Phonons, Optical Constants, and Composition Determination of $In_xGa_{1-x}As_{1-x}N_y$

G. Leibiger[1*], V. Gottschalch[1]
M. Schubert[2]

[1]University of Leipzig, Faculty for Chemistry and Mineralogy, Linn¾str. 3, 04103 Leipzig, Germany
[2]University of Leipzig, Faculty for Physics and Earthsciences, Linn¾str. 5, 04103 Leipzig, Germany

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*E-mail: pge97jrk@studserv.uni-leipzig.de

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Motivation

→ InGaAsN as new material for long-wavelength Lasers and high-efficiency solar cells

→ Optical constants are needed for precise device design.

→ X-ray diffraction fails to give relyable nitrogen- and indium concentrations, which are prerequisite for a better understanding of the complex MOVPE growth mechanism.

→ Phonon properties of InGaAsN are still unknown.

Outline

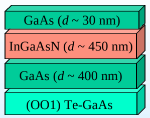
→ Deposition of $In_xGa_{1-x}As_{1-y}N_y$ (d ~ 450 nm, x ~ 0.1, y < 0.03) single layers on GaAs substrates using metal-organic vapor-phase epitaxy (MOVPE)

→ Derivation of complex dielectric functions for 0.75 eV ≤ E ≤ 1.3 eV and 100 cm^{-1} ≤ ω ≤ 600 cm^{-1} using near (NIR)- and far (Fir)-infrared spectroscopic ellipsometry (SE), respectively

→ Two-mode phonon behaviour (GaAs- a. GaN-like phonon)

→ Calculation of nitrogen and indium concentrations combining the results from high-resolution x-ray diffraction (HRXRD) and FIRSE

Samples/MOVPE

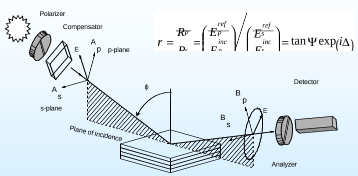


N- and In-Concentrations

Sample	A	B	C	D	E
x_{In}	0.09	0.11	0.11	0.12	0.09
y_N	0.013	0.019	0.022	0.024	0.029

→ Precursors: TMGa; TMIn; Arsine; 1,1-DMHy
→ Growth temperatures: T_G = 560-600ºC
→ Reactor pressure: p_{tot} = 50 mbar
→ V/III ratios: V/III = 110-180
→ Gas flow: f_{tot} = 7 l/min
→ Carrier gas: H_2

Ellipsometry



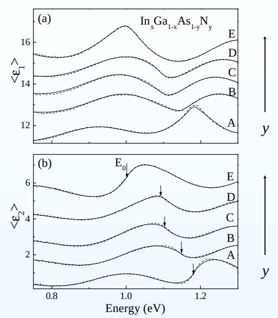
$\Psi, \Delta = f[d_j, \varepsilon_j(\omega), \Phi_a] \rightarrow d_j, \varepsilon_j(\omega)$, model parameter

Model calculation
d…layer thickness
ε…dielectric function
Φ…angle of incidence

NIR-Ellipsometry

Pseudodielectric Function

$$\langle\varepsilon\rangle = \{[(1-\rho)/(1+\rho)]^2 \sin^2\Phi_a + \cos^2\Phi_a\}\tan^2\Phi_a$$



→ redshift of E_o with increasing y

Model Dielectric Function

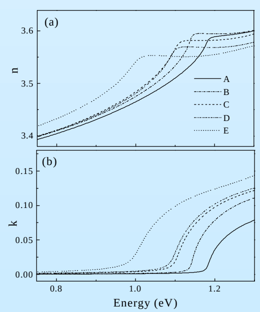
$$\varepsilon(E) = \varepsilon_0(E) + \varepsilon_{\Delta 0}(E) + c + d\cdot E^2 + f\cdot E^4$$

$$\varepsilon_j(E) = A_j E_j^{-1.5}(\chi_j^{-2}[2 - (1+\chi_j)^{0.5} - (1-\chi_j)^{0.5}])$$

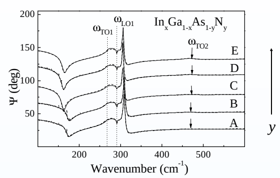
with $\chi_j = (E_j + i\Gamma_j)/E_j$ [j = "0", "Δ_0" for E_0 and $E_0+\Delta_0$, respectively].

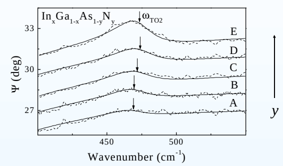
$\varepsilon_j(E)$ can be found, e. g., in S. Adachi, *Physical Properties of III-V Semiconductor Compounds* (Wiley, New York, 1992).

Optical Constants



FIR-Ellipsometry





→ two-mode phonon behaviour: GaAs-like (ω_{TO1} ~ 267 cm^{-1}) and GaN-like phonon (ω_{TO2} = 469...474 cm^{-1})

→ blueshift of ω_{TO2} with y due to alloying ($\omega_{TO}^{\beta\text{-GaN}}$ = 553 cm^{-1}) and compressive biaxial strain → lower ω_{TO2} - values of sample E due to lower In-concentration (lower compressive strain)

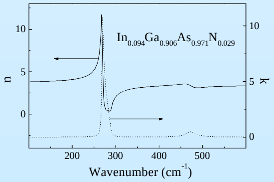
→ amplitude f of GaN-like resonance [$f = (\omega_{LO2} - \omega_{TO2})/\omega_{TO2}$] increases with y and with biaxial strain ε_{xx}, which is used to calculate N- and In-concentrations

Model Dielectric Function

F. Gervais and B. Piriou, J. Phys. C **7**, 2374 (1974).
D. W. Berreman and F. C. Unterwald, Phys. Rev. **174**, 791 (1968)

$$\varepsilon^L(\omega) = \varepsilon_\infty \prod_{i=1}^{2} \frac{\omega_{LOi}^2 - \omega^2 - i\,\omega\gamma_i}{\omega_{TOi}^2 - \omega^2 - i\,\omega\gamma_i}$$

Optical Constants



Determination of y_N and x_{In}

Starting Point

1.) FIR-Ellipsometry on GaAsN/GaAs and GaAsN/InAs/GaAs superlattices [J. Appl. Phys. **89**, 294 (2001)]:

→ amplitude f of the GaN-like phonon changes with y (N-concentration) and ε_{xx} (biaxial strain):

$$f = \alpha y + \beta\varepsilon_{xx} \text{ with } \alpha = 0.33,\ \beta = 0.51 \qquad (1)$$

→ Assumption: Validity of Eq. 1 for InGaAsN

2.) f-values resulting from FIR-Ellipsometrie on InGaAsN (this work)

3.) lattice misfit $(\Delta a/a)_\perp = (a_{InGaAsN}^\perp - a_{GaAs})/a_{GaAs}$ from HRXRD

Nitrogen-Concentrations

→ relation between $(\Delta a/a)_\perp$ and ε_{xx}:

$$e_{xx} \equiv \frac{a_{GaAs} - a_{InGaAsN}}{a_{InGaAsN}} = -\frac{a_{GaAs}}{a_{InGaAsN}} \cdot \frac{C_{11}}{C_{11} + 2C_{12}} \cdot \left(\frac{\Delta a}{a}\right)_\perp, \qquad (2)$$

with the elastical constants C_{11} and C_{12} (start values: GaAs → second iteration: linear interpolation between C_{1i} values of the binary end-compounds GaAs and β-GaN).

→ $y = (f - \beta\varepsilon_{xx})/\alpha$ with ε_{xx} from Eq. 2 (3)

Indium-Concentrations

→ Vegard's law für $a_{InGaAsN}$ following from Eq. 2:

$$a_{InGaAsN} = a_{GaAs}(1-x)(1-y) + a_{InAs}(1-y)x + a_{GaN}(1-x)y + a_{InN}xy \qquad (4)$$

→ N-concentration follows after rearrangement of Eq. 4 with respect to x

Comparison with Growth Properties and Band Gaps

Sample	A	B	C	D	E
$(\Delta a/a)_\perp$	$7.3\cdot10^{-3}$	$8.1\cdot10^{-3}$	$7.4\cdot10^{-3}$	$7.8\cdot10^{-3}$	$1.9\cdot10^{-3}$
x	0.09 (1)	0.11 (1)	0.11 (1)	0.12 (1)	0.09 (1)
y	0.013 (2)	0.019 (2)	0.022 (3)	0.024 (3)	0.029 (4)
x_g	0.105	0.105	0.105	0.091	0.077
y_g	0.96	0.907	0.936	0.936	0.96
T_G (°C)	600	560	560	560	560
E_g (eV)	1.180	1.146	1.103	1.094	1.003

1.) all samples: $x/x_g \approx 1$ but $y/y_g << 1$ (x_g, y_g: gas-phase concentrations)
→ cause: rel. high vapor pressure of nitrogen above InGaAsN surface

2.) samples B-E: calculated nitrogen concentrations increase with increasing gas-phase values, and correspondingly with decreasing band-gap energies

3.) sample A: lowest N-concentration (highest E_g) despite highest gas-phase value due to increased growth temperature

4.) sample E: strong increase of nitrogen-composition due to reduced In-concentration

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