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P. Kessler · K. Lorenz · S. M. C. Miranda · J. G. Correia · K. Johnston · R. Vianden ·

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Abstract The role of indium in GaN and AlN films is investigated with the method of the perturbed angular correlation (PAC). Using the PAC probe ^{111}In in addition to indium on substitutional cation sites a large fraction of probes is found in a distinctly different microscopic environment which was attributed to the formation of an indium nitrogen-vacancy (V_N) complex. The influence of an electron capture induced *after effect* is ruled out by additional measurements with the PAC probes $^{111\text{m}}\text{Cd}$ and ^{117}Cd and using GaN with different dopants. It is shown that the V_N is not bound to substitutional Cd impurities suggesting that the In- V_N complex formation is a particularity of In in GaN and AlN. Finally, a preliminary model is presented to explain the temperature behavior of the electric field gradient, observed in the In- V_N complex measured with ^{111}In .

Keywords GaN · AlN · Ternary semiconductors · Perturbed angular correlation (PAC) · Defect complex · Point defects

This work reports on PAC measurements in InGaN and AlInN using Cd-111m and Cd-117 probes. The results show that a signal previously observed using In-111 arises from an In-nitrogen-vacancy complex and is not influenced by an electronic aftereffect. The background, experimental method, and results are all described well. In short, the work reported is of high quality, and I recommend the manuscript for publication without modification.

P. Kessler (✉) · R. Vianden
Helmholtz-Intstitut für Strahlen- und Kernphysik, Universität Bonn, 53115 Bonn, Germany
e-mail: kessler@hiskp.uni-bonn.de

K. Lorenz · S. M. C. Miranda · J. G. Correia
Instituto Tecnológico e Nuclear, 2686-953 Sacavém, Portugal

K. Johnston
Technische Physik, Universität des Saarlandes, 66041 Saarbrücken, Germany

the ISOLDE collaboration
ISOLDE CERN, Geneva, Switzerland

1 Introduction

InGa_N based LEDs show a high luminous efficacy despite a high defect concentration. These defects, mainly threading-dislocations that form during the growth of Ga_N films on lattice mismatched substrates would normally lead to non-radiative recombination of excitons. However, it seems that such non-radiative recombination is efficiently suppressed in indium containing alloys [1]. The role of indium in Ga_N and Al_N films is investigated with the method of the perturbed angular correlation (PAC) [2].

In previous measurements [3–5] it was shown, that in addition to indium on substitutional cation (Ga/Al) sites a configuration attributed to an indium nitrogen-vacancy (V_N) defect complex (In– V_N) is observed with the PAC probe ^{111}In . The electric field gradient (EFG) of the In– V_N is oriented parallel to the c-axis of the crystal. It decreases with temperature and finally, above a critical temperature (Al_N: $T_{\text{critical}} = 1,100\text{ K}$, Ga_N: $T_{\text{critical}} = 500\text{ K}$) it cannot be distinguished from the EFG of the wurtzite lattice (W–EFG) any more. This is accompanied by a decrease of the In– V_N quadrupole interaction frequency's (QIF) distribution from $\delta = 40\%$ to a constant value of $\delta = 10\%$. When measured at room temperature again, after the increase of the temperature up to 1,100 K, this process is fully reversible, indicating that the complex is stable up to this temperature.

Additionally, after a temperature treatment in vacuum, to diffuse out nitrogen and therefore produce V_N , an increase of the probes on In– V_N sites can be observed. Co-implantation of stable ^{115}In decreases this fraction. PAC measurements with ^{181}Hf show no defect induced EFG and therefore are a first hint that the complex is indium specific [4].

In– V_N was also observed in AlIn_N where the characteristic signal is even more pronounced than in GaIn_N [5]. In– V_N could be a competing exciton trap and may be involved in the processes leading to the high efficacy. Nevertheless some doubts remain about the influence of *after effects* on the PAC measurements using the ^{111}In probe and about the fact that the actual PAC measurement occurs at an excited level of the cadmium daughter nucleus.

To rule out the possible influence of an electron capture (EC) induced *after effect* and to confirm the assumption that the In– V_N is a particularity of indium and that V_N are not bound to other heavy substitutional impurities in particular cadmium, in this work additional measurements with $^{111\text{m}}\text{Cd}$ and ^{117}Cd were performed at the ISOLDE facility.

2 Experimental details

^{111}In decays with $T_{1/2} = 2.83\text{ d}$ via EC to an excited state of ^{111}Cd with $T_{1/2} = 120\text{ ps}$ which decays via a γ – γ cascade with an intermediate excited state with $T_{1/2} = 85\text{ ns}$ to the ground state of ^{111}Cd . After the EC process a so called *after effect* can influence the measurement, which will be explained later in detail.

$^{111\text{m}}\text{Cd}$, decays with a half life of 49 min via the same intermediate state as the cascade of ^{111}In . So this probe is perfectly suited to evaluate, if the complex is indium specific and to check for a possible EC *after effect*. ^{117}Cd decays with $T_{1/2} = 2.49\text{ h}$ via beta decay to an excited state of ^{117}In with $T_{1/2} = 4.37\text{ ns}$. The following cascade's

Table 1 Properties of the samples

	GaN			AlN
Dopant	Mg (p-type)	Zn (high-res.)	Si (n-type)	None
Thickness [μm]	2–5	2–5	2.5–3.5 (a) 3–7 (b)	7
Carrier-concentration	$(0.5\text{--}5) \times 10^{18} \text{ cm}^{-3}$	Compensated	$(4\text{--}6) \times 10^{17} \text{ cm}^{-3}$ (a) $(1\text{--}9) \times 10^{18} \text{ cm}^{-3}$ (b)	

Two different Si doped GaN samples were used for each cadmium probe. (^{111}mCd : (a), ^{117}Cd : (b)). All samples were grown on sapphire substrate

intermediate state has a half life of $T_{1/2} = 53.6$ ns. This represents the opposite case to the indium to cadmium decay of ^{111}In [6].

The cadmium probes were produced at the ISOLDE facility at CERN. Commercially available¹ AlN samples and GaN samples grown by hydride vapor phase epitaxy and doped with Mg, Si, and Zn respectively were implanted with an energy of 30 keV and fluences between 10^{11} and 10^{12} ions/cm². The samples' properties are summarized in Table 1. All samples were annealed at 1,223 K under N₂-flow for 10–20 min. The PAC measurements with the probe ^{117}Cd were performed at a 6-detector BaF₂ [7] setup, the ^{111}mCd at a 4-detector BaF₂ setup, both at the ISOLDE facility.

The $e^- \gamma$ correlation measurements were carried out in a set-up described in [8]. The probe ^{111}mCd was chosen, because it “simulates” an EC *after effect*. When a conversion electron is emitted instead of a γ as start signal for the measurement, it leaves a hole in the shell of the atom. This hole is filled up successively with nearby electrons from the lattice. The mean half life of these holes depends on the concentration of electrons in the conduction band and their mobility. This process is similar to the *after effect* of the EC, where a hole in the inner shell, that is formed during the EC process, is filled up by electrons from the outer shells.

3 Results

The results of the $\gamma\text{--}\gamma$ PAC measurements at room temperature (RT) and at 473 K for ^{111}mCd and ^{117}Cd are shown in Fig. 1 for GaN doped with Mg (p-type), Zn (compensated semi-insulating) and Si (n-type). For the probe ^{111}mCd , no defect complex influence can be seen either in p-type, high resistive and n-type GaN. This is also the case for n-type samples (Si) measured with ^{117}Cd . The same behavior can be observed with undoped AlN samples. Small differences between the spectra are attributed to different strain states of the as-grown material and slightly varying implantation and annealing conditions.

This and the previous results mentioned in the introduction lead to the conclusion that the In–V_N indeed is a purely indium related defect complex, that is not formed

¹Technologies and Devices International Inc., 12214 Plum Orchard Drive, Silver Spring, MD 20904, USA.

Fig. 1 PAC-ratio function of differently doped GaN samples measured at room temperature and 473 K with the probes $^{111}\text{m}\text{Cd}$ and ^{117}Cd (last column). Only the W-EFG interaction frequency can be seen

Fig. 2 **a** $e^- - \gamma$ -ratio function of GaN and AlN measured at 10 K and 473 K respectively. No influence of a “simulated” *after effect* can be seen. **b** Proposed model to explain the temperature dependence of $\text{In}-\text{V}_\text{N}$. If V_N is annihilated quickly enough, the PAC ratio function shows only the interaction frequency of the lattice EFG

at cadmium. The QIF of the measurement is given in the figure. Noticeable is the increase of the QIF of the Mg doped sample both at RT and 473 K compared to undoped GaN.

The $e^- - \gamma$ measurements for GaN and AlN at 10 K and 473 K (Fig. 2a) support the assumption that the observed QIF is related to a defect complex and not due to an EC *after effect*. Since no influence of the measurement temperature other than the strength of the W-EFG either in the AlN or p-type GaN sample can be observed.

To explain the observed behavior of the probe ^{111}In in GaN/ AlN we suggest the following preliminary model shown in Fig. 2b. Since the actual measurement of the EFG is at the intermediate ^{111}Cd state and we showed above that the complex is not formed with cadmium there are two possible situations depending on the critical temperature T_critical . After the decay of ^{111}In to ^{111}Cd , the V_N is annihilated after a certain amount of time. If this is happening during the first 120 ps, the half life of the

first excited state of ^{111}Cd , only the lattice EFG is measured. If the temperature is too low so that the V_N is not filled up quickly enough, its influence is measured since the complex still exists.

4 Summary

PAC-measurements with different cadmium probes show that a defect complex seen in PAC measurements with ^{111}In is indium specific and does not form at the cadmium-site. No *after effect* can be observed with $^{111\text{m}}\text{Cd}$ e^- - γ measurements which leads to the possible explanation that the V_N is annihilated after the decay of ^{111}In to ^{111}Cd . Below a critical temperature the V_N remains bound to the probe atom during the measurement, since the V_N has not enough energy to leave the cadmium within the life time of the initial excited state of ^{111}Cd .

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