Optical response of nanostructured GaSb

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Optical measurements of nanostructured GaSb prepared by sputtering is presented. The optical response is studied by Mueller Matrix Ellipsometry (MME) in the visible range (430-850 nm), and by spectroscopic ellipsometry in the range 0.6-6.5 eV. The nano-structured surfaces reported in this work, consist of densely packed GaSb cones approximately 50 nm high, on bulk GaSb. The nanostructured surfaces are here shown to considerably modify the optical response of the surface, hence giving a strong sensitivity to the far field spectroscopic (Mueller matrix) ellipsometric measurements. The off-specular scattering and the depolarization is found to be low. The anisotropic response is particularly emphasized by studying nano-structured GaSb cones approximately 45 degrees tilted with respect to the surface normal. In the latter case, one observes upon rotating the sample around the surface normal, that the Mueller matrix elements m_{13} and m_{14} oscillate as a function of the rotation angle. Finally, Mueller matrix techniques have been applied to the measured data, in order to analyze the acquired Mueller matrix in terms of physical realisability and noise.

1 Introduction The field of research related to nano-structured materials, with their wide range of applications, including photonics applications, gives interesting problems also to the far field techniques such as e.g. spectroscopic ellipsometry (SE). In particular, traditional thin film properties, may be mimicked by nanostructures, and further supply new or enhanced properties. We are here particularly focusing on the optical response of nanostructured GaSb, as measured by MME and generalized SE. In this preliminary paper we primarily report the Mueller matrix measurements and their sensitivity to nano-structuration.

2 Experimental The samples were low ion energy sputtered crystalline GaSb(001). Under low ion energy sputtering conditions, one finds conditions where an apparently "smooth" surface forms, but which in reality consists of nano-structured cones (i.e. equivalent to a high quality nano-structured thin film on top of the substrate).

By sputtering at normal incidence, one could obtain typically cones normal to the sample surface (denoted "normal cones" in this paper), while by sputtering at 45 degrees incidence, with respect to the sample normal, close to 45 degrees tilted cones forms (denoted here the 45 degree cones). Such issues have been studied by Atomic Force Microscopy, Scanning Electron Microscopy (SEM), and Transmission Electron Microscopy (TEM) [1-3].

The optical far field measurements were performed using a commercial Photo-Elastic-Modulator Spectroscopic Ellipsometer (PMSE) in the range 0.6-6.5 eV (UVISEL), at 55 degrees angle of incidence. The complete Mueller matrix was also measured using a commercial ferroelectric liquid crystal retarder based Mueller matrix Ellipsometer (MM16) in the range 850-430 nm (1.46-2.88 eV), at 70° angle of incidence. The sample orientation with respect to the incoming beam was carefully recorded, and the sample was rotated manually in steps of 45°, with a total sample rotation in all cases of at least 360 degrees.

The PMSE measurements were performed in the UVISEL setup, polarizer-sample-PEM-analyzer, where the angle of the fast axis of the PEM with respect to the analyzer is fixed to 45°. Measurements were performed in the standard PMSE configurations (M = 0°, A = 45°), determining $I_s = -m_{43} = \sin2\Psi\sin\Delta$ and $I_{c1} = m_{33} = \sin2\Psi\cos\Delta$. For the normal cones, measurements were additionally performed in the configuration (M = 45°, A = 45°), determining $I_{c2} = -m_{12} = \cos2\Psi$. In the case of the 45 degrees tilted cones, a complete set of 8 measurements were performed, in order to determine the 3 first columns of the full Mueller matrix [4, 5].

3 Results and discussion Figure 1 shows standard PMSE measurements of the nano-structured sample for the "normal cones". The "normal cones" were determined from high resolution-TEM to be approximately 50 nm tall. Figure 1 is of major importance, since it demonstrate that spectroscopic ellipsometry is highly sensitive to nanostructuration. Furthermore, it is evident from Fig. 1 that these nanostructured surfaces strongly modify the optical response of the system.
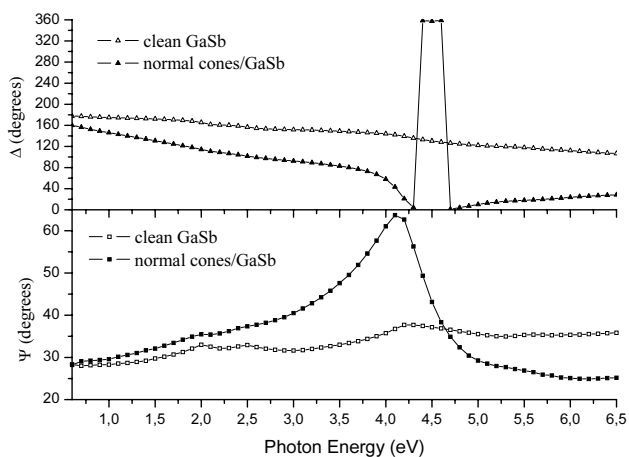

Figure 1 Measured Ψ and Δ from c-GaSb with approximately 7 nm oxide (hollow symbols), and normal GaSb cones on bulk GaSb (full symbols).

The sample depolarization was found to be low, and negligible below 3.5 eV (see Fig. 2). In fact, using He-Ne laser light, and a goniometer, the angular scattering was measured, and it was found to be similar to the one from c-Si. Hence, the nano-structured surface is expected to be well modelled within an appropriate effective medium approximation. Indeed, the low off specular scattering, was correlated to negligible depolarisation in the visible. The latter was confirmed by Lu-Chipman product decomposition [6] of the Mueller matrix measured in the visible. As a result of the decomposition, the total depolarisation was determined from the diagonal elements of the depolarization matrix. For photon energies above 4 eV, an apparent trend of depolarization was observed for the normal cones,

while not observed for the tilted cones. The "degree of polarization" as defined from

$$p = \sqrt{I_s^2 + I_{c1}^2 + I_{c2}^2} \qquad (1)$$

is shown in Fig. 2. Equation (1) is applicable only when the Jones matrix is diagonal, i.e. for normal cones, or when the cones lie in the incidence plane. The degree of polarization for the tilted cones, was thus only calculated from the PMSE data (Mueller elements m_{12}, m_{33} and m_{43}) corresponding to the cones in the incidence plane (see also below).Since the spacing of the cones and the typical cone sizes are much smaller than the wavelength of light, it is speculated that the depolarization arises rather from the statistical distribution of the cones, and appears to follow a trend similar to Rayleigh scattering, and such an effect is possibly more pronounced for the normal cones. The dip in the degree of polarisation around 4 eV for the normal cones, may possibly be a result of small ripple effects in the "layer thickness" (cone height) across the measurement spot [4]. This dip coincides with the photon energy where $I_c = 1$ and $I_s = 0$.
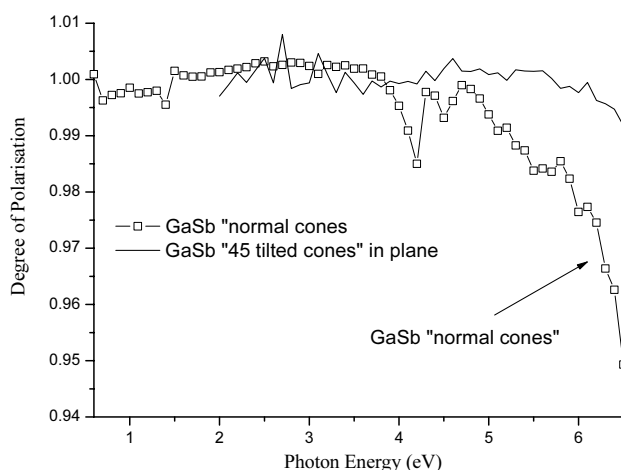
Figure 2 The degree of polarization (p), as defined by equation (1), for the normal cones (hollow squares), 45 degree cones (full line) .

The 45 degrees tilted cones, were both measured by complete visible MME, and by generalized PMSE. The complete Mueller matrix at 450 nm (2.755 eV), as a function of rotation of both the "normal cones" and the "tilted cones" samples is shown in Fig. 3. The accuracy of the manual rotation of the sample was estimated to +/- 5 degrees. The movement of the ellipsometric spot was carefully monitored to remain within a uniform sample area. Evidently, the oval shape of the ellipsometric spot makes it impossible to probe the exact same sample area upon rotation.The Mueller matrix was normalized by m_{11}. It is as expected, observed from Fig. 3, that there is very little change in the MM elements upon rotation of the "normal cones". On the other hand, large variations in the MM elements are observed as a function of rotation of the sample with the "45 degrees tilted cones". In particular, it is observed that the elements m_{13} and m_{14} are quite smooth
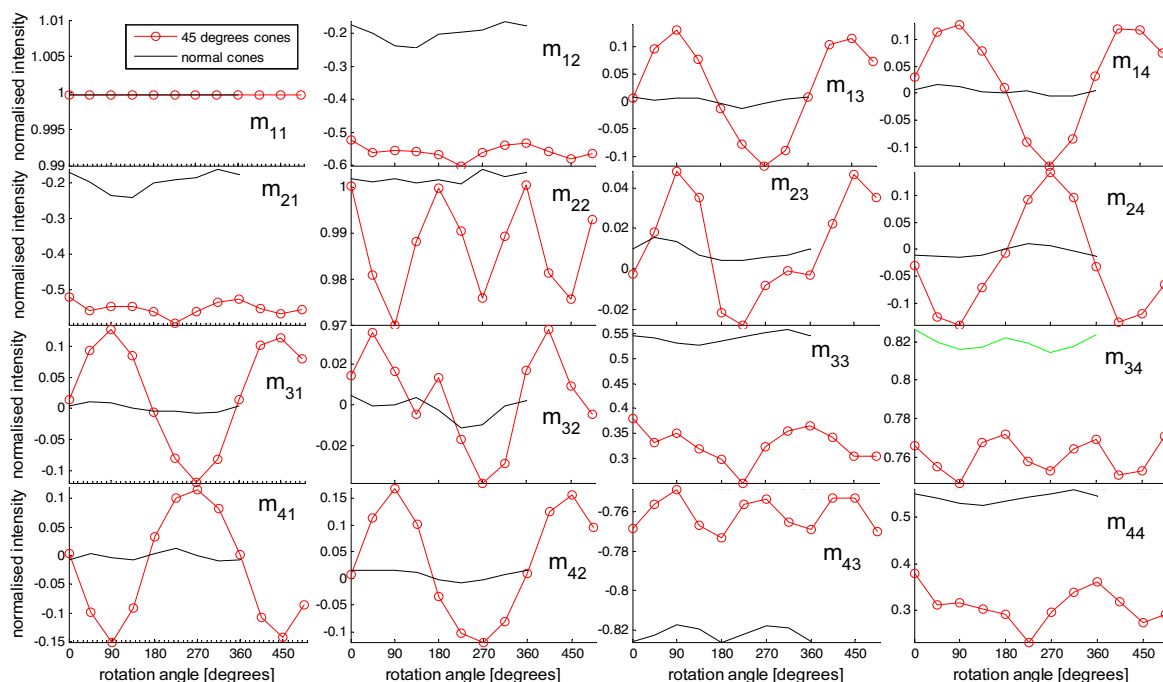
Figure 3 The measured (normalized) Mueller matrix at 450 nm (2.755 eV) as a function of sample rotation around the sample normal, for "normal cones" (full line), and 45 degrees tilted cones (hollow circles).

oscillating functions of the sample rotation. Both m_{13} and m_{14} are zero when the cones lie in the incidence plane, while they are maximum and anti-symmetric for the cones pointing in a plane perpendicular to the incidence plane. Details of the analysis and the estimated multilayer anisotropic effective medium models and fits, in direct comparison to SEM data, will be reported elsewhere [3].

Generalized PMSE was also used to characterize the "45 degrees tilted cones", in the range 0.6-6.5 eV. A set of sufficient configurations were recorded, as a function of the sample being rotated around the sample normal, thus allowing the three first columns of the Mueller matrix to be determined for each angle of rotation. The normalised Mueller matrix for a general non-depolarising anisotropic sample may be written as:

$$m_{ij}^J = \frac{1}{2}Tr\left(J^\dagger \sigma_i J \sigma_j\right) \quad (1)$$

where σ_i are the Pauli matrices here defined as in the work by Cloude [7], and J is the normalised Jones matrix [8]. This matrix is described by the complex reflections coefficients:

$$J = \frac{1}{r_{ss}}\begin{bmatrix} r_{pp} & r_{ps} \\ r_{sp} & r_{ss} \end{bmatrix} = \begin{bmatrix} \gamma_{pp}e^{i\delta_{pp}} & \gamma_{ps}e^{i\delta_{ps}} \\ \gamma_{sp}e^{i\delta_{sp}} & 1 \end{bmatrix} \quad (2)$$

where $\gamma_{pp}=|r_{pp}|/|r_{ss}|$, and δ_{pp} is the complex argument of r_{pp}/r_{ss}. The other quantities are defined in an equal manner. The complete normalised Mueller-Jones matrix only depends on these six parameters, whereas the missing elements of the measured Mueller matrix can then be found

by solving a set of equations, as described by Jellison and Modine [4]. The last column of the Mueller matrix in Fig. 4, shows the result of the calculated Mueller-Jones matrix, using the latter approach (hollow circles). It is particularly observed that the elements m_{14} and m_{24} are noisy and not well determined. The method of solving the set of equations appears thus in some cases to give unstable solutions (probably due to poor conditioning). An alternative approach that may better handle noise and small amounts of depolarisation has been developed. It is here proposed to fit the normalized complex Jones elements as a function of wavelength, to the measured 3 first columns of the Mueller matrix, by minimization of :

$$\alpha^2 = \sum_{i=1}^{4}\sum_{j=1}^{3}\left(m_{ij}^{exp} - m_{ij}^J\right)^2, \quad (3)$$

where the Mueller-Jones elements m_{ij}^J are calculated from the fitted elements of J, according to Eq. (1). Figure 4 also shows the latter estimation of the Mueller-Jones matrix. It is observed from the last column in Fig. 4, that this procedure appears more numerically stable than the direct solution from the set of equations. It is further observed from Figure 4, that a particular enhanced sensitivity to the nanostructuring of GaSb is found around 4 eV. The Mueller matrices measured from both the visible-MME and estimated from generalized SE, were tested for their physical realisability, using the method described by Cloude and Pottier [9]. The entropy and physical realisability of the Mueller matrices, as defined by Cloude et al. [9], was used to determine any possible depolarization effects. For the

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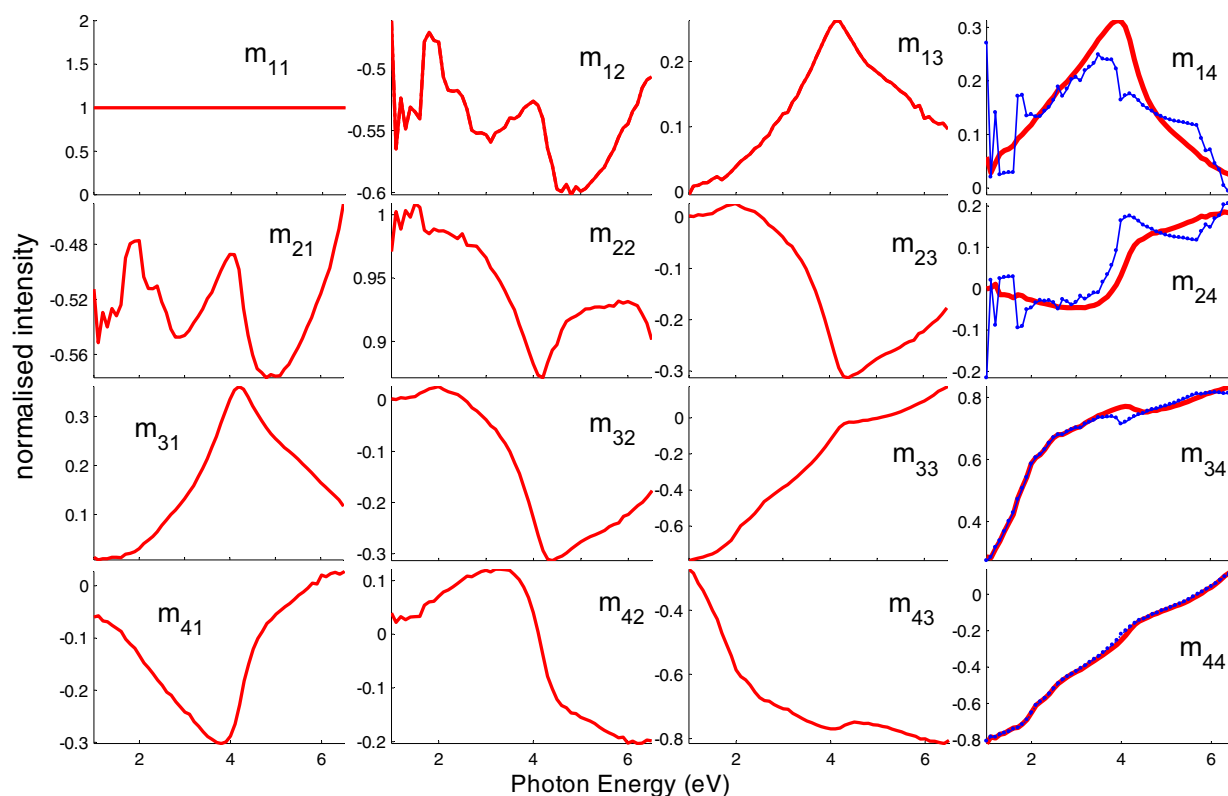
Figure 4 Mueller-Jones matrix of the "normal cones" derived from the 12 elements from generalized PMSE, at 55° angle of incidence. The last column shows the result of solving the set of equations as in Ref. [4] (hollow circles, blue), while the full curves (red) are the result of the proposed method based on fitting the normalised elements of the Jones Matrix, and generating the last column of the Mueller-Jones Matrix.

visible MME measurements in the limited range 1.46-2.88 eV, the entropy was found to be particularly low, and the physical realisability good. Low entropy indicates no depolarization, while the physical realisability indicates little measurement noise. This was also the case for the PMSE Mueller-Jones matrices in the energy range from 1 to 2.5 eV. In the range 2.5 eV to 6.5 eV, the physical realisability was within reasonable limits, i.e. accepting the matrices as proper Mueller matrices that allow detailed analysis, but a higher noise level might hide minor depolarizing effects.

4 Conclusion Spectroscopic ellipsometry and generally Mueller matrix ellipsometry have been shown to be a useful technique for the characterization of nanostructured surfaces such as nanocones of GaSb on GaSb.

References

[1] S. Facsko, H. Kurz, and T. Dekorsy, Phys. Rev. B **63**, 165329 (2001).
[2] N. Brun, E. Søndergård, A. Lelarge, and E. Barthel, to be submitted.
[3] I. S. Nerbø, M. Kildemo, E. Søndergård, L. Holt, J. Walmsley, and I. Simonsen, manuscript in preparation.
[4] G. E. Jellison, Jr. and F. A. Modine. Appl. Opt. **36**, 8190 (1997).
[5] A. Laskarakis, S. Logothetidis, E. Pavlopoulou, and M. Gioti. Thin Solid Films **455/456**, 43 (2004).
[6] S. Lu and R. A. Chipman. J. Opt. Soc. Am. A **13**, 1106 (1995). S. Lu and R. A. Chipman. J. Opt. Soc. Am. A **11**, 766 (1993).
[7] S. R. Cloude, in: Polarization Considerations for Optical Systems II, edited by R. A.Chipman, Proc. SPIE **1166**, 177 (1989).
[8] P. S. Hauge, R. H. Mueller, and C. G. Smith, Surf. Sci. **96**, 81 (1980).
[9] S. R. Cloude and E. Pottier, Opt. Eng. **34**, 1599 (1995).