Mueller Matrix Ellipsometry – a universal optical characterization technique

Question Asked	Answer Given
For the tilted silicon columns, how does the model account for an oxide layer covering the silicon columns as it is not a uniform layer like in a case of a oxide layer present over a thin film.	Oxide layers can be treated in the same way as the column itself, meaning as another constituent in the anisotropic Bruggeman EMA. Instead of fitting a thickness, the volume fraction would be the parameter of interest. If the dimensions of the columns are known, the oxide thickness can be estimated. Grading the volume fraction in a "parametric" grading would be a way to account for changes throughout the effective layer.
Has MM ellipsometry been performed on 1D grating samples that show guided mode resonances where the excited resonances couple to the underlying substrates. Can these resonances show their presence in the MM data?	I don't know for sure, but I would assume so. Any effect that is correctly reproduced using an RCWA or FEM solver can be measured and characterized. The specific measurement geometry with highest sensitivity would depend on the sample structure but can be simulated.
If you cannot access M44, can't you obtain the same information from M24^2 + M34^2? This would be nice for use with ellipsometers with a single compensator.	We can perform full data analysis on anisotropic samples with ellipsometry data even if we cannot access the m44 element since the same information is often contained in other Mueller elements. In fact, we often only need a single sample orientation and normal incidence data to characterize the retardance and fast axis orientation since the equations for the contribution of this information to different Mueller matrix elements form an equation system that is overdetermined.
What angles of analysis do you recommend for MM ellipsometry? This example uses 45, 55, 65, and 75.	This can depend a little on your final goal. In this case, we want to be sensitive to the optical constants in the different orientations, so we gain sensitivity to this at more oblique angles (just like standard ellipsometry). Thus, a good range of 45 to 75 degrees was sensitive to the optical constants from the surface. If you know for sure that your sample is uniaxial with optical axis in the surface plane a single angle, say 65deg, would be sufficient to get a unique answer. The specific angle is not that important as long as you are not measuring exactly at the Brewster angle where the data gets a little noisy.
Do you always assume that the k ellipsoid coincides with the n ellipsoid? If so, what is the validity of that assumption?	Yes, I believe this is correct. The n and k ellipsoids coincide. However, the polarizable resonances do not have to all be along the same orientations. There is an excellent paper by the Schubert group (University of Nebraska) that we can send you that discusses this in more detail; Stefan: I am not entirely sure; this picture of an index ellipsoid works well for an orthorhombic material where absorption occurs only along the crystal axes. I assume in this case that statement would be correct. For materials with less symmetry, the picture might be a bit more complicated, check out this paper by Sturm and Grundmann on the optical properties of monoclinic Ga2O3: https://doi.org/10.1103/PhysRevA.93.053839 Maybe the answer is in there. In any case, we don't make any assumptions about any index ellipsoids. What we use to calculate the optical response of a layered system is the 4x4 transfer matrix algorithm developed by Berreman. The input quantities in that algorithm are the elements of the dielectric function tensor, or in more general terms, the 36 elements of the 6x6 magneto electric tensor representation similar to what is outlined, for example, in this paper by Arteaga:https://doi.org/10.1364/JOSAA.32.002049 The dielectric function tensor in this algorithm can have many symmetries depending on the problem at hand.
In this example, don't you get a lot of interference also from the glass substrates?	In transmission, you can go to near normal incidence and have little-to-no effect from the glass (isolating the liquid crystal)
Can the angle information of manually rotated samples be added to the dataset afterwards?	Yes, we have that capability in our software.
How do you fix polymer films during measurement? Often times such polymer films are not flat...	I always start by using the vacuum stage. If the polymer foils are too thin, they may crinkle. In this case, I may stretch them across a "cut-out" hole and tape around the edges. However, many of these problems are only witnessed for reflection-based measurements. Thus, we get plenty of benefits by using transmission-based MM-SE. It has high sensitivity to the anisotropic properties of such films AND it does not affect the transmitted light path (even when crinkled) like with reflected beam.
How do you fix polymer film samples during the measurement? Such films are often not flat...	Use vacuum or tape. For transmission measurements, it is not as critical. It is more important for reflection-based measurements.
How can you be sure that the gap between two substrates does not change after is being filled with liquid (liquid crystal). One would expect the gap to shrink when the air in the gap is replaced with liquid.	This is true. If the gap is thin enough, you may still have some coherent reflections from the glass "gap" even though it is filled with liquid crystal. This gives the most accurate measure of the liquid crystal because you can also get correct thickness of the "filled" gap. If too thick that the coherent oscillations are suppressed, then the air-gap measurement at least gives you a better nominal thickness to use in your model than just guessing based on the spacer size that was used.
Do you strongly recommend using sample rotation to get the full potential of MM ellipsometry?	If I have an automated sample rotator, I like to collect data over the full rotation. However, when I don't have access to such a stage, I can often get enough information by rotating manually to 0, 45, 90 and 135 degree sample orientations. Stefan: Many problems in transmission only require a single orientation since the problem is overdetermined. See similar question above
Regarding the case 4. Have you tried to anneal the sample to observe a transition of the a-Si optical properties, found by you, to the poly-Si ones? This should, I guess, be an indicator of the model correctness.	I don't believe this has been attempted. Stefan: assuming this refers to the GLAD films. Annealing was attempted on this films, but they tend to start to deform before a transition can occur. XRD measurements did not show any crystallinity.
I think I might have missed this, but is all of this data being collected in transmission?	For this example, yes all data is measured in transmission through this optical element
Can you please explain what is the difference between Ordinary and Extraordinary RI?	Ordinary = Refractive index in direction perpendicular to optical axis, Extraordinary = refractive index in direction along optical axis, The optical constants are different along the three principle directions: x, y and z. For a uniaxial material, two of the indexes are equal (say, nx=ny, or nx=nz, or ny=nz). Those are the ordinary index, or no. The extraordinary index is the index that is different; that would be called ne .

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Could you comment on non-ideal model parameters effect on the Mueller matrix? (spectral bandwidth, angular spread, back surface contribution, roughness) For example, is there a way to isolate depolarization coming from the bulk of a thin film?	Mueller matrix already contains the information about depolarization. In our software, we provide couple depolarization metrics: the depolarization index which requires the complete MM and Jones quality factor which does not require the complete MM. The non-ideal model parameters mentioned in the question are rather fit parameters where our modeling interest is rather than treated as noise to be removed. However, such noise or depolarization could be removed through Mueller matrix decomposition such as the Lu-Chipman, Cloude, or differential decomposition. Currently, our software does not promote these methods.
Can you give insight as to why for a uniaxial material with c-axis out of plane, ellipsometry isn't sensitive to this axis even at oblique angles? For example of the TiO2 with 001 orientation Stefan showed	The high index of refraction leads causes the beam in the material to be close to the normal even at high angles of incidence. There is only s very small fraction of the E field in the probing beam that would be along the z-direction. Check out this paper by Aspnes which estimates why the out-of-plane component can't be determined: https://doi.org/10.1364/JOSA.70.001275
Which are the ranges of the ellipsometer angles delta and psi? has a psi larger of 45 degree any sense?	Psi -> 0-90deg, Delta -> 0-360deg......Psi can be above 45deg, especially when you have interference effects caused by a thick film. The Delta parameter can cover a full 360° range (often graphed from -90 to 270). Psi can cover 90°. Yes, it is okay for Psi to be above 45 degrees, but this depends on the measurement configuration. It is common for Psi to be above 45 degrees for thin film ellipsometry experiments. You can also have Psi above 45 degrees for MM-SE in transmission for a polarizer where the s-polarized light is absorbed and the p-polarized component is transmitted. Stefan: For a substrate, a Psi value above is direct prove that the sample is anisotropic. Specifically in the IR where strong phonon modes occur, we often see this happening at the edge of the reststrahlen bands. In that case, we actually have some sensitivity to the out-of-plane optical properties caused by some phonon absorption along that direction.
Given an arbitrary MM, it is possible to recover the delta and psi uniquely?	If the sample is isotropic or pseudo-isotropic, then yes. For an anisotropic sample, Psi and Delta are insufficient to describe a general Mueller Matrix. Stefan: If the sample is anisotropic but not depolarizing, we can define similar quantities for each element of the Jones matrix, which also accounts for the cross-polarization terms. Instead of rp/rs, we define the generalized ellipsometry equations by three quantities: rpp/rss (this is very similar to the standard ellipsometry definition), rsp/rss, and rps/rpp ; for each of these we can define "Psi" and "Delta" values analog to the standard ellipsometry equation
What are the prominent limitations when evaluating structured metasurfaces?	This is hard to answer in general. The exact material combinations, dimensions, and complexity of the structure will define how sensitive we are to individual model parameters. Commercial solvers often include simulation capabilities to estimate sensitivity up front.
What ellipsometer was used for most of the data sets in this presentation? In particular, what instrument was used for the metasurface examples discussed?	RC2 was used for all data presented in this webinar
Is there an analogous system to the RC2 for responses in the IR?	We have the IR-VASE Mark II spectroscopic ellipsometer available. It is capable of measuring 12 of the 16 Mueller matrix elements. We do not have a dual-compensator system (to collect full MM) in the infrared. There is plenty of redundancy in the MM for most samples and measuring 3 columns (or 3 rows) of the MM is adequate.
what's the angle resolution of the optical axis of rutile TiO2 by RC2?	In reflection, I would guess maybe on the order of one degree, in transmission less than 0.2 deg. It depends on how different the index components in the different directions are. The angle would be relative to the surface normal, not relative to the crystal system, i.e. miscuts would be included in the reported angle.
main difference between standard spectroscopic ellipsometry and Muller matrix ellipsometry.	Standard ellipsometry assumes the off-diagonals elements of the Mueller Matrix are zero. Mueller Matrix ellipsometry measures the off diagonals with no assumptions
but you can also do analysis by SE of anisotropic materials with by measuring considering the material is anisotropic and the adequate modeling, cant you?	If the sample creates "cross-polarization" between the p- and s- waves, standard ellipsometry does not provide reliable ellipsometry data. Stefan: if you can orient uniaxial sample or biaxial sample of at least orthorhombic symmetry so, that the material crystal axes are aligned exactly with the ellipsometer coordinate system, you can use the standard ellipsometry data since no cross-polarization occurs. This was done in the early day of generalized ellipsometry development. Today, we would recommend to measure the Mueller matrix and add multiple orientations to take advantage of the off-diagonal elements in the Mueller matrix that are created by an anisotropic sample. This improves sensitivity to the different tensor elements and is direct evidence of the anisotropic nature of the sample without any assumptions or sensitivity to misalignment.
is it possible to determine the crystalline texture in a polycrystalline film using MM?	Usually you cannot do this, because the beam illuminates an area containing many crystallites with random orientation, so you only measure the average. Stefan: If there are preferred orientations, for example twinning, an effective medium approach that combines multiple orientations of the same tensor can indicate the amount of each crystal orientation in a sample and orientation. I specifically remember a case, where this was demonstrated for a polycrystalline InN film utilizing strong phonon absorption and anisotropy in the IR https://doi.org/10.1103/PhysRevB.90.195306 This is pretty hard to model correctly though.
What utility is MM analysis for instruments that cannot access all matrix elements (e.g. the Alpha-SE)?	With these instruments, you can still accurately measure the accessible elements of the Mueller matrix. This is especially necessary if the sample is anisotropic. Even when you can't measure the full MM, there is still information about the anisotropic properties and depolarization effects that appear in the MM. If you look carefully at the 16 elements of the full MM, there is plenty of redundancy for most sample types.