Optical phonons in $Al_xIn_yGa_{1-x-y}N$ films

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Our message

➢ We present the first study of the phonon mode properties of hexagonal $Al_xIn_yGa_{1-x-y}N$ films employing infrared spectroscopic ellipsometry (IRSE).

➢ Wurtzite $Al_xIn_yGa_{1-x-y}N$ ($x \leq 0.40$, $y \leq 0.17$) as well as $Al_xGa_{1-x}N$ ($x \leq 0.21$) possess an intricate phonon mode behavior, which does not fit into the simple "one-mode" or "two-mode" behavior scheme:

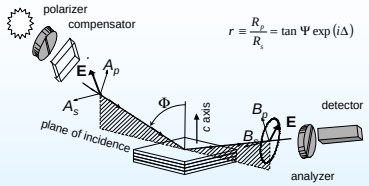
Two polar E_1 phonon branches are observed, which are GaN- and AlN-like, respectively. In between both E_1(TO) modes, a broad, possibly disorder-related phonon mode band occurs, which has already been predicted theoretically.

➢ The influence of strain and alloying on the GaN-like E_1(TO) mode frequency is differentiated.

➢ Incorporation of In reduces TO phonon mode broadening indicating improvement of the crystal quality.

➢ LO-phonon-plasmon coupling is observed for MOCVD grown $Al_xIn_{0.12}Ga_{0.88-x}N$ films. Assuming the effective electron mass, the free-electron concentrations in the films are estimated.

Infrared Ellipsometry



$$r \equiv \frac{R_p}{R_s} = \tan\Psi \exp(i\Delta)$$

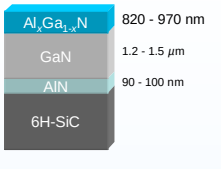
Model dielectric function for a-$Al_xGa_{1-x}N$ and a-$Al_xIn_yGa_{1-x-y}N$:

$$\varepsilon_j(w) = \varepsilon_{\infty,j} \prod_{i=1}^{l} \frac{w_{LO,ij}^2 - w^2 + i g_{LO,ij} w}{w_{TO,ij}^2 - w^2 + i g_{TO,ij} w} - \varepsilon_{\infty,j} \frac{w_{p,j}^2}{w(w + i g_{p,j})} \qquad j = \perp, \parallel$$

phonon contribution — free-carrier contribution "Drude term"

$\varepsilon_\perp(w)$: $l = 3$, one of which being non-polar ($w_{TO} = w_{LO}$)
$\varepsilon_\parallel(w)$: $l = 1$

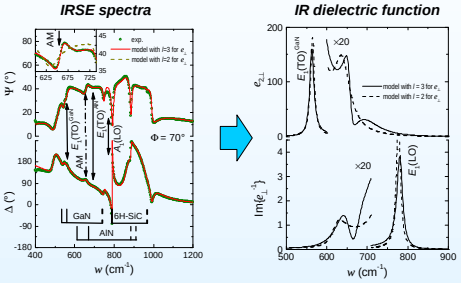
0.05 ≤ x ≤ 0.21, y = 0

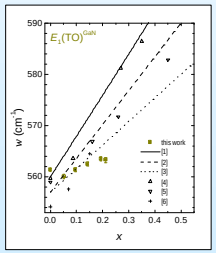


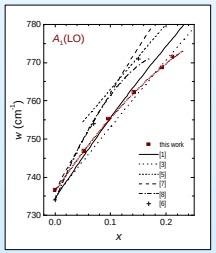
820 - 970 nm
1.2 - 1.5 μm
90 - 100 nm

• MOCVD
@ North Carolina State University
• strain gradients along growth direction

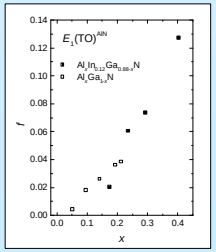
IRSE spectra — ***IR dielectric function***







Composition dependences of the GaN-like E_1(TO) phonon mode frequency (graph above), the A_1(LO) phonon mode frequency (graph diagonally above) and the polarity of the high-frequency E_1 phonon mode branch (right graph).



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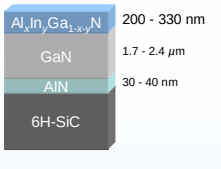
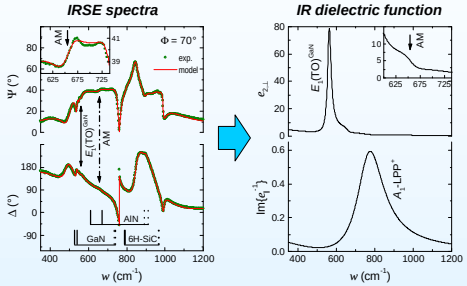
0 ≤ x ≤ 0.40, y » 0.12

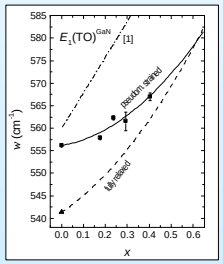


200 - 330 nm
1.7 - 2.4 μm
30 - 40 nm

• MOCVD
@ University of Stuttgart
• constant In flux and varied Al and Ga fluxes
• pseudomorphically strained quaternary layers
• T_{gr} = 770°C

IRSE spectra — ***IR dielectric function***





Left graph: Differentiation of the influence of strain and alloying on the GaN-like E_1(TO) phonon mode frequency:

$\Delta w(x,e_{xx}) = a_0 x + a_1 x(1-x) + b e_{xx}$

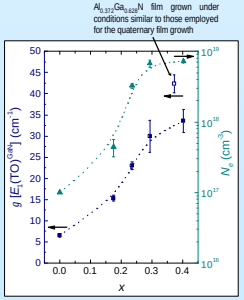
with the least-squares best-fit parameters a_0 = 80 (±25) cm^{-1}, a_1 = -48 (±19) cm^{-1}, and b = -1403 (± 150) cm^{-1}.

[1] Theoretical result for strain-free a-$Al_xGa_{1-x}N$; H. Grille *et al.*, Phys. Rev. B **61**, 6091 (2000)

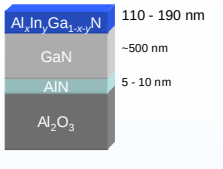
Right graph: Composition dependence of the GaN-like E_1(TO) phonon mode broadening g and the free-electron concentration N_e in the quaternary films.

The results obtained for g indicate an increase of compositional fluctuations and/or lattice imperfections with increasing x. However, the incorporation of In into ternary $Al_xGa_{1-x}N$ implies a significant improvement of the crystal quality.

The free-electron concentration increases by approximately two orders of magnitude from $N_e \leq 1\times10^{17}$ cm^{-3} for x = 0 to $N_e \sim 7\times10^{18}$ cm^{-3} for x = 0.40.



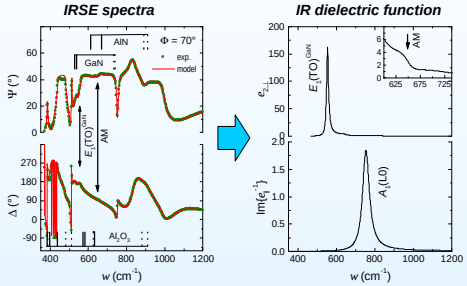
x = 0.12...0.17, 0.004 ≤ y ≤ 0.145

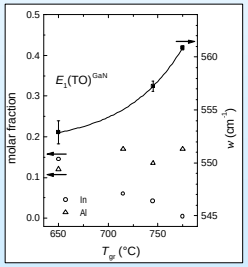


110 - 190 nm
~500 nm
5 - 10 nm

• plasma-induced MBE
@ WSI, TU Munich
• constant In, Al, and Ga fluxes
• different strain states
• T_{gr} = 650 ... 775°C

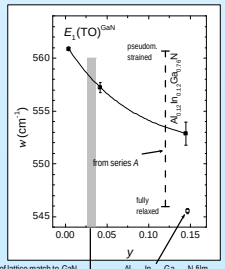
IRSE spectra — ***IR dielectric function***





GaN-like E_1(TO) phonon mode frequency vs. growth temperature (left graph) and In content (graph below), respectively.

Due to the comparatively high In vapor pressure, the incorporation of In into the quaternary films is diminished with increasing growth temperature. As expected from the behavior of the ternary compounds, the GaN-like E_1(TO) mode is subject to a significant red-shift with increasing y.



According to the results obtained from the $Al_xIn_{0.12}Ga_{0.88-x}N$ series, the E_1(TO) phonon mode frequency value measured for the $Al_{0.12}In_{0.145}Ga_{0.728}N$ epilayer indicates partially compressive in-plane film strain with e_{xx}=-4.9 (±0.8)×10^{-3}. This value agrees well with results from X-ray reciprocal space maps (e_{xx}=-6.6×10^{-3}).