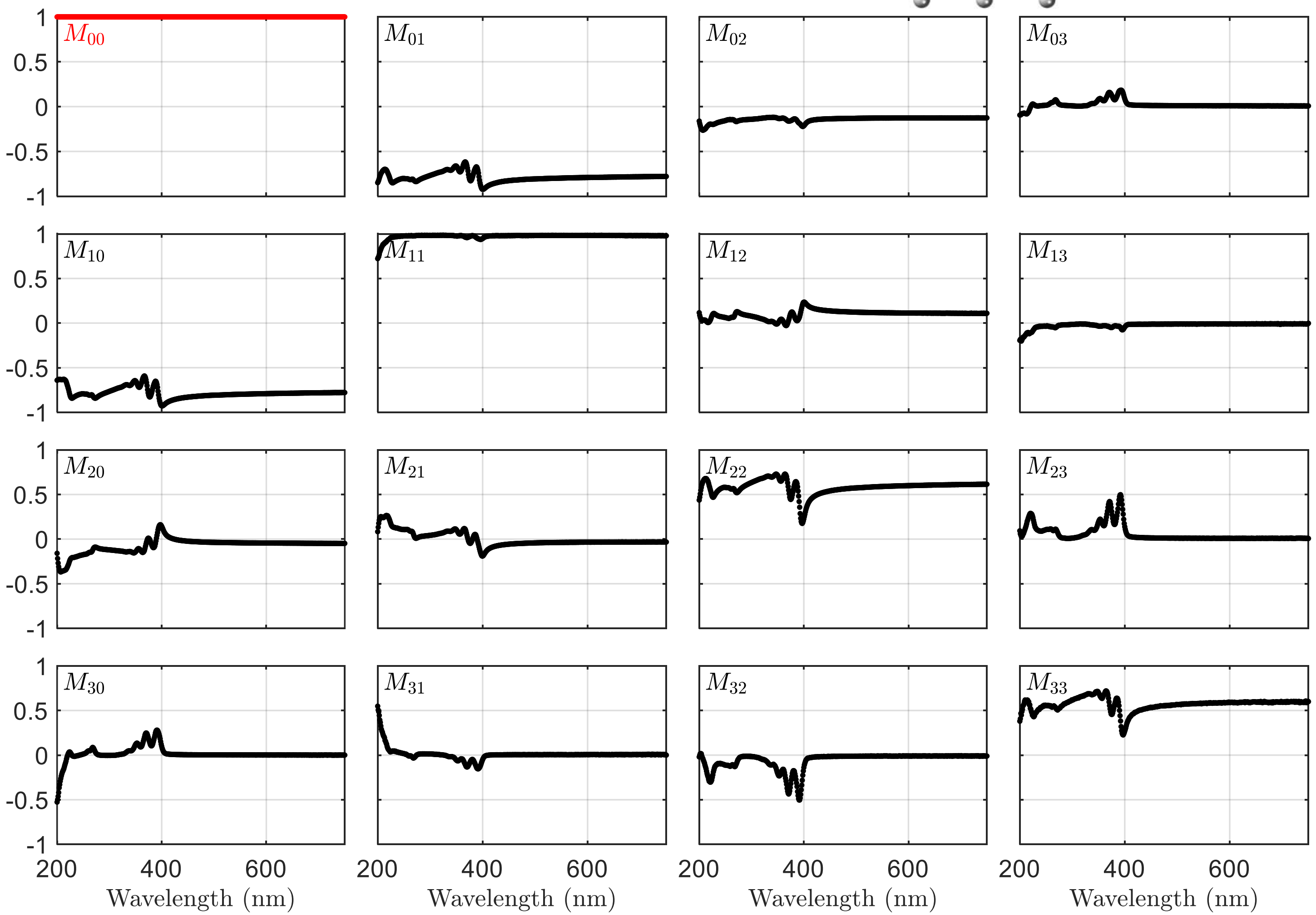
MM Symmetries



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

Example Antracene (monoclinic)

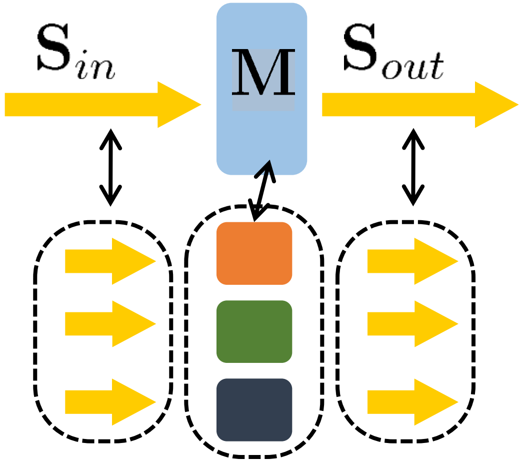


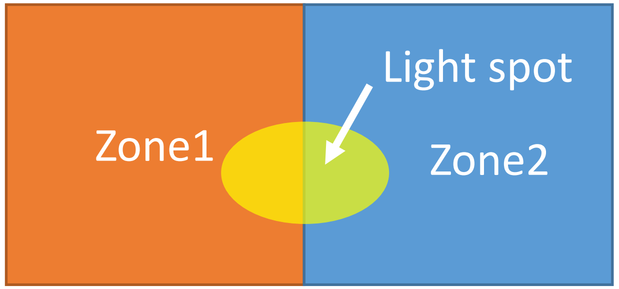


Outline

Depolarization

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





Incoherent sum of waves

↓

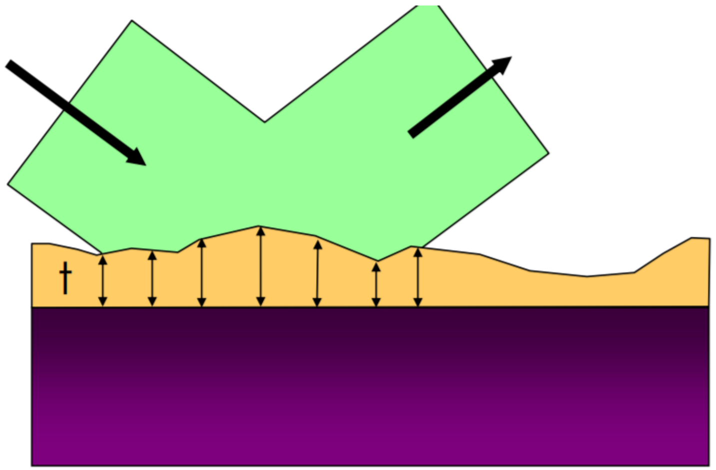
Depolarization

Incoherent sum: Mueller matrices: $\mathbf{M}=\mathbf{M}_1+\mathbf{M}_2$

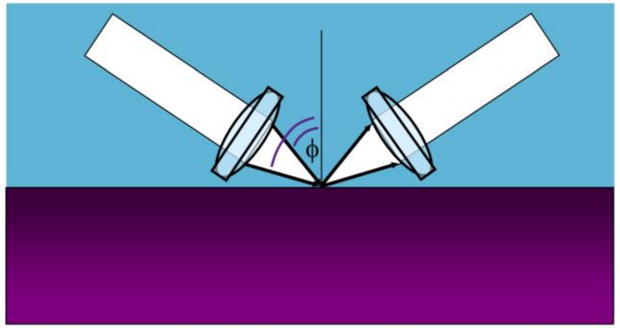
$$\frac{1}{2}\begin{bmatrix} 1 & -N_1 & 0 & 0 \\ -N_1 & 1 & 0 & 0 \\ 0 & 0 & C_1 & S_1 \\ 0 & 0 & -S_1 & C_1 \end{bmatrix} + \frac{1}{2}\begin{bmatrix} 1 & -N_2 & 0 & 0 \\ -N_2 & 1 & 0 & 0 \\ 0 & 0 & C_2 & S_2 \\ 0 & 0 & -S_2 & C_2 \end{bmatrix} = \begin{bmatrix} 1 & -(N_1+N_2)/2 & 0 & 0 \\ -(N_1+N_2)/2 & 1 & 0 & 0 \\ 0 & 0 & (C_1+C_2)/2 & (S_1+S_2)/2 \\ 0 & 0 & -(S_1+S_2)/2 & (C_1+C_2)/2 \end{bmatrix}$$

$$N_1^2+C_1^2+S_1^2=1 \qquad N_2^2+C_2^2+S_2^2=1 \qquad \frac{1}{4}[(N_1+N_2)^2+(C_1+C_2)^2+(S_1+S_2)^2]\leq 1$$

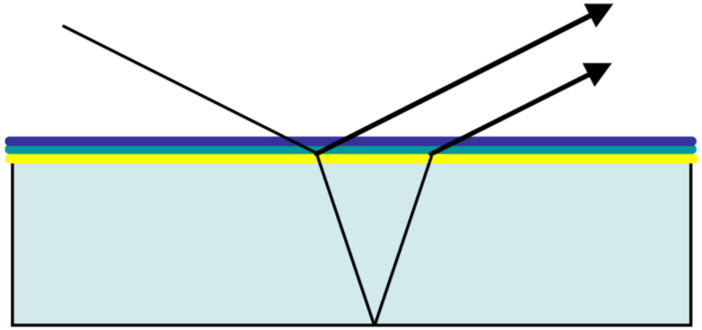
Depolarization



Thickness non uniformity



Angular spread



Backside reflections

* *Images from J. A. Woollam tutorial*

Parasitic light contamination

$$\frac{1}{2}\begin{bmatrix} 1 & -N_1 & 0 & 0 \\ -N_1 & 1 & 0 & 0 \\ 0 & 0 & C_1 & S_1 \\ 0 & 0 & -S_1 & C_1 \end{bmatrix} + \frac{1}{2}\begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix} = \begin{bmatrix} 1 & -N_1/2 & 0 & 0 \\ -N_1/2 & 0.5 & 0 & 0 \\ 0 & 0 & C_1/2 & S_1/2 \\ 0 & 0 & -S_1/2 & C_1/2 \end{bmatrix}$$

Depolarization

In anisotropic systems, depolarization will “usually” will NOT break the symmetries

A symmetric matrix
- Can be non-dep.
- Can be depolarizing

A non-symmetric matrix
- Can be non-dep.
- Can be depolarizing

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

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

Depolarization index

$$DI = \frac{\sqrt{\sum_{ij} m_{ij}^2 - m_{00}^2}}{\sqrt{3} m_{00}}$$

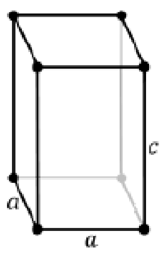
$$0 \leq DI \leq 1$$

Symmetric situations have the advantage that even depolarizing samples can be studied without full MM systems

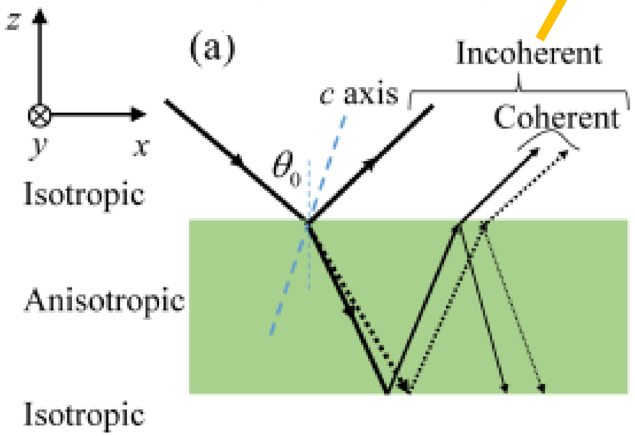
Depolarization

Example general case

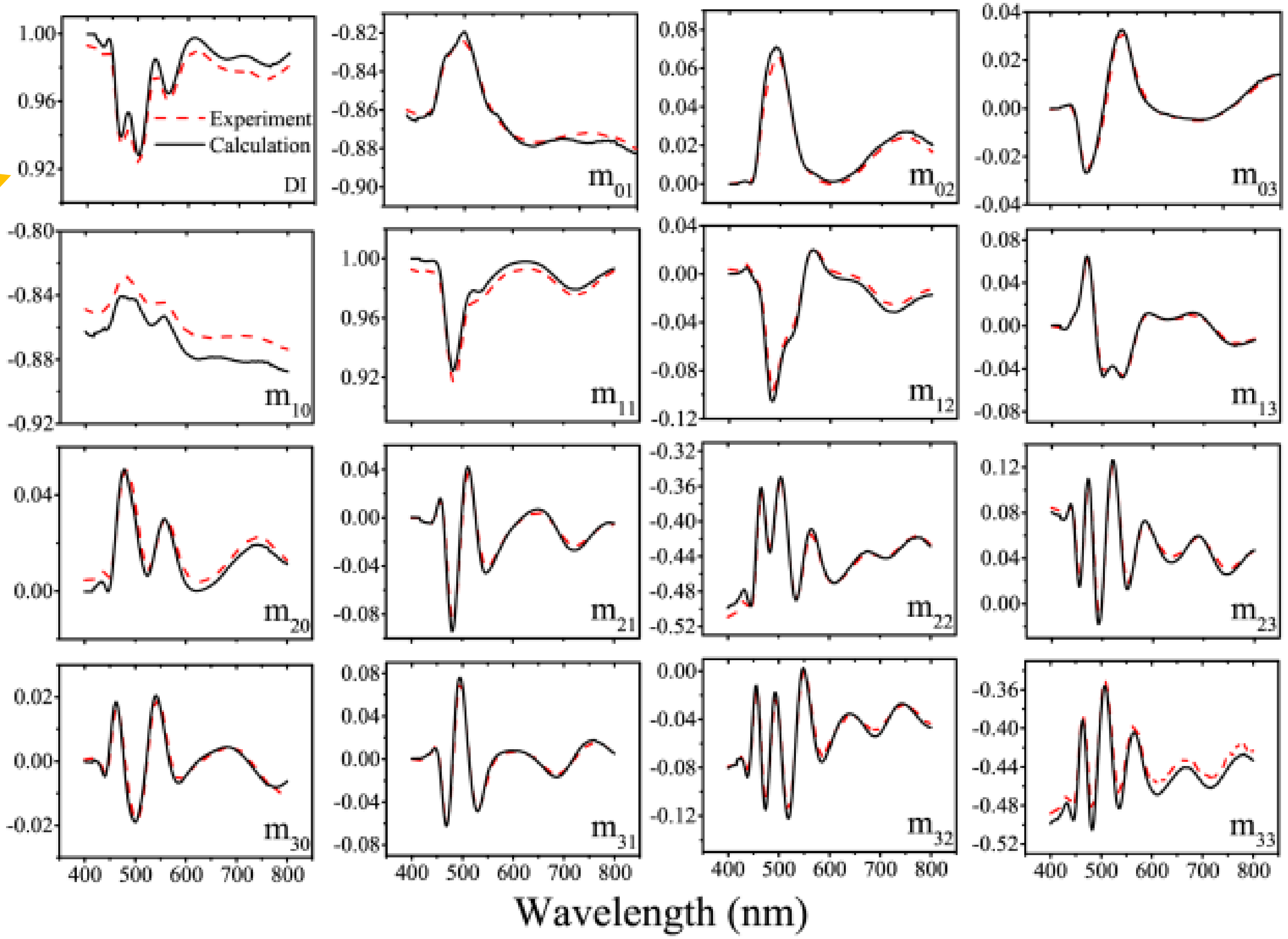
6H-SiC wafer (uniaxial)



$$\begin{pmatrix} \epsilon_{\perp} & 0 & 0 \\ 0 & \epsilon_{\perp} & 0 \\ 0 & 0 & \epsilon_{\parallel} \end{pmatrix}$$



Depolarization



H. Li, et al. Journal of Applied Physics, 128(23), 235304. (2020)

No symmetries if the OA is not parallel/perpendicular to the plane of incidence /sample plane

Outline

1. Introduction. MM types

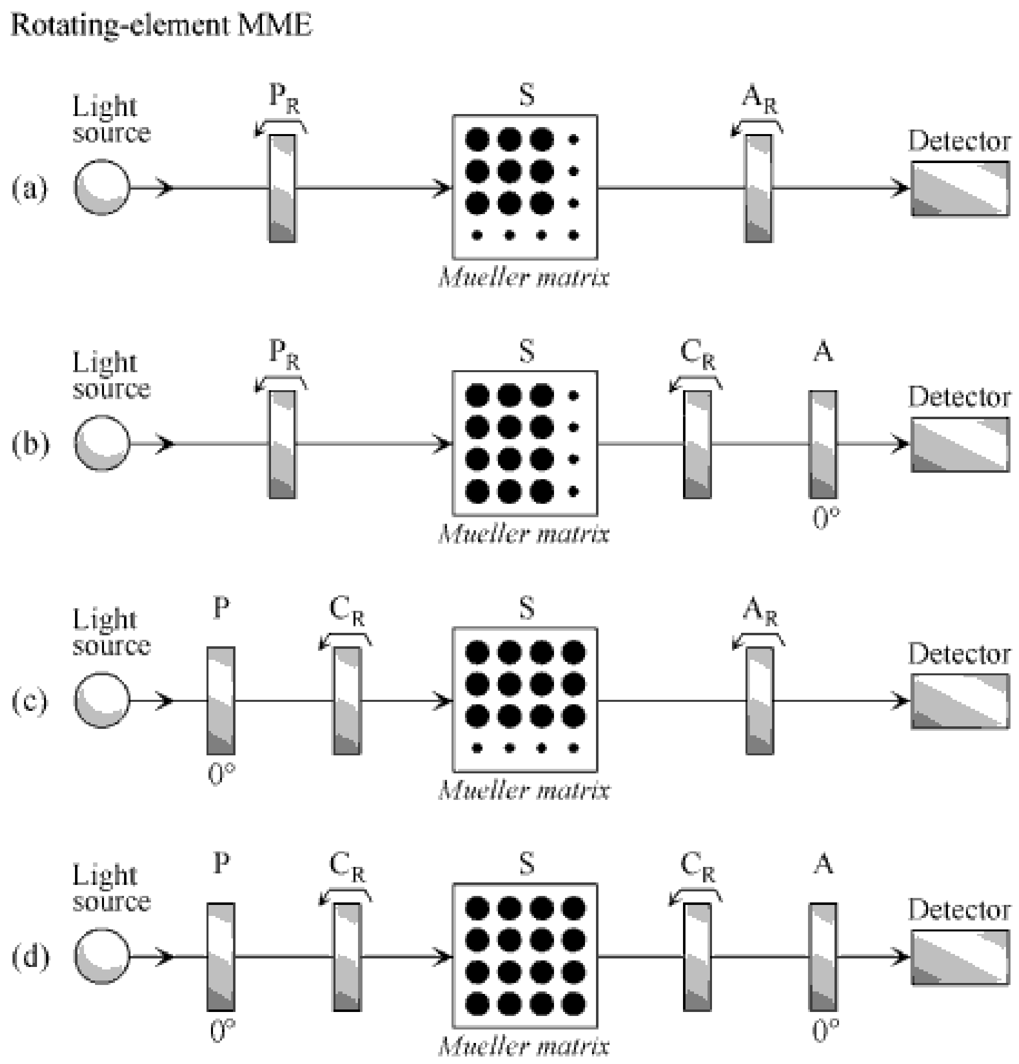
2. MM symmetries

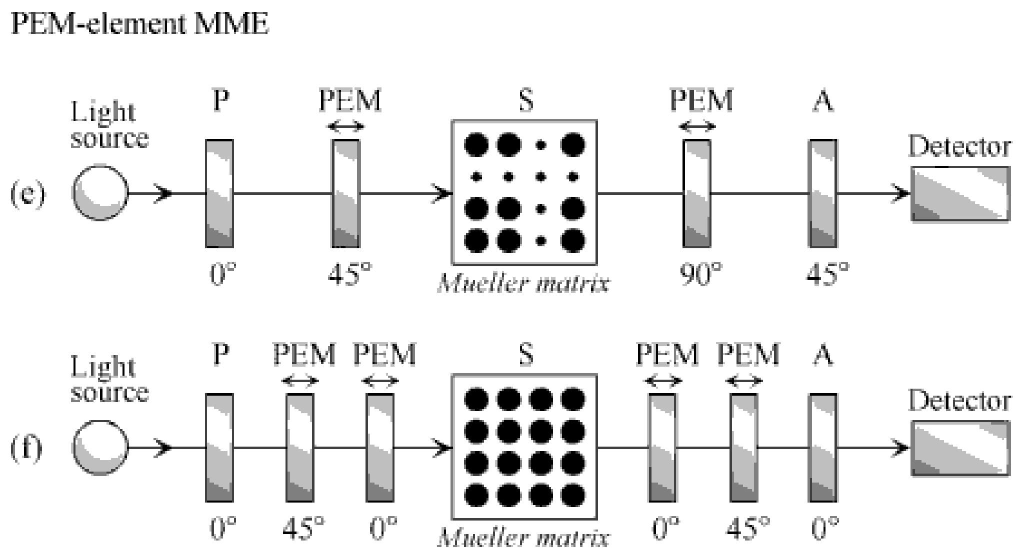
3. Depolarization

4. Complete vs incomplete MMs

5. Conclusions

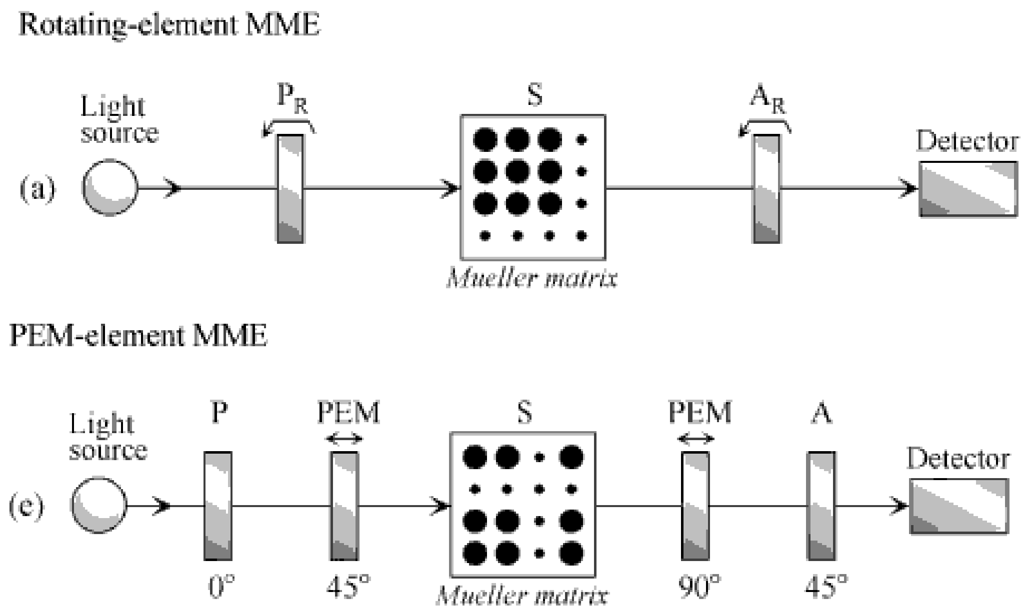
Complete vs incomplete MMs





P. S. Hauge, Surf. Sci. 96(1-3), 108–140 (1980)

Complete vs incomplete MMs



9 ELEMENTS MEASURED

For non depolarizing samples the full MM can be reconstructed

R. Ossikovski and O. Arteaga, "Complete Mueller matrix from a partial polarimetry experiment: the nine-element case," J. Opt. Soc. Am. A 36, 403-415 (2019)

R. Ossikovski and O. Arteaga, "Completing Experimental Non-Depolarizing Mueller Matrix with Both a Row and a Column Missing," J. Opt. Soc. Am. A accepted (2026)

(very fast reconstruction, no delay in the acquisition
+ see my talk on Thursday for more examples)

But there are two solutions!

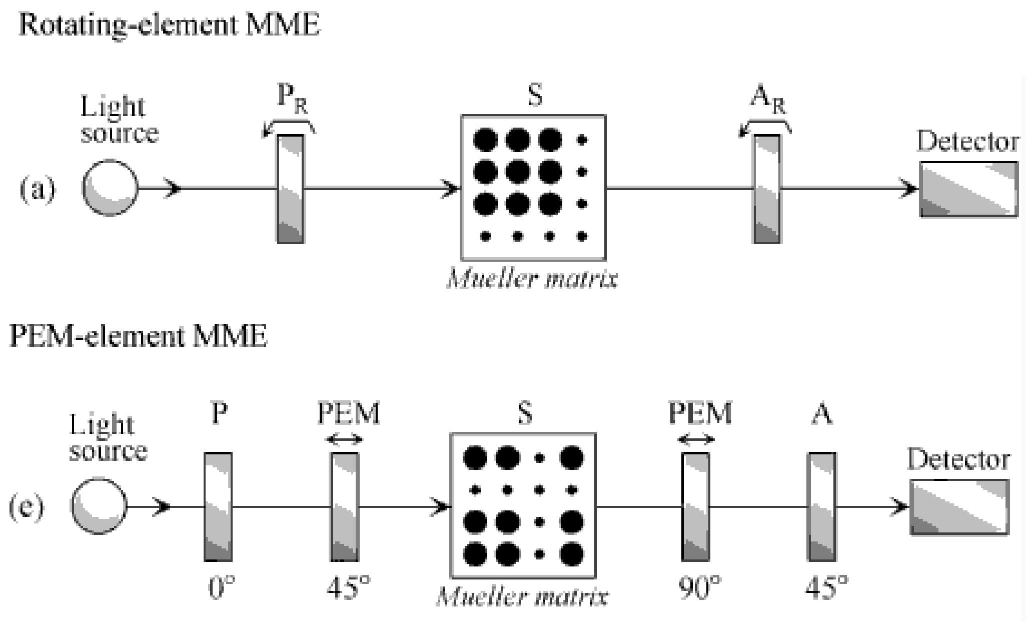
$$\begin{bmatrix} \bigcirc & \bigcirc & \bigcirc & \pm m_{03} \\ \bigcirc & \bigcirc & \bigcirc & \pm m_{13} \\ \bigcirc & \bigcirc & \bigcirc & \pm m_{23} \\ \pm m_{30} & \pm m_{31} & \pm m_{32} & m_{33} \end{bmatrix}$$

$$\begin{bmatrix} \bigcirc & \bigcirc & \pm m_{02} & \bigcirc \\ \pm m_{10} & \pm m_{11} & m_{12} & \pm m_{13} \\ \bigcirc & \bigcirc & \pm m_{22} & \bigcirc \\ \bigcirc & \bigcirc & \pm m_{32} & \bigcirc \end{bmatrix}$$

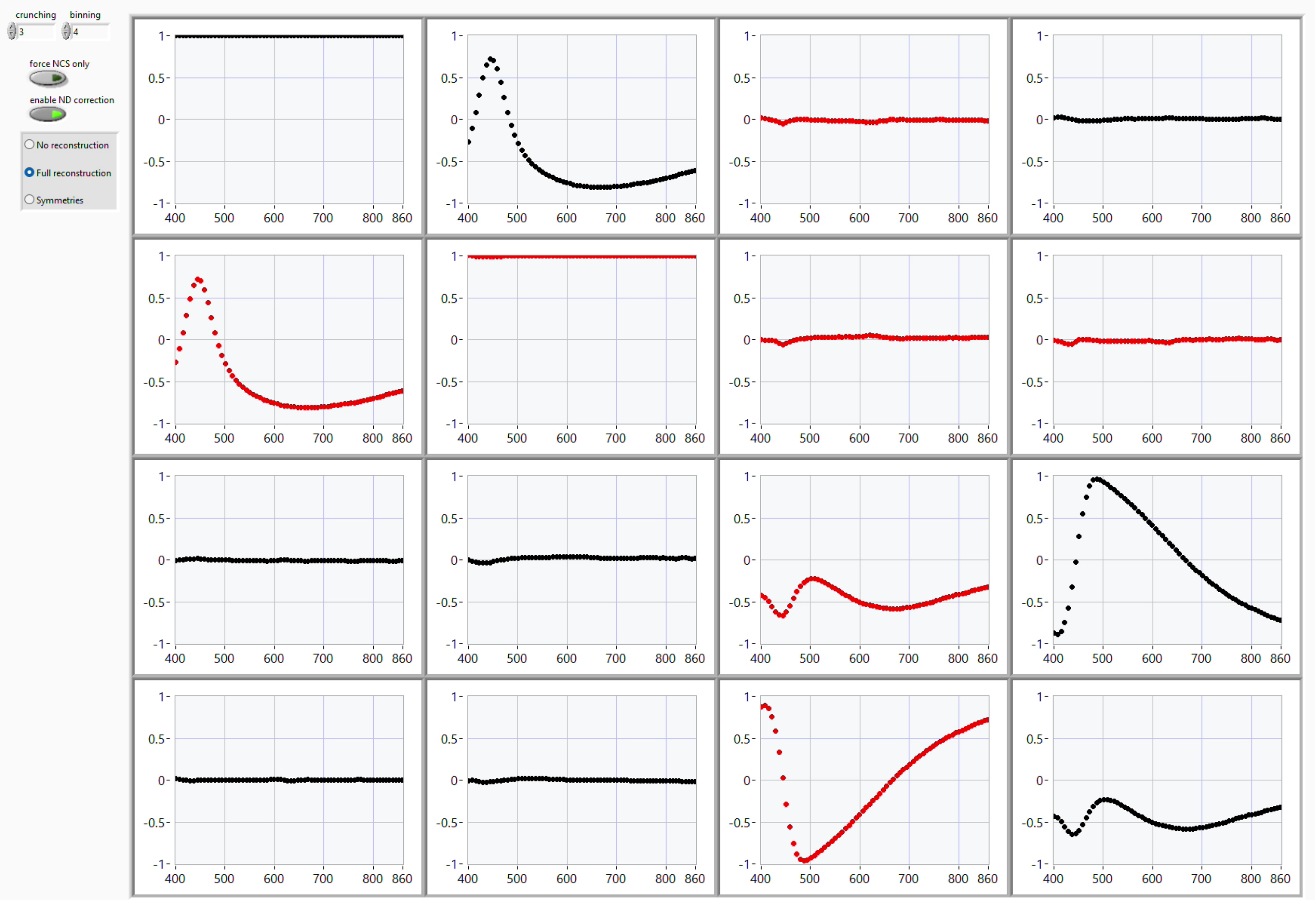
Here we cannot always chose the right solution! (retardance ambiguity!)

Here we can always chose the right solution!

Complete vs incomplete MMs



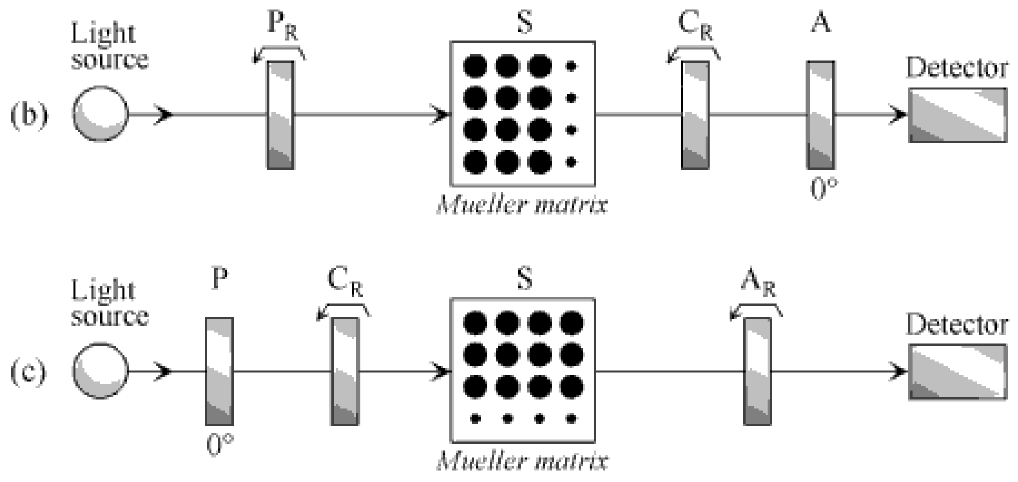
9 ELEMENTS MEASURED



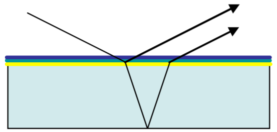
Black points are measured, red points are real-time reconstructed (under the non-depolarization assumption)

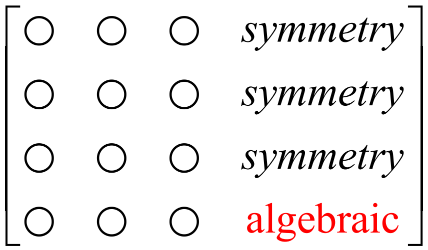
Talk on Thursday!

Complete vs incomplete MMs



For **depolarizing** samples the full MM can be reconstructed if using symmetries





To be published

12 ELEMENTS MEASURED

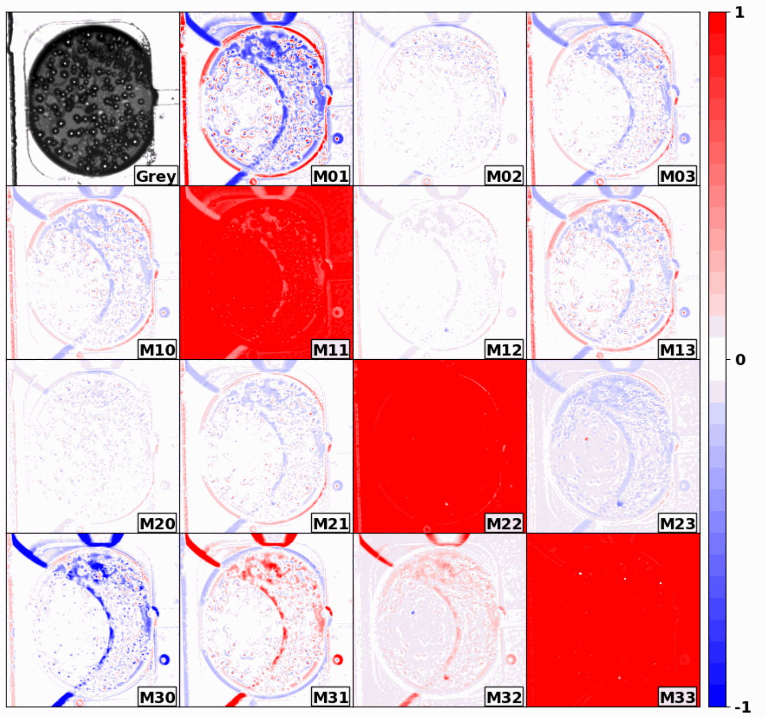
For **non depolarizing** samples the full MM can be reconstructed

Razvigor Ossikovski, Oriol Arteaga; Completing an experimental nondepolarizing Mueller matrix whose column or row is missing. *J. Vac. Sci. Technol. B* 1 (2019)

O. Arteaga and R. Ossikovski, "**Complete** Mueller matrix from a partial polarimetry experiment: the 12-element case," J. Opt. Soc. Am. A 36, 416-427 (2019). Very fast

Only one solution!

(very fast, no delays)



Outline

Conclusions

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

- A phenomenological understanding of Mueller matrix elements is relatively easy to obtain. In many cases, ***interpreting the Mueller matrix is actually the easiest part of the experiment analysis***.
- 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 misalignement of the sample

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Thank you for your attention