Optical Properties of Solids: Lecture 3

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http://ellipsometry.nmsu.edu

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Optical Properties of Solids: Lecture 3

- Maxwell's Equations in Vacuum, Plane Waves Polarized Light
- Stokes Parameters, Poincare Sphere Jones Vectors, Jones Matrix, Mueller Matrix
- Decoherence and Depolarization



References: Maxwell's Equations and Ellipsometry

Standard Texts on Electricity and Magnetism:

- J.D. Jackson: *Classical Electrodynamics*
- Landau & Lifshitz, Vol. 2: Classical Theory of Fields
- D.E. Aspnes: **Practical** Electrodynamics (forthcoming)

Optics:

- E. Hecht: Optics
- M. Born, E. Wolf: *Principles of Optics*

Ellipsometry and Polarized Light:

- R.M.A. Azzam and N.M. Bashara: *Ellipsometry and Polarized Light*
- H.G. Tompkins and E.A. Irene: Handbook of Ellipsometry (chapter by Josef Humlicek)
- H. Fujiwara, Spectroscopic Ellipsometry
- H.G. Tompkins and J.N. Hilfiker: Spectroscopic Ellipsometry
- H. Fujiwara and R.W. Collins: *Spectroscopic Ellipsometry for PV* (Vol 1+2)
- Zollner: Propagation of EM Waves in Continuous Media (Lecture Notes)



Scalars and Vectors

- Scalar: Invariant under rotations and inversion
- **Vector:** Transforms like x,y,z under rotation. Sign change under inversion
- **Pseudoscalar:** Invariant under rotations, sign change under inversion
- **Pseudovector:** Transforms like x,y,z under rotation. Invariant under inversion.

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	O _h (m-3m)	#	1	4	2 ₁₀₀	3	2 ₁₁₀	-1	-4	m ₁₀₀	-3	m ₁₁₀	functions	
BSW	Mult.	K	1	6	3	8	6	1	6	3	8	6		
Γ ₁	A _{1g}	Γ ₁ +	1	1	1	1	1	1	1	1	1	1	1, $x^{2}+y^{2}+z^{2}$	Scalar
Γ <mark>,</mark> '	A _{1u}	۲ ₁ -	1	1	1	1	1	-1	-1	-1	-1	-1		Pseudoscalar
Γ ₂	A _{2g}	Г2 ⁺	1	-1	1	1	-1	1	-1	1	1	-1		
Γ <mark>2</mark> ΄	A _{2u}	Γ ₂ -	1	-1	1	1	-1	-1	1	-1	-1	1	хуz	
Γ ₁₂	Eg	Г ₃ +	2	0	2	-1	0	2	0	2	-1	0	(2z ² -x ² -y ² ,x ² -y ²)	
Γ ₁₂ '	Eu	Г ₃ -	2	0	2	-1	0	-2	0	-2	1	0	-	
Г ₂₅	T _{2u}	Г5 ⁻	3	-1	-1	0	1	-3	1	1	0	-1	-	
Γ ₂₅ '	T _{2g}	Г ₅ +	3	-1	-1	0	1	3	-1	-1	0	1	(xy,xz,yz)	
Γ ₁₅	T _{1u}	Γ4-	3	1	-1	0	-1	-3	-1	1	0	1	(x,y,z)	Vector
Γ ₁₅ '	T _{1g}	Г4+	3	1	-1	0	-1	3	1	-1	0	-1	(J_x, J_y, J_z)	Pseudovector
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Scalar and Vector Waves

- **Field:** Scalar or vector depends on position **r**.
- Physical quantities are always real Scalar: energy, charge, etc.
 Vector: momentum, current density, electric field, etc.
- Scalar wave

$$s(\vec{r},t) = A\cos\left(\vec{k}\cdot\vec{r} - \omega t + \varphi\right)$$

• Vector wave

$$\vec{E}(\vec{r},t) = \vec{E}_0 A \cos\left(\vec{k}\cdot\vec{r} - \omega t + \varphi\right)$$

• Where do the complex notations come from?



Fourier Series of Periodic Functions

- A real-valued scalar function f(t) is called periodic with period T, if f(t)=f(t+T) for all values of t.
- A periodic scalar function with period T can be written as a Fourier Series

$$f(t) = \frac{1}{2}A_0 + \sum_{m=1}^{\infty} [A_m \cos(m\omega t) + B_m \sin(m\omega t)]$$

with angular frequency $\omega = 2\pi/T$ and Fourier coefficients
$$A_m = \frac{\omega}{\pi} \int_{-\frac{\pi}{\omega}}^{\frac{\pi}{\omega}} f(t) \cos(m\omega t) dt$$
$$B_m = \frac{\omega}{\pi} \int_{-\frac{\pi}{\omega}}^{\frac{\pi}{\omega}} f(t) \sin(m\omega t) dt$$
$$ackson, E&M, 1975$$

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-1

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x in periods

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Fourier Series of Periodic Functions

- Dealing with harmonic functions (sin, cos) is not convenient, because
 - We need two functions for each harmonic.
 - Taking derivatives is not easy, because sin and cos switch at each order.
- A periodic scalar function with period T can be written as a Fourier Series

$$f(t) = \sum_{m=-\infty}^{\infty} c_m \exp(-im\omega t)$$

with **complex** Fourier coefficients

$$c_m = \frac{\omega}{\pi} \int_{-\frac{\pi}{\omega}}^{\frac{\pi}{\omega}} f(t) \exp(im\omega t) dt = \begin{cases} \frac{A_0}{2} & m = 0\\ \frac{1}{2}(A_m + iB_m) & m > 0\\ \frac{1}{2}(A_{-m} - iB_{-m}) & m < 0 \end{cases}$$

- The Fourier coefficients are now complex, but the function f(t) is still real.
- The imaginary parts all cancel, if the complex coefficients c_m are defined correctly.

Jackson, E&M, 1975



Fourier Transforms of Non-Periodic Functions

- If the function f(t) is not periodic, then the period T becomes infinite and the frequency spacing ω between overtones becomes very small.
- The Fourier series now becomes a Fourier Integral.

 $f(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} F(\omega) \exp(-i\omega t) d\omega$

with the Fourier transform $F(\omega)$

$$F(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} f(t) \exp(i\omega t) dt$$

- The prefactors $1/\sqrt{2\pi}$ before the integral can vary (depends on convention).
- The Fourier transform function F(ω) is allowed to be complex, because it is not a meaningful physical quantity.
- Orthogonality and completeness:

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} \exp[i(\omega - \omega')t] dt = \delta(\omega - \omega')$$
$$\frac{1}{2\pi} \int_{-\infty}^{\infty} \exp[i\omega(t - t')t] d\omega = \delta(t - t')$$

Jackson, E&M, 1975





Orthonormal basis of **Hilbert Space** of real functions



Math with Fourier Transforms

Convolution theorem:

The Fourier transform of a convolution equals the product of the Fourier transforms.

 $(f * g)(t) = \int_{-\infty}^{\infty} f(t')g(t - t')dt'$

$$\frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} (f * g)(t) \exp(i\omega t) dt = \sqrt{2\pi} F(\omega) G(\omega)$$

The Fourier transform of the derivative of f(t) equals $-i\omega F(\omega)$.

$$\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty}f'(t)\exp(i\omega t)dt = -i\omega F(\omega)$$

The complex conjugate of the Fourier transform equals $F(-\omega)$. ٠

 $\overline{F(\omega)} = F(-\omega)$



Fourier Series in Multiple Dimensions

- A real-valued scalar field s(r) in a Bravais lattice (with Bravais lattice vectors T and reciprocal lattice vectors G) is called periodic, if s(r+T)=s(r) for all Bravais lattice vectors T.
- A real-valued periodic scalar field s(r) in a Bravais lattice can be written as a Fourier sum in reciprocal space

$$s(\vec{r}) = \sum_{\vec{G}} s_{\vec{G}} \exp\left(i\vec{G}\cdot\vec{r}\right)$$

with complex Fourier coefficients

$$s_{\vec{G}} = \frac{1}{V} \int_{C} s(\vec{r}) \exp\left(-i\vec{G} \cdot \vec{r}\right) d^{3}\vec{r}$$

where C is the unit cell with volume V. G is a reciprocal lattice vector.

The same equations apply to a real-valued periodic vector field $\mathbf{E}(\mathbf{r})$.

$$\vec{E}(\vec{r}) = \sum_{\vec{G}} \vec{E}_{\vec{G}} \exp(i\vec{G} \cdot \vec{r})$$
$$\vec{E}_{\vec{G}} = \frac{1}{V} \int_{C} \vec{E}(\vec{r}) \exp(-i\vec{G} \cdot \vec{r}) d^{3} \vec{r}$$
shcroft & Mermin, Appendix D



Fourier Transforms in Multiple Dimensions

Fourier transforms can also be generalized to multiple dimensions for scalar fields

$$s(\vec{r}) = \left(\frac{1}{\sqrt{2\pi}}\right)^3 \iiint_{-\infty}^{\infty} S\left(\vec{k}\right) \exp\left(i\vec{k}\cdot\vec{r}\right) d^3\vec{k}$$

$$S(k) = \left(\frac{1}{\sqrt{2\pi}}\right)^3 \iiint_{-\infty}^{\infty} s(\vec{r}) \exp\left(-i\vec{k}\cdot\vec{r}\right) d^3\vec{r}$$

and vector fields

$$\vec{E}(\vec{r}) = \left(\frac{1}{\sqrt{2\pi}}\right)^3 \iiint_{-\infty}^{\infty} \vec{E}\left(\vec{k}\right) \exp\left(i\vec{k}\cdot\vec{r}\right) d^3\vec{k}$$

$$\vec{E}(k) = \left(\frac{1}{\sqrt{2\pi}}\right)^3 \iiint_{-\infty}^{\infty} \vec{E}(\vec{r}) \exp\left(-i\vec{k}\cdot\vec{r}\right) d^3\vec{r}$$

- The fields $s(\mathbf{r})$ and $\mathbf{E}(\mathbf{r})$ in real space have real values. ٠
- The Fourier transforms S(k) and E(k) have complex values, but their imaginary parts cancel out in the summation.



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Microscopic Maxwell's Equations (in Vacuum)

- Electric field strength **E**(**r**)
- Magnetic field strength $H(\mathbf{r})$
- Current density $\mathbf{j}(\mathbf{r})$, charge density $\rho(\mathbf{r})$
- Permittivity of free space ε_0 , permeability of free space μ_0 .

 $\vec{\nabla} \cdot \vec{E} = \frac{\rho}{\varepsilon_0} = 0$ Gauss' Law (Coulomb) $\vec{\nabla} \cdot \vec{H} = 0$ Gauss' Law (magnetic field) $\vec{\nabla} \times \vec{E} = -\mu_0 \frac{\partial \vec{H}}{\partial t}$ Faraday's Law (Lenz) $\vec{\nabla} \times \vec{H} = \vec{j} + \varepsilon_0 \frac{\partial \vec{E}}{\partial t} = \varepsilon_0 \frac{\partial \vec{E}}{\partial t}$ **Ampere's Law**

- Homogeneous (in vacuum), linear, first-order, constant coefficients, partial DEQ.
- Vector analysis can be used (Stokes' Theorem) to transform Maxwell's equations into integral form.
- Integration. Introduce speed of light $C = \frac{1}{\sqrt{\varepsilon_0 \mu_0}}$

Jackson, E&M, 1975

Wave Equations (in Vacuum)

- Electric field strength $\mathbf{E}(\mathbf{r})$; Magnetic field strength $\mathbf{H}(\mathbf{r})$.
- Maxwell's equations can be combined to obtain the vacuum wave equations (second order, linear, homogeneous, constant coefficients).

$$\vec{\nabla}^2 \vec{E} - \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = 0$$
$$\vec{\nabla}^2 \vec{H} - \frac{1}{c^2} \frac{\partial^2 \vec{H}}{\partial t^2} = 0$$

$$c = \frac{1}{\sqrt{\varepsilon_0 \mu_0}}$$

Plane wave solutions:

$$\vec{E}(\vec{r},t) = \vec{E}_0 \exp\left[i\left(\vec{k}\cdot\vec{r}-\omega t\right)\right]$$
$$\vec{H}(\vec{r},t) = \vec{H}_0 \exp\left[i\left(\vec{k}\cdot\vec{r}-\omega t\right)\right]$$

Why are the solutions complex ?

- Plane wave is not physical (infinite, monochromatic). Form Gaussian wave packets.
- Poynting vector indicates energy flow:

$$\vec{S} = \vec{E} \times \vec{H}$$

Jackson, E&M, 1975



Plane-Wave Solutions to Maxwell's Equations (Vacuum)

- Electric field strength $\mathbf{E}(\mathbf{r})$; Magnetic field strength $\mathbf{H}(\mathbf{r})$.
- Any electric and magnetic field strength can be written as a Fourier-transform

$$\vec{E}(\vec{r},t) = \left(\frac{1}{2\pi}\right)^2 \int d\omega \iiint d^3\vec{k} \ \vec{E}\left(\vec{k},\omega\right) \exp\left[i\left(\vec{k}\cdot\vec{r}\right) - \omega t\right]$$
$$\vec{E}\left(\vec{k},\omega\right) = \left(\frac{1}{2\pi}\right)^2 \int dt \iiint d^3\vec{r} \ \vec{E}(\vec{r},t) \exp\left[-i\left(\vec{k}\cdot\vec{r}\right) - \omega t\right]$$

- The Fourier transforms are complex, but the E(r) and H(r) fields are not.
- Signs: Nebraska convention as modified by Aspnes. Kinetic energy of free particle in quantum mechanics is positive. Classical wave travels along k.
- The complex plane waves

$$\vec{E}(\vec{r},t) = \vec{E}_0 \exp\left[i\left(\vec{k}\cdot\vec{r}-\omega t\right)\right]$$
$$\vec{H}(\vec{r},t) = \vec{H}_0 \exp\left[i\left(\vec{k}\cdot\vec{r}-\omega t\right)\right]$$

are just one term in the Fourier transform. The entire integral is real. (Add complex conjugate.)

Solutions to Maxwell's equations are superpositions of plane waves. ٠

Jackson, E&M, 1975 Stefan Zollner, February 2019, Optical Properties of Solids Lecture 3



Fourier-transform Maxwell's Equations

• Substitute plane wave solutions into the differential form of Maxwell's Equations:



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Fourier-transform Maxwell's Equations

 $\vec{E}(\vec{r},t) = \vec{E}_0 \exp\left[i\left(\vec{k}\cdot\vec{r}-\omega t\right)\right]$ $\vec{H}(\vec{r},t) = \vec{H}_0 \exp\left[i\left(\vec{k}\cdot\vec{r} - \omega t\right)\right]$ $\vec{k} \cdot \vec{E}_0 = 0$ Gauss' Law (Coulomb) $\vec{k} \cdot \vec{H}_0 = 0$ Gauss' Law (magnetic field) $\vec{k} \times \vec{E}_0 = \omega \mu_0 \vec{H}_0$ Faraday's Law $\vec{k} \times \vec{H}_0 = -\omega \varepsilon_0 \vec{E}_0$ Ampere's Law $k^2 = \frac{\omega^2}{c^2}$ Wave equation (Dispersion relation)

Any solution to Maxwell's equation in vacuum can be written as a superposition of plane waves.

EM waves are transverse (E, H perpendicular to k).

 $\mathbf{E} \perp \mathbf{H}$, $\mathbf{E}_0 = Z_0 \mathbf{H}_0$, $Z_0 = \sqrt{(\mu_0 / \epsilon_0)} = 377 \ \Omega$ impedance of vacuum.

Jackson, E&M, 1975

Polarized Light; Jones Vectors

$$\vec{E}(\vec{r},t) = \vec{E}_0 \exp\left[i\left(\vec{k}\cdot\vec{r}-\omega t\right)\right]$$

• Select **k** along the z-axis. Then two field components E_x and E_y are sufficient.

$$\vec{E}(\vec{r},t) = \begin{pmatrix} E_{0x} \\ E_{0y} \end{pmatrix} \exp[i(kz - \omega t)]$$

- An EM wave is described by seven (7) real quantities:
 - Direction of wave vector (two angles ϕ and θ).
 - Magnitude of wave vector (and angular frequency).
 - Two complex amplitudes E_{0x} and E_{0y} (Jones vector).
 - One of these (absolute phase) cannot be measured; leaving six parameters.

$$\begin{pmatrix} E_{0x} \\ E_{0y} \end{pmatrix} = E_0 \begin{pmatrix} X \exp i\Delta_X \\ Y \exp i\Delta_Y \end{pmatrix} = E_0 \begin{pmatrix} \sin \psi \exp i\Delta \\ \cos \psi \end{pmatrix} \exp i\Delta_y$$

- We don't care about the light intensity and the absolute phase.
- ψ and Δ are called the ellipsometric angles; describe polarization of wave.
- ψ =arctan(X/Y); Δ=Δ_X-Δ_Y; ρ=tanψexp(iΔ);

J. Humlicek, in Tompkins & Irene (Handbook of Ellipsometry)



Solids Lecture 3



Polarized Light; Jones Vectors

The polarization state of polarized light can be described with two parameters ψ and Δ called ellipsometric angles.

H. Fujiwara

Polarization Ellipse $\vec{E}(z=0,t) = E_0 \begin{pmatrix} \sin\psi \exp i\Delta \\ \cos\psi \end{pmatrix} \exp[-i\omega(t-\tau)t]$

At z=0, the electric field vector traces out an ellipse.



Parameters of the ellipse:

• Azimuth ϑ

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 Ratio tanγ major/minor axis Ellipticity e=tanγ=B/A can be calculated from ψ,Δ.

J. Humlicek

Representation of Polarized Light by Complex Numbers

 $\vec{E}(z=0,t) = \frac{E_0}{\cos\psi} \left(\frac{\sin\psi \exp i\Delta}{\cos\psi} \right) \exp[-i\omega(t-\tau)t]$



$$\chi = \frac{\exp i\Delta}{\tan \psi} = \frac{\tan \vartheta + i \tan \gamma}{1 - i \tan \vartheta \tan \gamma}$$

Complex number χ is related to ellipticity and azimuth of the polarization ellipse.

Also Jones ratio:

$$\rho = \tan \psi \exp i\Delta = \frac{E_x}{E_y}$$

J. Humlicek





Decoherence and Depolarization

$$\vec{E}(\vec{r},t) = \vec{E}_0 \exp\left[i\left(\vec{k}\cdot\vec{r}-\omega t\right)\right]$$

In practice, light sources are superpositions with several frequencies, called **wave packets**.

Similarly, light sources have mixed polarization states.





Stokes Parameters

$$\vec{E}(\vec{r},t) = \vec{E}_0 \exp\left[i\left(\vec{k}\cdot\vec{r} - \omega t\right)\right]$$

$$S_{0} = I_{x} + I_{y} = E_{x}E_{x}^{*} + E_{y}E_{y}^{*}$$

$$S_{1} = I_{x} - I_{y} = E_{x}E_{x}^{*} - E_{y}E_{y}^{*}$$

$$S_{2} = I_{45^{\circ}} - I_{-45^{\circ}} = E_{x}E_{y}^{*} + E_{x}^{*}E_{y}$$

$$S_{3} = I_{R} - I_{L} = 2 \operatorname{Re}(E_{x}^{*}E_{y})$$

Total intensity s-polarized minus p-polarized Diagonal difference Right minus left circular

$$0 \le p = \frac{\sqrt{S_1^2 + S_2^2 + S_3^2}}{S_0} \le 1$$
$$\tan(2\vartheta) = \frac{S_2}{S_1}$$
$$\tan(2\gamma) = \frac{S_3}{\sqrt{S_1^2 + S_2^5}}$$

Degree of Polarization (%)

The Stokes parameters are related to the azimuth ϑ and ellipticity γ of the polarization ellipse.



H. Fujiwara

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Poincare Sphere



 $S_{1} = \cos 2\gamma \cos 2\vartheta = -\cos 2\psi$ $S_{2} = \cos 2\gamma \sin 2\vartheta = \sin 2\psi \cos \Delta$ $S_{3} = \sin 2\gamma = -\sin 2\psi \sin \Delta$

The Stokes parameters for completely polarized light, taken as coordinates, define a point on the surface of a sphere.

Poles: Circularly polarized Equator: Linearly polarized

H. Fujiwara



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Partially Polarized Light



(a) Totally polarized light (b) Partially polarized light (c) Unpolarized light

Totally polarized light: point on the surface of Poincare sphere. Partially polarized light: region in the sphere. Completely unpolarized light: Point in the center of the sphere.



Jones Matrix

Optical element	Corresponding Jones matrix	
Linear polarizer with axis of transmission horizontal ^[1]	$\begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}$	
Linear polarizer with axis of transmission vertical ^[1]	$\begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}$	
Linear polarizer with axis of transmission at $\pm 45^{\circ}$ with the horizontal ^[1]	$rac{1}{2}inom{1}{\pm 1}inom{\pm 1}{\pm 1}$	
Quarter-wave plate with fast axis vertical ^{[2][note 1]}	$e^{rac{i\pi}{4}}inom{1}{0}inom{0}{-i}$	
Quarter-wave plate with fast axis horizontal ^[2]	$e^{-rac{i\pi}{4}}inom{1}{0}inom{0}{i}$	
Half-wave plate with fast axis at angle $ heta$ w.r.t the horizontal axi	$_{\rm S}^{[3]} = e^{-rac{i\pi}{2}} \begin{pmatrix} \cos^2 heta - \sin^2 heta & 2 \\ 2\cos heta\sin heta & \sin heta \end{pmatrix}$	$\left(2\cos\theta\sin\theta\right)^2 \theta - \cos^2 heta ight)$



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Ellipsometry Measurement (simplified)



Ellipsometry: How does it work?



Ellipsometry Instrumentation



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Mueller Matrix Ellipsometry



Mueller Matrices

Isotropic sample, no depolarization

$\mathbf{J}_{sample} = \begin{bmatrix} r_p & 0\\ 0 & r_s \end{bmatrix}$	s	s
$\mathbf{M}_{sample} = \begin{bmatrix} 1 & -N & 0 & 0 \\ -N & 1 & 0 & 0 \\ 0 & 0 & C & S \\ 0 & 0 & -S & C \end{bmatrix}$	$N = \cos(2\psi)$ $S = \sin(2\psi)\sin(\Delta)$ $C = \sin(2\psi)\cos(\Delta)$ $N^{2} + S^{2} + C^{2} = 1$	Standard ellipsometry:Thickness measurements of thin filmsOptical functions of isotropic materials
$\rho = (\rho_{real} + i\rho_{imag}) = \frac{r_p}{r_s} = \tan(\psi$	$)e^{i\Delta} = \frac{C+iS}{1+N}$	This Mueller matrix depends only on 2 parameters

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O. Arteaga

Summary

- Fourier series and Fourier transforms
- Plane waves
- Maxwell's Equations in vacuum (general and plane-wave format)
- Polarized light
 - Jones and Stokes vectors
 - Polarization ellipse
 - Poincare sphere
- Ellipsometry experiments
 - Jones and Mueller matrices





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