Connecting ab inito surface structures to photoemission experiment by one-step model calculations: GaN(0001)

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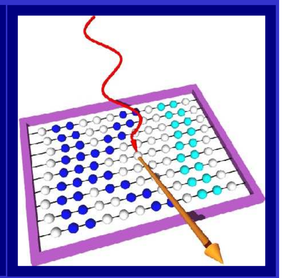


In 1997 Dhesi et al. [1] presented an extensive experimental examination of the electronic structure of the GaN(0001) surface by angle resolved photoelectron spectroscopy. The sample were grown using molecular-beam epitaxy obtaining a surface with a sharp 1x1 low-energy electron diffraction pattern (LEED). In the spectra a non dispersive feature was identified as emission from a surface state near the valence-band maximum.

Using scanning tunneling microscopy and total energy calculations within the local density functional theory, Smith et al. [2] predicted an Ga adlayer structure for the 1x1 surface. Using this model as a starting point, we calculated the photocurrent in normal and off-normal emission within the one-step model. By comparing our detailed interpretation with the experiment by Dhesi et al. we could connect the geometric and electronic structure to the measured photocurrent [3]. Especially, the surface state pointed out near the upper valence band edge could be identified in theory and experiment, which underlines the plausibility of the surface model by Smith et al.

Later, a second experimental examination by angular resolved photoemission by Chao et al. [4] yielded rather different spectra. The GaN thin films where grown by metal-organic chemical-vapor deposition, which also show a very sharp 1x1 LEED pattern. Two new surface bands were measured, one of them being highly dispersive throughout most of the valence band. Wang et al. [5] performed a systematic ab initio examination of the formation energy and band structure of several clean and Ga and N covered 1x1 GaN(0001) and GaN(000-1) surface configurations. Only the surface band structure of the clean nitrogen terminated GaN(000-1)-(1x1) is in favourable agreement with the measured data. But still theory and experiment show larger deviations, such as near the K point, where the measurements show a strong dispersing band which has no counterpart in theory.

Starting from the ab initio surface models by Wang et al. we calculate and analyse the photocurrent within the one-step-model.

First Results:

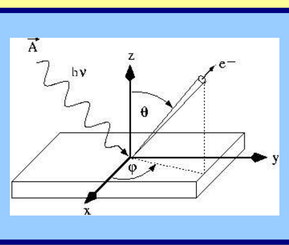
•One-step-model calculations based on the Ga adlyer model explain the experimental details for measurements taken from MBE samples by Dhesi et al.

•Characteristic emissions from the Ga adlayer are absent in the experiment from MOCVD samples by Chao et al.

•Despite the good agreement between the experimental band mapping and ab initio suface band structure for a pure nitrogen terminated surface, the photoemission theory expect strong emission from a nitrogen dangling bond state which are not visible in experiment

•For lower excitation energy a pure Ga-terminated surface could explain some of the experimental results, but for higher photon energies a strong direct transition in theory is missed in experiment.

ARUPS



One-Step-Model

•Photocurrent calculated from the "Golden Rule"

$$I^{fi}(\vec{A}, E_f) \sim \alpha \sum_{n}^{occ} \left| \langle \Psi_f(E_f + \hbar\omega) | \vec{A}_0 \cdot \vec{p} | \Psi_n(E) \rangle \right|^2 \delta(E - E_n)$$

Calculation of the initial states:

•Halfspace-Greensfunction

•Empirical Tight-binding method

•Basis: layer resolved bloch sum

•Transferintegrals: Extended Hückel Theory

Calulation of the final states:

•Time reversed LEED state

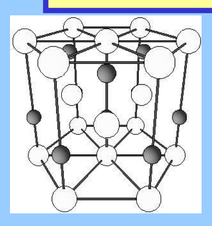
CKomplex band structure

•Pseudopotential

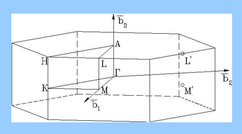
•Damping: optical potential

GaN Bulk properties

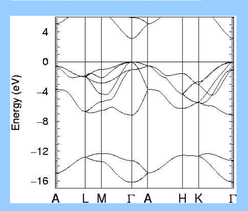
Geometry



Brillouinzone



Bandstructure



Complex final bands

References:

[1] S.S. Dhesi, C.B. Stagarescu, K.E. Smith, D. Doppalapudi, R. Singh and T.D. Moustakas; Phys. Rev. B 56, 10271 (1997).

[2] A.R. Smith, R.M. Feenstra, D.W. Greve, J. Neugebauer and J.E. Northrup; Phys. Rev Lett. 79, 3934 (1997).

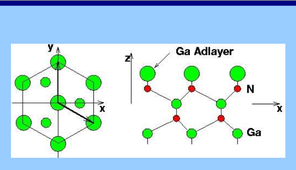
[3] T. Strasser, C. Solterbeck, F. Starrost, and W. Schattke; Phys. Rev. B 60, 11577 (1999).

[4] Y.-C. Chao, C.B. Stagarescu, J.E. Downes, P. Ryan, K.E. Smith, D. Hanser, M.D. Bremser, and R.F. Davis; Phys. Rev. B 59, R15586 (1999).

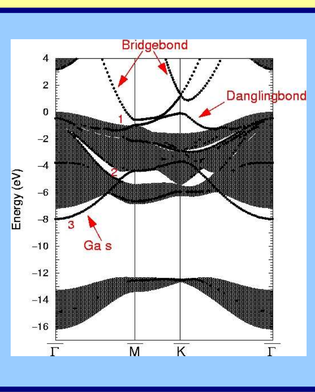
[5] Fu-He Wang, Peter Krueger, and Johannes Pollmann; Phys. Rev. B 64, 035305 (2001).

GaN(000$\bar{1}$)-(1x1)Ga

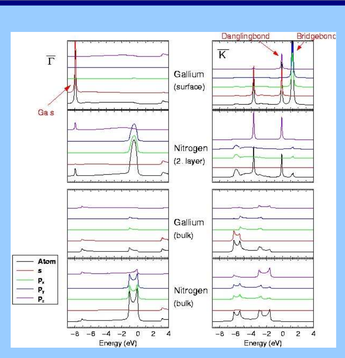
Geometry and Surface Brillouinzone



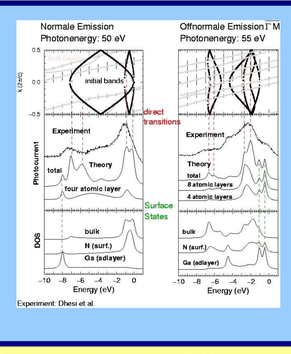
Surface Bandstructure



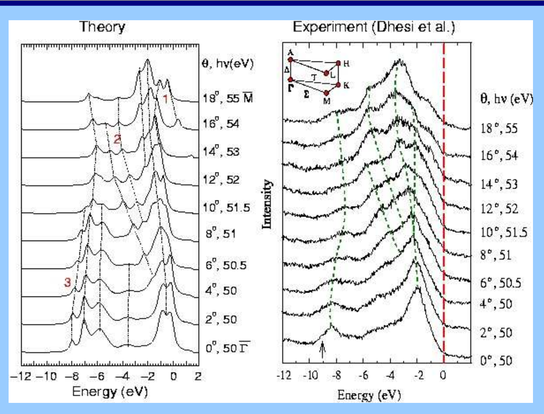
Density of States



Comparison of calculated photocurrents with experiments from MBE samples



Detailed analysis with DOS, layer resolved photocurrent and direct transitions

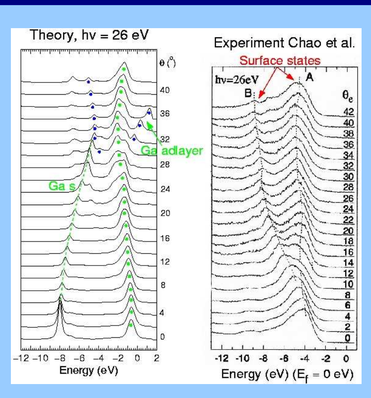


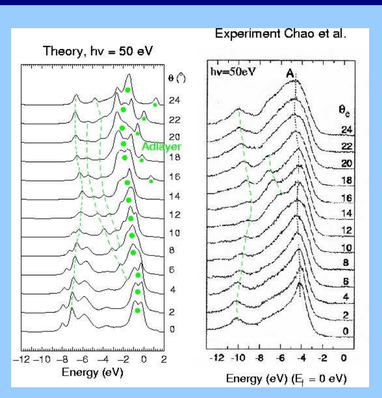
•Good agreement between theory and experiment for the Ga adlayer model and MBE samples

•The One-Step-Model explain all experimental structures by their microscopic details

•Photoemission calculations connect experiment to a surface structure and their electronic properties

Comparison of calculated photocurrents with experiments from MOCVD samples





For MOCVD samples:

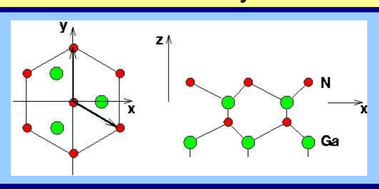
•Ga s emissions missed in experiment

•Surface state A corresponds to emission from the upper valence band edge

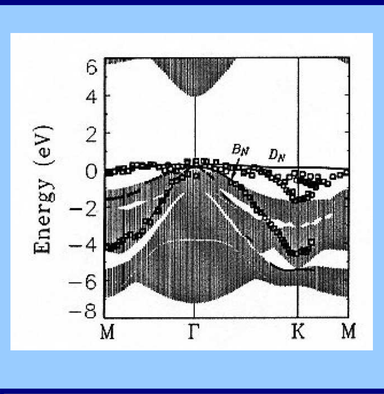
•Surface state B is only visible at high angles in theory

GaN(000$\bar{1}$)-(1x1) (N-terminated)

Geometry



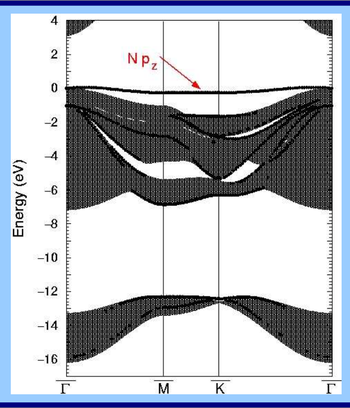
Theoretical band structure and experimental band mapping



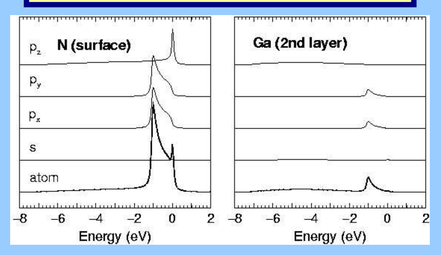
•The surface band structure by Wang et al. and the experimental band mapping by Chao et al. agrees well

•The weak dispersing surface state A in experiment is identified as an Nitrogen dangling bond state.

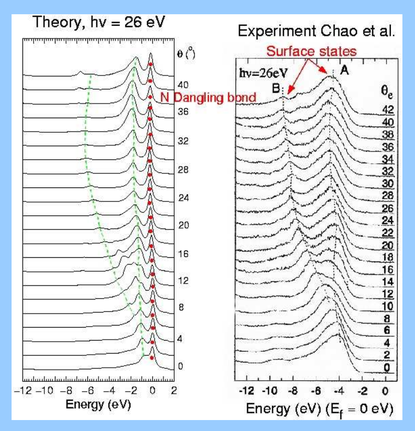
Surface Bandstructure

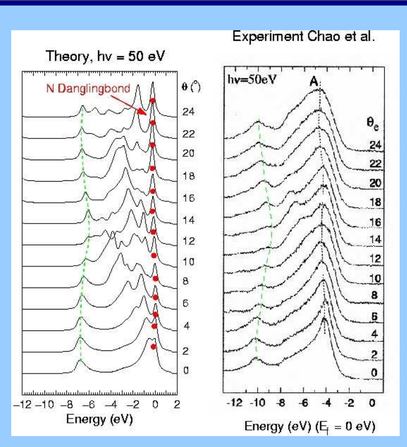


Density of States



Comparison of calculated photocurrents with experiments from MOCVD samples





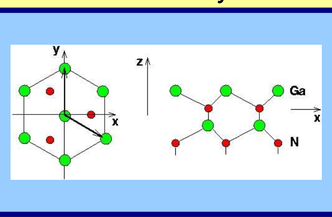
• Surface state B: weak resonance in theory

•Emission from the upper valence band edge correspond to the strong experimental emission A in experiment

•Strong emission from the N Dangling bond are missed in experiment or hidden in the broad structure A

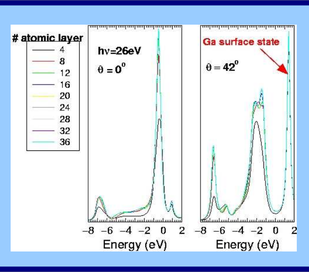
GaN(0001)-(1x1) (Ga-terminated)

Geometry

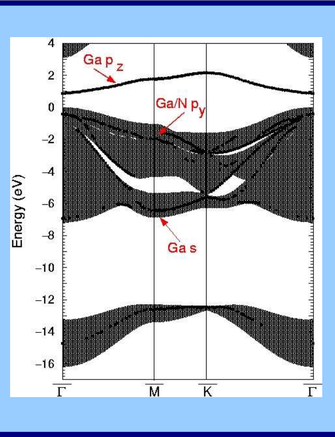


Layer resolved photocurrent

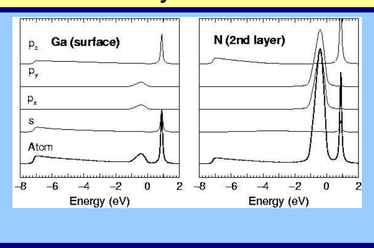
(without Fermi-function)



Surface Bandstructure



Density of States



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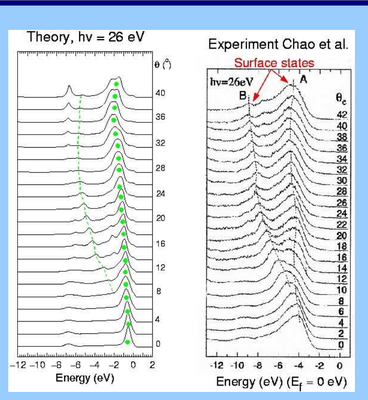

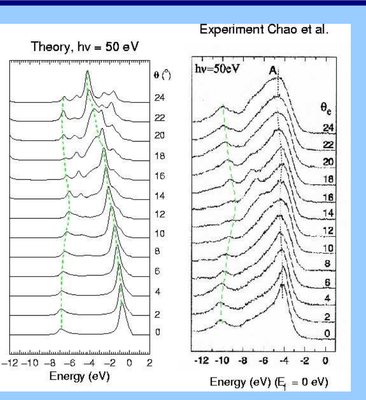

• Surface state B: weak resonance in theory

• Surface state A: emission from the upper valence band edge with strong contributions from the first atomic layers (26 eV photon energy)

• But: theoretical spectra for 50 eV excitation energy show a strong direct transition which could not be identified in experiment.