Dielectric function and band structure of cubic InN

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InN represents the least studied compound among the group-III nitride semiconductors. Determination of the complex dielectric function (DF) $\bar{\varepsilon}(\hbar\omega)=\varepsilon_1(\hbar\omega)+i\cdot\varepsilon_2(\hbar\omega)$ of InN from the infrared into the VUV spectral region by spectroscopic ellipsometry (SE) has become one of the most powerful methods for studying absorption related properties of In-compounds both around the band gap as well as in the range of the high-energy critical points (CP) of the band structure [1-3]. By analysing the optical anisotropy around the absorption edge we demonstrated unambiguously that the band gap of hexagonal InN (h-InN) amounts to only 0.68 eV at room temperature which is much lower than the long-time accepted value of 1.9 eV. Theoretical calculations of the DF for the range of the high-energy CP's proved that the anisotropy and the peak positions can only be reproduced if electron-hole interaction is consequently taken into account [4].

So far, much less is known about the cubic counterpart (c-InN). Only recently, high quality epitaxial films became available. Preliminary ellipsometry studies [5] revealed an even lower band gap than for h-InN of about 0.61 eV, which is very close to the theoretical predictions of 0.59 eV [4] and 0.53 eV [6]. Here, we report for the first time on the experimental determination of the DF for c-InN up to photon energies of 10 eV and a comparison to numerical results.

The investigated films were grown by plasma-assisted molecular beam epitaxy. First, a 600 nm thick c-GaN buffer layer was deposited on (001)-oriented 3C-SiC substrate. Then, for the two studied samples c-InN films of 75 nm or 130 nm were grown. High-resolution X-ray diffraction proved the high crystalline quality of the films. Only 5% and 9% of hexagonal inclusions were found. Further growth details and results of the structural characterisation can be found elsewhere [5]. The ellipsometric data above 4 eV were recorded with BESSY ellipsometer attached to the 3m-NIM-monochromator, excellent matching to the data for the energy range from 0.74 up to 4.5 eV obtained with a commercial ellipsometer (J.A. Wollam Co., Inc.) was achieved.

Determination of a reliable DF becomes only possible if the peculiarities of the semiconductor surface are taken into account. In a first step, organic contaminations and the native oxide has to be removed. As demonstrated by our previous studies of h-InN it can be achieved by heating the sample to about 350 °C for 10 minutes. Figure 1(a) shows a comparison of the measured imaginary parts of pseudo-DF at room temperature for the as-grown sample (red) and cleaned film (blue). From the increase of the peak intensities after treatment a removal of about 3 nm oxide is estimated. It emphasises the necessity of surface preparation. In the second step, the post-annealing data are fitted by multi-layer approach which takes the surface roughness into account in order to get the DF of c-InN. The final results for the real and imaginary part of the DF are shown in Fig.1(b) by the red and blue lines, respectively. The spectral dependence up to 4 eV is very similar to the behaviour found for h-InN [1]. We find a sharp increase of ε_2 around the gap followed by a plateau. The shape of ε_2 above 4 eV is however markedly different to both, the ordinary and extraordinary DF of h-InN. A pronounced double-peak structure is found between 5 eV and 6.5 eV followed by a shoulder at 7 eV and a third peak at 9 eV.

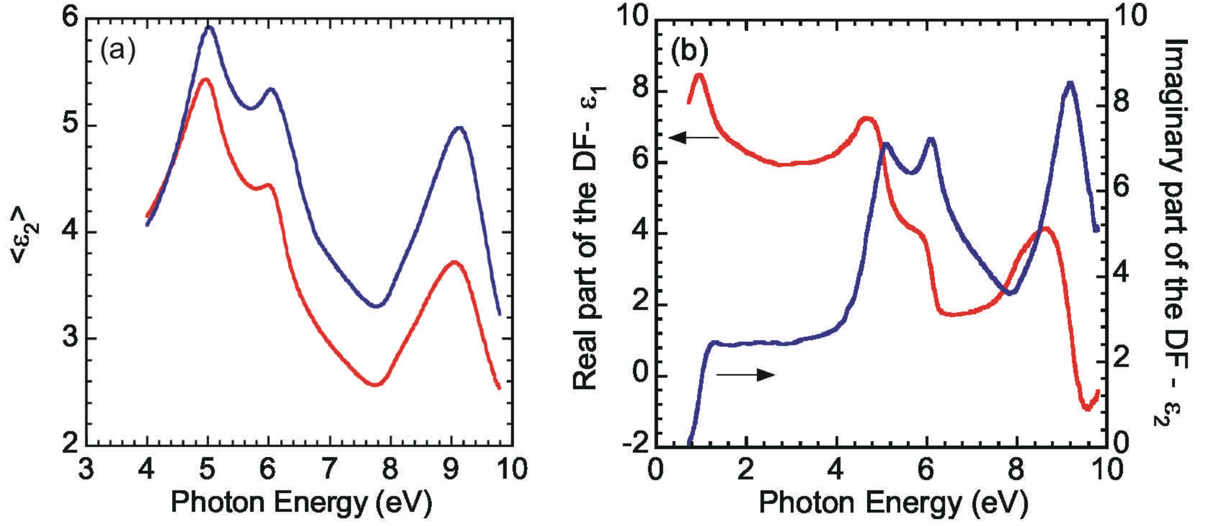

Fig. 1: (a) Comparison of the imaginary parts of the pseudo-DF prior (red) and after (blue) heating under UHV conditions. The data refer to room temperature. (b) Fitted real (red) imaginary part (blue)of the DF for cubic InN at room temperature.

Theoretical calculations of the optical properties of cubic InN starts from careful study of the quasi-particle band structure which has been described in more detail in Ref. 4. Taking the computed single-electron energies and wave functions only, one obtains an imaginary part of the DF as shown by the black line in Fig.2(a). Features attributed to transitions in the vicinity of the CP's are clearly. However, their spectral position deviates up to 800 meV from the experimental results (blue line). The discrepancy is lifted, if instead the electron-hole pair Hamiltonian is used for the DF calculation. Both, the peak positions and the peak intensity ratios, are in excellent agreement. As already observed for h-InN, only the magnitudes differ slightly to the experimental data. The current results for cubic InN are a further manifestation of our previous observation that the inclusion of excitonic effects over the whole spectral range is fundamental for the understanding of optical properties of nitride semiconductors.

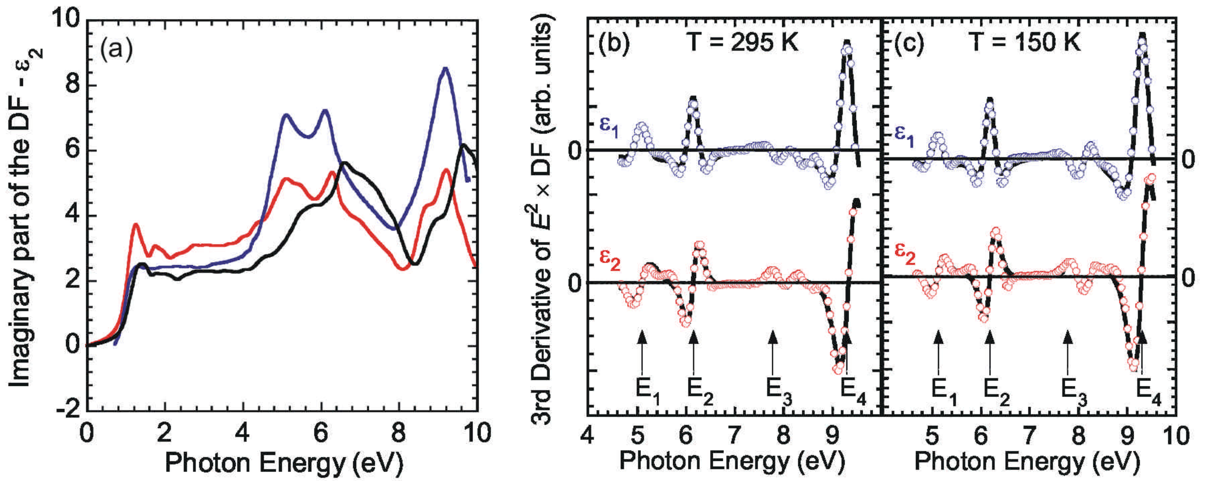

Fig. 2: (a) Imaginary part of the DF of cubic InN. Comparison of Coulomb-correlated electron-hole pairs (red) and independent quasiparticles spectra (black) to the experimental result (blue). Fit of the third derivative of the DF for room temperature (b) and T=150 K (c). The circles represent the experimental data, and solid lines are the best fit. The transition energies of the critical points are marked by arrows.

For a high-resolution determination of the transition energies of the critical points following accurate approach was used. One calculate the third derivative of the point-by-point obtained DF (after surface roughness correction) multiplied by the square of the photon energy *E*. The resulting spectra can be fitted [7] via

$$\frac{d^3}{d(\hbar\omega)^3}\left((\hbar\omega)^2\bar{\varepsilon}\right)=\sum_{j=1}^{4} e^{i\cdot\phi_j}\cdot\frac{C_j}{\left(\hbar\omega+i\cdot\Gamma_j-E_j\right)^{n/2}} \quad , \qquad (1)$$

where ϕ_j, C_j, Γ_j, and E_j denote the phase angle, the magnitude, the broadening energy, and the transition energy of the j-th CP, respectively. With n=6 corresponding to a two-dimensional critical point, we get the fit results for T=295 K and T=150 K as depicted in Fig. 2(b) and (c), respectively. At room temperature, the transition energies amount to 5.09 eV (E_1), 6.14 eV (E_2), 7.83 eV (E_3), and 9.28 eV (E_4). Especially the E_3 transition becomes much sharper in the numerical derivatives if the sample temperature is reduced to T=150 K. The low-temperature spectrum is characterized by Van Hove singularities at 5.12 eV (E_1), 6.18 eV (E_2), 7.85 eV (E_3), and 9.31 eV (E_4), i.e. the average shift between both temperatures amounts to about 30 meV. For comparison, similar studies of hexagonal InN yielded 40 meV.

In summary, we have determined the DF of cubic InN exhibiting a band gap of only 0.61 eV. Efficient surface cleaning was achieved by heating the samples up to 350 °C under UHV conditions. The DF shows in the high-energy part of spectrum contributions from at least four critical points of the band structure. Between 295 K and 150 K the transitions undergo a shift to higher energies by about 30 meV. It was demonstrated that the experimentally determined spectral dependence of the DF is well reproduced by theoretical calculation if electron-hole interaction is consequently taken into account.

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