Modelling optical response of periodic nanosurfaces using FEM

Modelling of spectroscopic Mueller matrices of ordered nanoplasmonic surfaces using the finite element method

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Master thesis presentation, 15. November 2017

Outline

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Problem formulation
Achievement goals

- Simulate optical properties of periodic plasmonic nanosurfaces
    - Reflection from sample at oblique polar angle of incidence and full azimuthal angle rotation.
- Retrieve the sample Mueller matrix.
- Verify model by reproducing experimental results, mainly by comparing Mueller matrices.
- Apply model to other interesting structures.

Outline

Why metasurfaces?

- Fifth item.

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- Retardation effects; i.e. kan løse for bølgelengder på størrelse med nanostrukturen

Mueller-Stokes formalism

- short explanation

Plasmonics

Localized surface plasmon resonances

- Plasmonics explores how EM fields may be confined and/or enhanced over sub-wavelength dimensions
- This effect is based on the interaction between light and the conduction electrons at a metallic interface or metallic nanoparticle
- A *localized surface plasmon resonance* (LSPR) is a non-propagating excitation of the electron plasma of metallic nanostructures coupled to an incident EM field
  - Time-varying E-fields associated with light waves exert a force on the electrons within a metallic nanoparticle and drive them into oscillation. At specific frequencies this oscillation is resonantly driven to produce a very strong charge displacement and associated field concentration, known as a LSPR.

Rayleigh anomalies

- Basic intuitive explanation of concept

Rayleigh anomalies

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- Rayleigh-line equation

Finite element method (FEM)

- Basic intuitive explanation of concept
- pretty pictures

Outline

Gold particles on a substrate

Gold hemispheroidal particles on SiO_2 substrate

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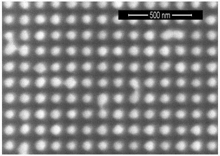
- Sample 6



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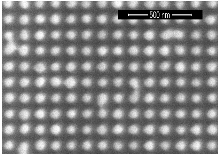

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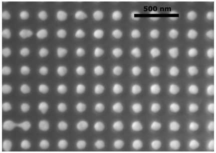

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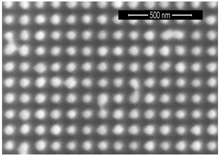

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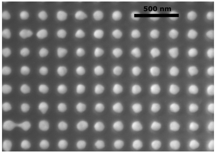
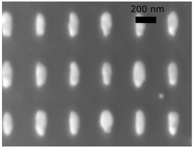


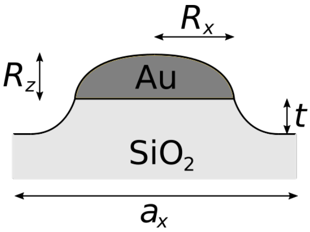
- Sample 5B



Sample geometries

Dielectric mound

- Each Au particle on all samples lie on top of a dielectric mound



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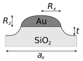

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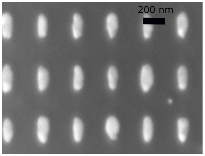
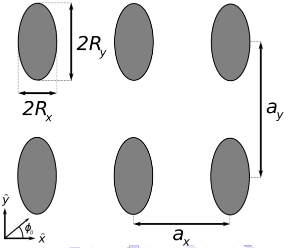
- Height of mound varies for each sample

Parameters

- Reflection from sample at oblique angle of incidence and full azimuthal angle rotation
  - Incident photon wavelengths $\lambda = 210 - 1700$ nm
  - Incident polar angle $\theta_0 = 55°$
  - Incident azimuthal angle $\phi_0 = 0° - 360°$





Outline

COMSOL model overview

1. Build the geometry

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2. Physics setup

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3. Meshing

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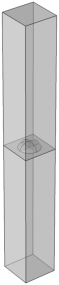
5. S-parameter conversion

COMSOL model overview

The nanostructures are modelled as unit cells with periodic boundary conditions simulating infinite periodicity



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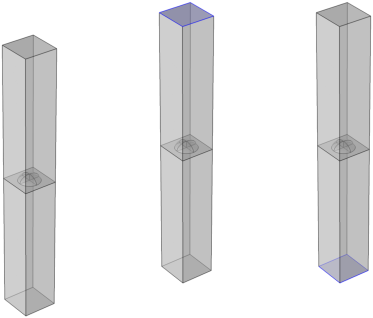

port 1 port 2

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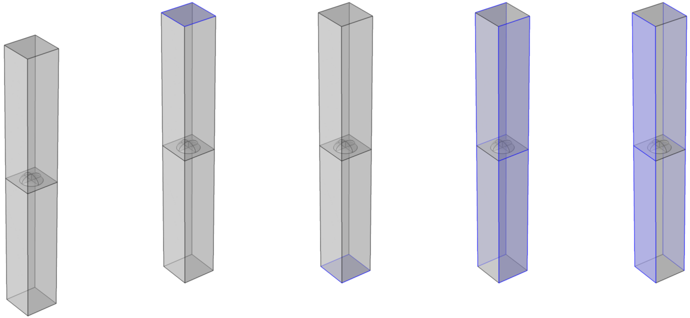

port 1 port 2 Floquet periodicity

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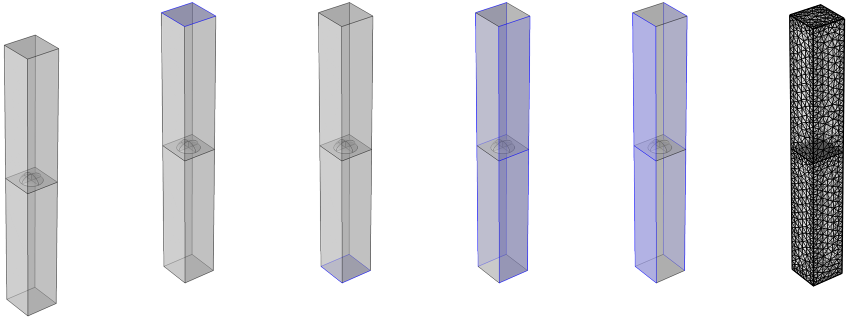
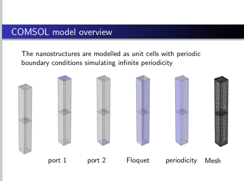


port 1 port 2 Floquet periodicity Mesh



- Porter ved utgang/inngang til modellen
- de eksiterer bølgen (inngang) og absorberer den påvei ut, samt leser av data i form av S-parametre
- Port 1 eksiterer og leser av reflekterte bølge
- Port 2 leser av transmitterte bølge
- Periodiske grensebetingelser som etterlikner den ordnete strukturen
- (Floquet periodisitet forsikrer en faseskift mellom de tangentielle bølgevektor-komponentene k_x, k_y. Faseskiftet bestemmes av en bølgevektor og avstanden mellom kilde og destinasjon.)
- Diskretisere geometrien til et mesh, der hvert element løses i henhold til FEM teori
- Dette er absolutt minstekrav for å modellen, skjelettet om du vil

COMSOL model overview

- Study steps:
    - $\lambda = 210 - 1700$ nm, with stepsize $\Delta\lambda = 5$ nm
    - $\phi_0 = 0^\circ - 180^\circ$, with stepsize $\Delta\phi_0 = 5^\circ$

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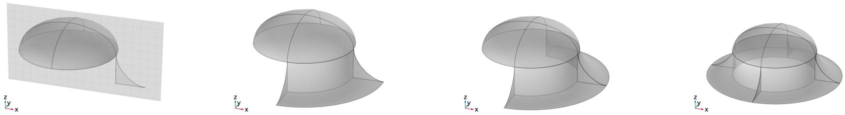
- Videre må vi bestemme hva som skal studeres.
- Study steps setter opp for hvilke betingelser modellen skal løses for
- Vi setter liknende parametersveip som ellipsometriet brukt til å karakterisere de eksperimentelle prøvene
- Tilstrekkelig åløse opp til 180° pga symmetri
- De 4 refleksjonskoeffisientene krever TE og TM

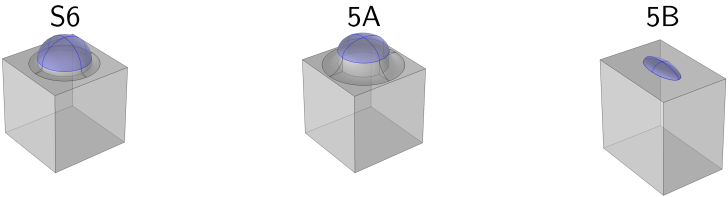
Outline

Unit cell geometries

- Mound geometry:



- Particle+mound+substrate:



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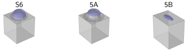
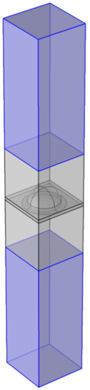
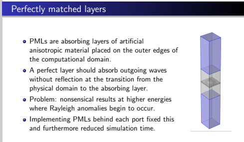


Perfectly matched layers

- PMLs are absorbing layers of artificial anisotropic material placed on the outer edges of the computational domain.
- A perfect layer should absorb outgoing waves without reflection at the transition from the physical domain to the absorbing layer.
- Problem: nonsensical results at higher energies where Rayleigh anomalies begin to occur.
- Implementing PMLs behind each port fixed this and furthermore reduced simulation time.





- PMLs består av flere lag av kunstig anisotropt absorberende materiale; en bølge som propagerer gjennom lagene vil bli eksponensielt dempet.
- Lagene ligger på utsiden av det fysiske domenet, altså på baksiden av portene
- PMLs kan på en måte replikere uendelig rom (infinite space)
- Problemet: Tror det oppstod uønsket numeriske refleksjoner ved portene pga diffraksjonsmodene

Optimization

- FEM effective at full-wave solutions for single wavelengths, but not so much at solving several hundred iterations of it.
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Optimization

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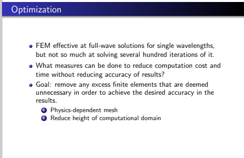
- What measures can be done to reduce computation cost and time without reducing accuracy of results?
- Goal: remove any excess finite elements that are deemed unnecessary in order to achieve the desired accuracy in the results.
    1. Physics-dependent mesh
    2. Reduce height of computational domain



- Fant raskt ut at simuleringstidene var upraktisk lange.
- Helt i begynnelsen tok det fort 24 timer for både TE og TM å løse $\approx$ 280 iterasjoner av bølgelengde for EN innfallsvinkel
-
- Kan ikke forsøke åforbedre algoritmene innebygd i programmet ettersom den er hard-coded i selve COMSOL
- Antar uansett at den er rimelig godt optimalisert av COMSOL-folka
-
- Ha sågrovt mesh som mulig uten at det påvirker resultatet
-
- To forbedringer som har prøvd å   håndtere dette
- 1. Mesh mht det fysiske systemet og ikke bare udiskrimenerende uniformt fin-detaljert mesh over hele modellen
- 2. Hvis man kan redusere høyden til luft/substrat domenene uten at det påvirker resultatet vil det kraftig redusere antall elementer

Physics-dependent mesh

A physically reliable solution requires a mesh that can properly resolve the wavelength, i.e. the maximum mesh element size should be limited to a fraction of the wavelength.

1. Mesh should take into account local material properties
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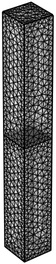

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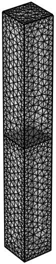

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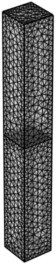
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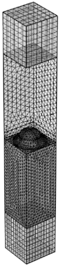

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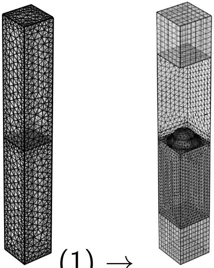

(1) → (2) →

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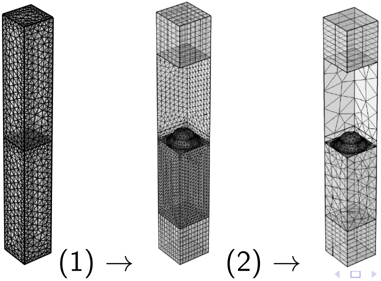
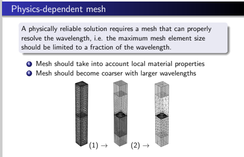
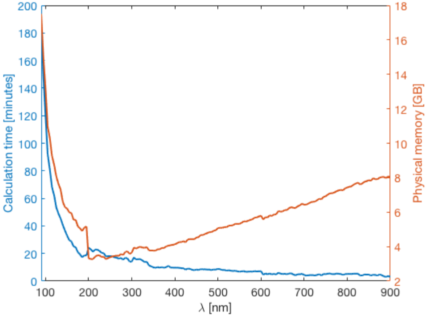




- (1) lokale egenskaper til materiale, avhengig av om elementet befinner seg i luft, glass eller gull.
- Som standard auto-genererer COMSOL et uniformt mesh med ønsket detaljenivå
- Vi velger at enhvert element ikke kan bli større enn $\frac{\lambda}{6n_i}$
-
- (2) Videre vil vi oppdatere mesh for hver ny λ slik at MES blir større/grovere for hver iterasjon
- (210 nm $\rightarrow$ 800 nm)
- Dette vil ha stor effekt på parametersveip over stort spektrum
- Hadde også lyst til at MES tar hensyn til dispersive brytningsindekser, slik at MES $= \frac{\lambda}{6n_i(\lambda)}$, men nåvaerende versjon av COMSOL støttet ikke for at mesh size parameteren kunne bli avhengig av variabler eller funksjoner.

Example: tilted GaSb cones

Wavelength-dependent mesh



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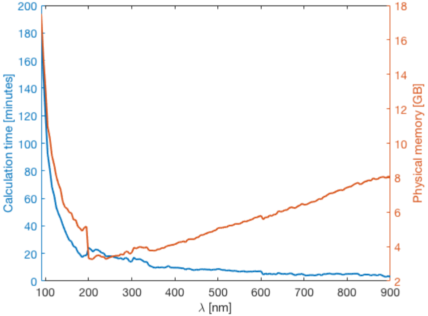
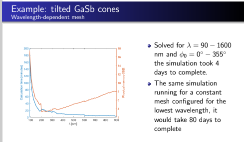
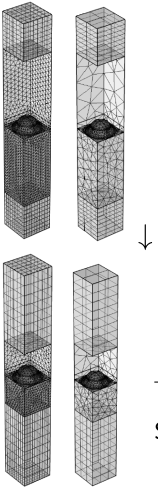


- Solved for $\lambda = 90 - 1600$ nm and $\phi_0 = 0^\circ - 355^\circ$ the simulation took 4 days to complete.
- The same simulation running for a constant mesh configured for the lowest wavelength, it would take 80 days to complete



- en modell som ble simulert for bølgelengder 90-1600 nm
- Såpass krevende simulering at den måtte deles opp i tre intervaller av bølgelengder
- Mesh ble oppdatert for hver iterasjon av bølgelenge
- Tydelig at dette har stor påvirkning på total simuleringstid
- Skulle dette bli simulert med MES $= \frac{\lambda_{\text{min}}}{6n_i}$ for alle bølgelengder ville hele prosessen tatt 80 dager! I motsetning til 4 dager.
- ikke mulig å simulere ned til 90 nm uten volumreduksjon (neste slide)

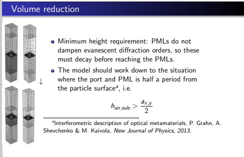
Volume reduction



- Minimum height requirement: PMLs do not dampen evanescent diffraction orders, so these must decay before reaching the PMLs.
- The model should work down to the situation where the port and PML is half a period from the particle surface[a], i.e.

$$h_{\text{air,sub}} > \frac{a_{x,y}}{2}$$

[a] Interferometric description of optical metamaterials, P. Grahn, A. Shevchenko & M. Kaivola, *New Journal of Physics, 2013.*



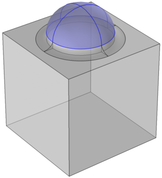
- Målet er å redusere volum av det fysiske domenet slik at antall elementer å kalkulere blir kraftig redusert.
- Hvor lite kan domenet vaere uten at man får feil resultat?
- For nanostrukturer med sub-wavelength enhetsceller burde modellen virke så lenge avstanden fra refleksjonspunkt til port er en halv gitterkonstant.
- Størst effekt for lavere bølgelengder: ser på figuren at elementer endrer seg mest for 210 nm mesh enn for 800 nm mesh

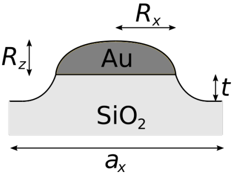
Optimization summary

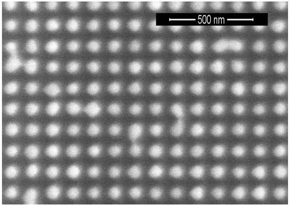
- *PMLs* fixed errors at higher frequencies and reduced computation time.
- *Material-dependent mesh* removed unnecessary elements and thus reduced computation time.
- *Wavelength-dependent mesh* greatly reduced computation time for larger wavelengths.
- *Volume reduction* greatly reduced computation time for lower wavelengths.

Outline

Sample 6

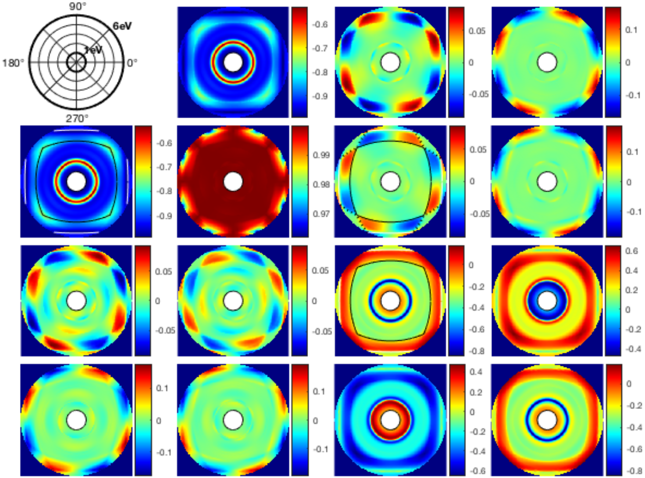




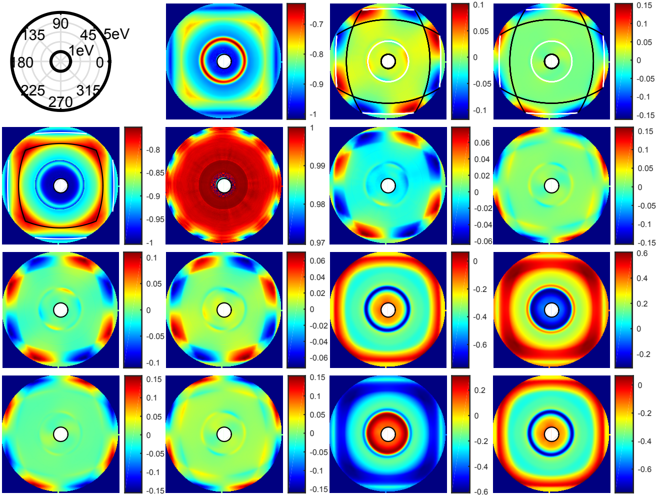


	Sample 6
a_x	125
a_y	125
R_x	38
R_y	38
R_z	32
t	8

The Mueller matrix



Mueller matrix of experimental sample

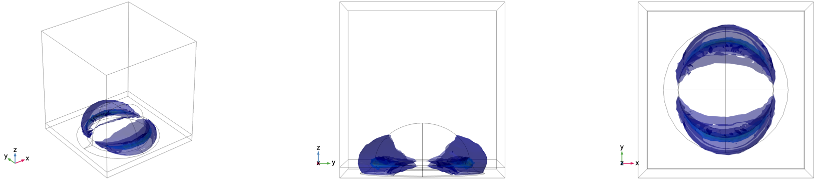


Optical response

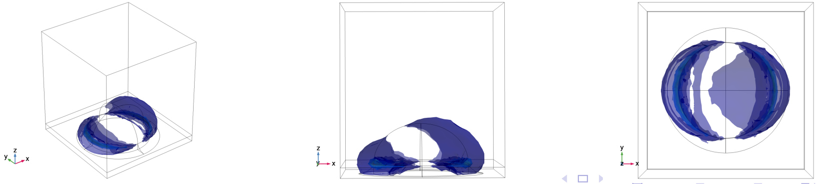
- Good correspondence between experimental and simulation results
- LSPR at around $E = 2.1$ eV, fluctuating slightly
- Pseudo-isotropic along the X- and M- points, i.e. $\phi_0 = 0^\circ, 45^\circ$, etc.
- Polarization coupling observed for azimuthal angles $\phi_0 = 22.5^\circ, 67.5^\circ$, etc around energies 2.1 eV (very weak) and at the meeting point between the extended Rayleigh line for glass and reduced Rayleigh line for air around 5.8 eV.
- Optical response at energies $E > 2.5$ eV attributed to the Rayleigh anomalies related to grazing diffracted waves.
- approximately equal conversion between both polarizations of the reflected light, $R_{sp} \approx R_{ps}$ for energies $E > 2.5$ eV

Field distribution around Au particle at LSPR

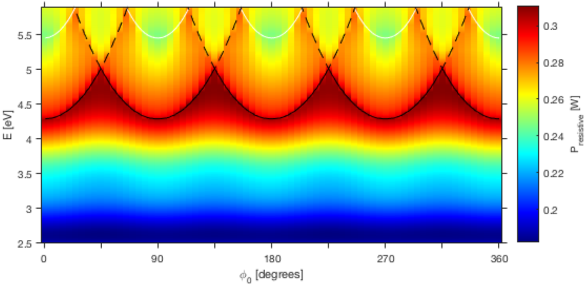
TE incident along $\hat{x}$



TM incident along $\hat{x}$



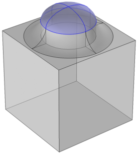
Resistive heating of Au particles

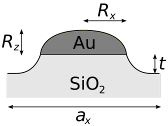


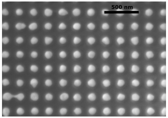
Outline

1. Introduction
   - Problem formulation
   - Motivation
2. Brief background theory
3. Model development
   - The experimental samples
   - Developing the COMSOL model
   - Improvements & optimization
4. Sample results
   - Sample 6
   - Sample 5A
   - Sample 5B
   - Tilted GaSb cones

Sample 5A

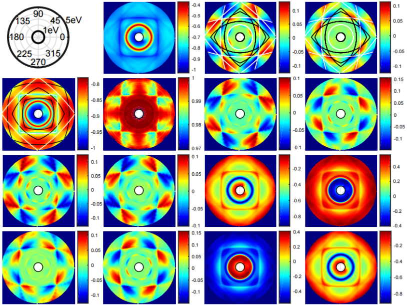




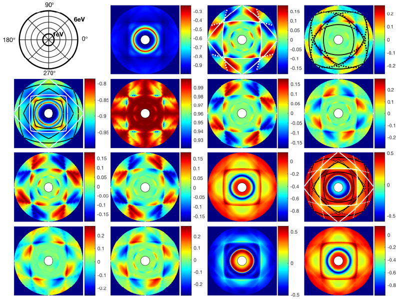


	Sample 6	Sample 5A
a_x	125	207.2
a_y	125	209.9
R_x	38	60.3
R_y	38	61.3
R_z	32	34.8
t	8	36.9

Experimental Mueller matrix



Experimental



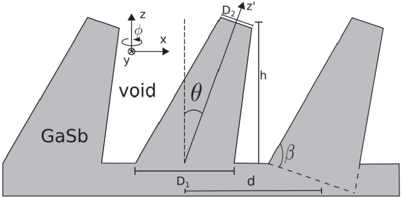
COMSOL

Outline

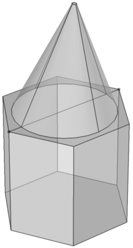
Outline

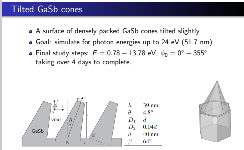
Tilted GaSb cones

- A surface of densely packed GaSb cones tilted slightly
- Goal: simulate for photon energies up to 24 eV (51.7 nm)
- Final study steps: $E = 0.78 - 13.78$ eV, $\phi_0 = 0° - 355°$ taking over 4 days to complete.



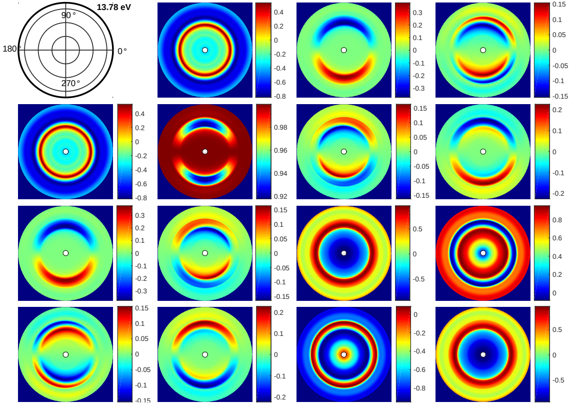
h	39 nm
θ	4.8°
D_1	d
D_2	$0.04d$
d	40 nm
β	64°





- Gallium antimonide
- Substrat og kjegler av samme materialet
- Ble karakterisert med spektroskopisk ellipsometri, fra ca. $1 - 6$ eV. Vi vil utvide dette til 24 eV om mulig.
- Samme prinsipp som før, bare med hexagonal symmetri:
- Floquet periodisitet med vegger på motsatt side av hverandre
- Utfordring å sørge for at disse veggene hadde identisk mesh samtidig som at veggkantene matchet meshet til sidemannen

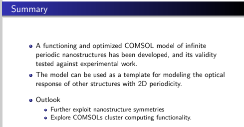
The Mueller matrix



- Polarization coupling revealed around $6 - 10$ eV

Summary

- A functioning and optimized COMSOL model of infinite periodic nanostructures has been developed, and its validity tested against experimental work.
- The model can be used as a template for modeling the optical response of other structures with 2D periodicity.
- Outlook
  - Further exploit nanostructure symmetries
  - Explore COMSOLs cluster computing functionality.



- Hovedmålet var å lage en fungerende modell i COMSOL og teste den opp mot eksperimentelle resultat (check), for så å prøve ut modellen på andre strukturer (check, dog bare én).
- Dette hadde ikke vært mulig, særlig sistnevnte, uten forbedringene som ble implementert. De reduserte beregningskostnadene åpnet også opp for mer krevende study steps og utvider mulighetene for hvilke andre strukturer den kan simulere.

For Further Reading I

A. Author.
Handbook of Everything.
Some Press, 1990.

S. Someone.
On this and that.
Journal of This and That, 2(1):50–100, 2000.