Characterization of Mueller matrices in retroreflex ellipsometry

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Abstract

Ellipsometry is a widely-used technique for characterizing materials and thin films. The principle is based on the polarization changes after light is reflected or transmitted at a sample. In general, the shape of the sample should be flat or nearly flat because ellipsometry is sensitive to the angle of incidence, tilt angle and the sample position (height variation). For nonplanar surfaces, retroreflex ellipsometry was proposed to solve the problem of the alignment. Despite of the Mueller matrix, the coherency matrix is often used for depolarization and noise reduction. In retroreflex ellipsometry, the measured Mueller matrix can be seen as a dual-rotation transformation. Therefore, it is important to discuss the changes of reference frames for Mueller matrices. In this report, the polarization model of retroreflex ellipsometry will be introduced. Decompositions and invariant quantities of a Mueller matrix with a dual-rotation transformation will be discussed.

1 Introduction

Ellipsometry is a widely-used technique for characterizing materials and thin films, e.g., in the semiconductor industry, biology and nanotechnology. The principle is based on the polarization changes after light is reflected or transmitted at a sample. The polarization characteristics can be described by Fresnel equations. The advantages of ellipsometry are non-destructive, fast measurements, and high accuracy and sensitivity. In general, the geometric shape of samples should be flat in order to fulfill the law of reflection or Snell's law. For nonplanar samples, the curvatures of the surface alter the reflected or transmitted light which causes experimental errors due to the misalignment. The worst-case scenario is that the detector cannot receive any signal. This restriction limits the feasibility of in-line measurements for industrial applications. In the last two decades, many approaches were proposed to overcome the shape restriction [19, 15, 10, 26, 14, 23, 16, 8]. However, these studies have some constraints, e.g., small measurement ranges, a short working distance, and complicated system design. In order to conquer these drawbacks, the concept of retroreflex ellipsometry

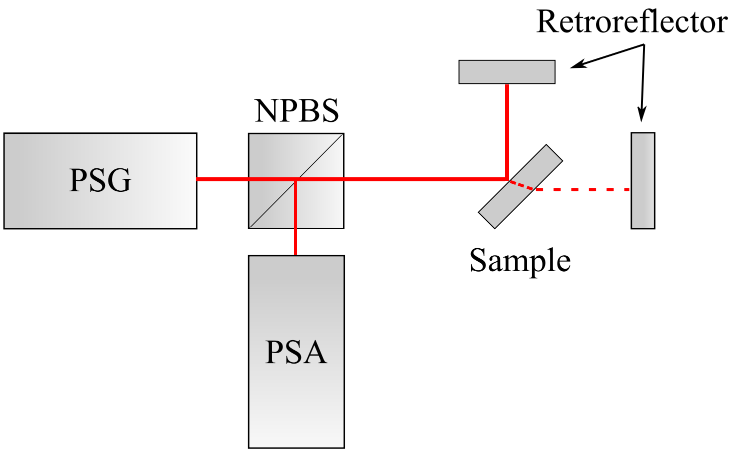

Figure 1.1: Schematic of the retroreflex ellipsometer in the reflection and transmission configurations, showing the polarization state analyzer (PSA), polarization state generator (PSG) and non-polarizing beam-splitter (NPBS).

(RRE) has been proposed at Fraunhofer IOSB[13, 4, 5, 18, 17]. Figure 1.1 shows the configuration of RRE whose concept is based on return-path ellipsometry [20, 2]. The key element in RRE is a retroreflector which returns the light along the same path and has the same polarization characteristics as an ideal mirror regardless of the angle of incidence (within an angular range up to $\pm 30°$). Therefore, the alignment of angles and position between the detector and the sample is automatically achieved. In this paper, the polarization model of retroreflex ellipsometry will be introduced, and decompositions and invariant quantities of the measured Mueller matrices will be discussed.

2 Polarization model of retroreflex ellipsometry

The polarization characteristics of optical elements or the interaction at the boundaries can be described by Jones vectors $\mathbf{E}$, Jones matrices $\mathbf{J}$, Stokes vectors $\mathbf{S}$ and Mueller matrices $\mathbf{M}$ [3, 9]. Jones vectors and Jones matrices can only be used for completely polarized light and nondepolarizing systems while Stokes vectors and Mueller matrices can be used for partially polarized light and depolarizing systems. A Jones matrix can be converted to the Mueller matrix by the transformation:

$$\mathbf{M} = \mathbf{A}(\mathbf{J} \otimes \mathbf{J}^*)\mathbf{A}^{-1}, \tag{2.1}$$

where $\otimes$ denotes the Kronecker product, the asterisk denotes complex conjugation, and $\mathbf{A}$ is the transformation matrix given by

$$\mathbf{A} = \begin{bmatrix} 1 & 0 & 0 & 1 \\ 1 & 0 & 0 & -1 \\ 0 & 1 & 1 & 0 \\ 0 & i & -i & 0 \end{bmatrix}. \tag{2.2}$$

Stokes vectors $\mathbf{S}$ (4×1 vector) describe the polarization state of the electromagnetic waves including fully polarized, partially polarized, or unpolarized light. Mueller matrices $\mathbf{M}$ (4×4 matrix) characterize the interaction between mediums and polarized light.

$$\mathbf{S} = \begin{bmatrix} s_0 \\ s_1 \\ s_2 \\ s_3 \end{bmatrix}, \mathbf{M} = \begin{bmatrix} m_{00} & m_{01} & m_{02} & m_{03} \\ m_{10} & m_{11} & m_{12} & m_{13} \\ m_{20} & m_{21} & m_{22} & m_{23} \\ m_{30} & m_{31} & m_{32} & m_{33} \end{bmatrix} \tag{2.3}$$

The Mueller matrix of an isotropic sample $\mathbf{M}_S$ can be expressed by the NSC representation:

$$\mathbf{M}_S = \begin{bmatrix} 1 & -N & 0 & 0 \\ -N & 1 & 0 & 0 \\ 0 & 0 & C & S \\ 0 & 0 & -S & C \end{bmatrix}, \tag{2.4}$$

where $N = \cos 2\varPsi$, $S = \sin 2\varPsi \sin \varDelta$, and $C = \sin 2\varPsi \cos \varDelta$. $\varPsi$ and $\varDelta$, which are functions of the angle of incidence and the refractive index of the sample, represent amplitude ratio and phase difference. When Mueller matrices are presented with different coordinate frames, the coordinate transformation should be applied. The Mueller matrix of a coordinate rotation $\mathbf{M}_R(\alpha)$ can be described as

$$\mathbf{M}_R(\alpha) = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos 2\alpha & -\sin 2\alpha & 0 \\ 0 & \sin 2\alpha & \cos 2\alpha & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix}, \tag{2.5}$$

where α is the rotation angle.

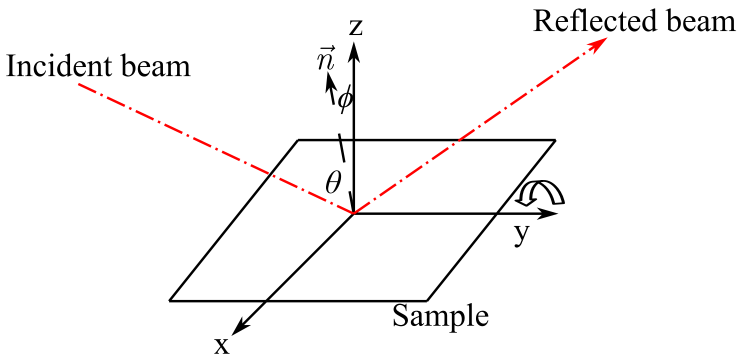

Figure 2.1: Definitions for the angle of incidence θ and the tilt angle ϕ of a tilted sample rotated around the y-axis.

Figure 2.1 shows that a flat sample on the x-y plane rotates around y-axis. If the surface is in parallel to x-y plane, the surface normal is the z-axis and the plane of incidence is determined by the z-axis and the incident beam. When the sample rotates around the y-axis, the surface normal of the sample becomes the vector $\vec{n}$. The tilt angle ϕ is defined by the surface normal $\vec{n}$ and the z-axis, and the angle of incidence θ is determined by the surface normal $\vec{n}$ and the incident beam. This model can be extended to nonplanar surfaces. A nonplanar surface can be seen as a flat surface rotates around y-axis. The incident angle θ and the tilt angle ϕ define the surface normal. The Mueller matrix model of the tilt sample for RRE is shown as [14]

$$\mathbf{M}_R(\alpha_{\mathrm{PSA}}) \cdot \mathbf{M}_S \cdot \mathbf{M}_{Retroreflector} \cdot \mathbf{M}_S \cdot \mathbf{M}_R(\alpha_{\mathrm{PSG}}), \tag{2.6}$$

where α_{PSA} and α_{PSG} are the rotation angles of the polarization state analyzer (PSA) and the polarization state generator (PSG). Assuming that the system is perfectly aligned, we can use the relation $\mathbf{M}_R(\alpha_{\mathrm{PSA}}) = \mathbf{M}_R(\alpha_{\mathrm{PSG}}) = \mathbf{M}_R(\phi)$ to simplify the equation as

$$\mathbf{M}_2 = \mathbf{M}_R(\phi) \cdot \mathbf{M}_1 \cdot \mathbf{M}_R(\phi), \tag{2.7}$$

where $\mathbf{M}_1 = \mathbf{M}_S \cdot \mathbf{M}_{Retroreflector} \cdot \mathbf{M}_S$. The ellipsometric parameters (Ψ, Δ) and the tilt angle ϕ from the Mueller matrix $\mathbf{M}_2$ can be solved by a numerical fitting method. $\mathbf{M}_2$ can be seen as a dual-rotation transformation of $\mathbf{M}_1$[11].

3 Decompositions of Mueller matrices with dual-rotation transformations

Figure 3.1 shows the different domains of 4×4 matrices. Mueller matrices are a subset of real 4×4 matrices because Mueller matrices contain physical properties (polarization). Mueller-Jones matrices are matrices which are derivable from Jones matrices. Therefore, not all Mueller matrices can be transformed from Jones matrices because Jones matrices only deal with nondepolarized systems. There have been many studies discussing necessary and sufficient conditions for a Mueller matrix [6, 21]. A Mueller matrix is a linear transformation of Stokes vectors. Hence, Mueller matrices must fulfill Stokes criterion:

$$s_0 \geq (s_1^2 + s_2^2 + s_3^2)^{\frac{1}{2}}. \tag{3.1}$$

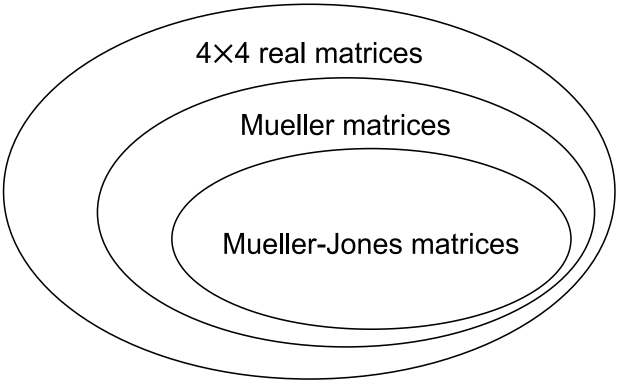

Figure 3.1: Different domains for 4×4 matrices.

For every Stokes vector $\mathbf{S}$ satisfies the criterion, the product of the Mueller matrix $\mathbf{M}$ and Stokes vector $\mathbf{S}$ also satisfies the criterion. Then the Mueller matrix fulfills the Stokes criterion.

In order to analyze depolarization, the wave coherency matrix $\mathbf{\Phi}$ is proposed as [24]:

$$\mathbf{\Phi} = \mathbf{E}(t) \otimes \mathbf{E}(t)^* = \begin{bmatrix} E_x(t)E_x^*(t) & E_x(t)E_y^*(t) \\ E_y(t)E_x^*(t) & E_y(t)E_y^*(t) \end{bmatrix} = \begin{bmatrix} j_{00} & j_{01} \\ j_{10} & j_{11} \end{bmatrix}, \tag{3.2}$$

where $\mathbf{E(t)}$ is a quasi-monochromatic wave whose amplitudes and phases depend on the time t. We can see depolarisation is related to second order products of the quasi-monochromatic wave. This concept can be applied to Mueller matrices. The covariance matrix $\mathbf{H}$ is defined as Kronecker product of the corresponding Jones covariance vector [22]:

$$\mathbf{H} = \frac{1}{2}\mathbf{T} \otimes \mathbf{T}^*, \tag{3.3}$$

where $\mathbf{T} = [j_{00} \;\; j_{01} \;\; j_{10} \;\; j_{11}]^T$ and j_{ij} is the element of the 2×2 Jones matrix. Hence, $\mathbf{H}$ is a 4×4 matrix. It is obvious that $\mathbf{H}$ and $\mathbf{M}$ are linearly related. Therefore, $\mathbf{H}$ can be written in terms of the elements m_{ij} of $\mathbf{M}$ as:

$$\mathbf{H}(\mathbf{M}) = \frac{1}{2}\begin{bmatrix} \begin{matrix} m_{00}+m_{01} \\ +m_{10}+m_{11} \end{matrix} & \begin{matrix} m_{02}+m_{12} \\ +i(m_{03}+m_{13}) \end{matrix} & \begin{matrix} m_{20}+m_{21} \\ -i(m_{30}+m_{31}) \end{matrix} & \begin{matrix} m_{22}+m_{33} \\ +i(m_{23}-m_{32}) \end{matrix} \\ \begin{matrix} m_{02}+m_{12} \\ -i(m_{03}+m_{13}) \end{matrix} & \begin{matrix} m_{00}-m_{01} \\ +m_{10}-m_{11} \end{matrix} & \begin{matrix} m_{22}-m_{33} \\ -i(m_{23}+m_{32}) \end{matrix} & \begin{matrix} m_{20}-m_{21} \\ -i(m_{30}-m_{31}) \end{matrix} \\ \begin{matrix} m_{20}+m_{21} \\ +i(m_{30}+m_{31}) \end{matrix} & \begin{matrix} m_{22}-m_{33} \\ +i(m_{23}+m_{32}) \end{matrix} & \begin{matrix} m_{00}+m_{01} \\ -m_{10}-m_{11} \end{matrix} & \begin{matrix} m_{02}-m_{12} \\ +i(m_{03}-m_{13}) \end{matrix} \\ \begin{matrix} m_{22}+m_{33} \\ -i(m_{23}-m_{32}) \end{matrix} & \begin{matrix} m_{20}-m_{21} \\ +i(m_{30}-m_{31}) \end{matrix} & \begin{matrix} m_{02}-m_{12} \\ -i(m_{03}-m_{13}) \end{matrix} & \begin{matrix} m_{00}+m_{01} \\ +m_{10}+m_{11} \end{matrix} \end{bmatrix} \tag{3.4}$$

The covariance matrix $\mathbf{H}$ provides necessary and sufficient conditions for a Mueller matrix to be derivable from a Jones matrix [1]. The form of $\mathbf{H}$ is a positive semidefinite Hermitian matrix, which means its eigenvalues are non-negative. In other words, a matrix is a physical realizable Mueller matrix if its coherency matrix $\mathbf{H}$ has non-negative eigenvalues. This concept can be used to determine physical Mueller matrices and reduce experimental errors [7].

Experimental errors in ellipsometry might induce nonphysical Mueller matrices (negative eigenvalues in corresponding covariance matrices $\mathbf{H}$). For example, a depolarizing Mueller matrix is measured due to the noise from the light source and the detector. The idea of sum decomposition or matrix filtering for experimental Mueller matrices is proposed by Cloude [7]. The covariance matrix of a physically realizable Mueller matrix can be decomposed to four covariance matrices of Mueller-Jones matrices as:

$$\mathbf{H} = \lambda_1\mathbf{H}_1 + \lambda_2\mathbf{H}_2 + \lambda_3\mathbf{H}_3 + \lambda_4\mathbf{H}_4. \tag{3.5}$$

If a Mueller matrix is nonphysical, at least one eigenvalue of its covariance matrix is negative. The filtering concept is to remove any negative contributions and covert the remaining term to a Mueller matrix, which can be described as:

$$\sum_{i=1}^{4}\frac{1}{2}(1+\mathrm{sgn}(\lambda_i))\mathbf{H}_i \Rightarrow \mathbf{M}_{filtering}, \tag{3.6}$$

where sgn is the sign function and λ_i is the eigenvalue of $\mathbf{H}$. Finally, The nearest non-depolarising Mueller matrix is obtained. This method is proved as an optimal filtering [25].

We can apply covariance matrices in retroreflex ellipsometry. The Mueller matrix for a gold sample at a wavelength of 632.8 nm and an incident angle of

$70°$ is given by

$$\mathbf{M}_{gold} = \begin{bmatrix} 1 & -0.094 & 0 & 0 \\ -0.094 & 1 & 0 & 0 \\ 0 & 0 & 0.802 & -0.589 \\ 0 & 0 & 0.589 & 0.802 \end{bmatrix}. \tag{3.7}$$

The eigenvalues of the covariance matrix $\mathbf{H}(\mathbf{M_{gold}})$ are $[1, 0, 0, 0]$, which means the matrix $\mathbf{M}_{gold}$ is a Mueller-Jones matrix. When the gold sample is tilted with $5°$. The Mueller matrix $\mathbf{M}_{gold}$ becomes

$$\mathbf{M}'_{gold} = \begin{bmatrix} 1 & -0.093 & -0.016 & 0 \\ -0.093 & 0.946 & 0.309 & -0.102 \\ 0.016 & -0.308 & 0.748 & -0.580 \\ 0 & -0.102 & 0.580 & 0.802 \end{bmatrix} \tag{3.8}$$

We can observe that the off-diagonal 2×2 blocks are nonzero elements. The eigenvalues of the covariance matrix $\mathbf{H}(\mathbf{M'_{gold}})$ are $[1, 0.015, -0.015, 0]$. Minus eigenvalue means that $\mathbf{M}'_{gold}$ is not a physically realizable Mueller matrix. The change of plane of incidence caused the anisotropic and depolarizing effect.

We can prove the Mueller matrix with a dual-rotation transformation $\mathbf{M}_2$ is not positive semi-definite by Sylvester's criterion [12]. A Hermitian matrix is positive semi-definite if and only if all principal minors of it are non-negative. For a 4×4 matrix, there are 15 principal minors D_k, where k is the order. The principal minors of $\mathbf{M}_2$ is shown as

$$\begin{aligned} D_1 &= [0, \frac{1}{2}(1-C)(1-\cos 4\phi), \frac{1}{2}(1-C)(1+\cos 4\phi), 1+C] \\ D_2 &= [0, 0, 0, 0, 0, 0] \\ D_3 &= [0, 0, 0, \frac{1}{2}S^2(1-C)(-1+\cos 8\phi)] \\ D_4 &= 0 \end{aligned} \tag{3.9}$$

Since $C, S \in [-1, 1]$ and $\phi \in [-90°, 90°]$, the principal minor in D_3 is negative when the tilt angle is not zero. The special cases are $S = 0$ and $C = 1$. $S = 0$ means $\Psi = 45°$ and the corresponding Mueller matrix is an ideal depolarizer which output randomly polarized light. $C = 1$ means $\Psi = 45°$ and $\Delta = 0°$ or

$360°$ and the Mueller matrix of this case is the same as the Mueller matrix of air. Except of these two cases, we can use this property to find the tilt angle by maximizing the $h_{fidelity}$ index[7]:

$$h_{fidelity} = 10\log\frac{\sum|\lambda_-|}{\sum\lambda_+}, \tag{3.10}$$

where λ_+ and λ_- are positive and negative eigenvalues of the correspingding covariance matrix.

4 Invariant quantities of a Mueller matrix with a dual-rotation transformation

There are polarimetric quantities which keep invariant under reference frame rotations. These invariant quantities can be used for determination of orientations of anisotropic materials and provide physical information. The Muller matrix model of a sample with a tilt angle in retrorelfex ellipsometry can be seen as a Mueller matrix with a dual-rotation transformation. The full form of $\mathbf{M}_1$ and $\mathbf{M}_2$ are expressed as:

$$\mathbf{M}_1 = \begin{bmatrix} 1+N^2 & -2N & 0 & 0 \\ -2N & 1+N^2 & 0 & 0 \\ 0 & 0 & S^2-C^2 & -2CS \\ 0 & 0 & 2CS & S^2-C^2 \end{bmatrix}, \tag{4.1}$$

$$\mathbf{M}_2 = \begin{bmatrix} 1+N^2 & -2N\cos 2\phi & 2N\sin 2\phi & 0 \\ -2N\cos 2\phi & 1-S^2+(1-C^2)\cos 4\phi & -(1-C^2)\sin 4\phi & 2CS\sin 2\phi \\ -2N\sin 2\phi & (1-C^2)\sin 4\phi & S^2-1+(1-C^2)\cos 4\phi & -2CS\cos 2\phi \\ 0 & 2CS\sin 2\phi & 2CS\cos 2\phi & S^2-C^2 \end{bmatrix}. \tag{4.2}$$

Compared $\mathbf{M}_1$ with $\mathbf{M}_2$, the following parameters are rotation invariant:

$$m_{00}, m_{03}, m_{30}, m_{33}, \tag{4.3}$$

$$m_{00}^2+m_{02}^2, m_{10}^2+m_{20}^2, m_{13}^2+m_{23}^2, m_{31}^2+m_{32}^2, \tag{4.4}$$

$$m_{11}^2+m_{12}^2+m_{21}^2+m_{22}^2, \tag{4.5}$$

$$\mathrm{Det}(\mathbf{M}), \tag{4.6}$$

$$\lambda_1 + \lambda_2 + \lambda_3 - 3\lambda_4, \tag{4.7}$$

where Det denotes determinant and λ_i is the eigenvalue of the correspingding covariance matrix.

From $\mathbf{M}_2$, the tilt angle ϕ can be obtained by

$$\phi = \tan^{-1}\frac{m_{02}}{m_{01}} = \tan^{-1}\frac{m_{20}}{m_{10}} = \tan^{-1}\frac{m_{13}}{m_{23}} = \tan^{-1}\frac{m_{31}}{m_{32}}. \tag{4.8}$$

The NSC parameters can be determined by

$$\begin{aligned} N^2 &= \frac{m_{01}^2 + m_{02}^2}{4} = \frac{m_{10}^2 + m_{20}^2}{4} \\ S^2 &= \frac{1}{2}(\sqrt{m_{31}^2 + m_{32}^2 + m_{33}^2} + m_{33}) = \frac{1}{2}(\sqrt{m_{13}^2 + m_{23}^2 + m_{33}^2} + m_{33}) \\ C^2 &= \frac{1}{2}(\sqrt{m_{31}^2 + m_{32}^2 + m_{33}^2} - m_{33}) = \frac{1}{2}(\sqrt{m_{13}^2 + m_{23}^2 + m_{33}^2} - m_{33}). \end{aligned} \tag{4.9}$$

Finally, the ellipsometric parameters Ψ and Δ can be determined by the NSC parameters. It is worthwhile to mention that the NSC parameters can be caulculated without knowing the tilt angle ϕ. In other words, NSC parameters are only related to the angle of incidence and material properties. If the refractive index of the sample is known, the angle of incidence can be solved analytically.

For isotropic materials, tilt angles induce anisotropic Mueller matrices. There are variant and invariant polarimetric quantities in the anisotropic matrices. These invariant quantites provide the information of rotation of reference frames. Moreover, the invariant quantities can be extended to anisotropic materials for separating azimuthal orientation and tilt angles.

5 Summary

In this report, the principle of retrorefelx ellipsometry, coherency matrix, Cloude's decomposition and invariant quantities for nonplanar surfaces have been introduced. The concept of RRE can measure samples with nonplanar shapes. The retroreflector acts as an ideal mirror regardless the incident angle ($< 30°$). The polarization model of the tilt sample can be seen as a dual-rotation transformation and the form of an isotropic Mueller matrix after rotation becomes anisotropic. The coherency matrix $\mathbf{H}$ provides necessary and sufficient conditions for a Mueller matrix which can be derivable from a Jones matrix. The Cloude's decomposition can reduce the experimental noise by filtering nonphysical contribution (negative eigenvalues) and can also be used to determine the tilt angle ϕ. While the sample has a tilt angle, $\mathbf{H}$ becomes nonphysical. This nonphysical Mueller matrix provides indication of tilt angles. Compared to the physical matrix (without tilting) and nonphysical matrix (with tilting), invariant quantities provide another way to calculate tilt angles and ellipsometric parameters. In the future, we plan to use these properties of Mueller matrices to improve the procedure of determination of the tilt angle ϕ and ellipsometric parameters (Ψ, Δ) and extend this method to anisotropic materials.

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