EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Addendum to the ISOLDE and Neutron Time-of-Flight Committee

The role of In in III-nitride ternary semiconductors

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Abstract

The present addendum to project IS481 will allow accomplishing the work of the running project which was not possible due to technical problems. Furthermore, the scope of the project will be extended from binary GaN and AlN to ternary AlGaN semiconductors. $\beta-\gamma$ Perturbed Angular Correlation (PAC) measurements using the ^{115}Cd(^{115}In) probe in GaN will allow the determination of the sign of the quadrupole interaction, an important parameter when comparing experimental results with density functional calculations of the electric field gradient. $\gamma-\gamma$ PAC using the probes ^{111m}Cd(^{111}Cd) and ^{117}Cd(^{117}In) will be used to investigate AlGaN ternaries. Special focus will lie on the investigation of implantation damage and alloy disorder. The presence and characteristics of In-V_N complexes in these alloys will be studied and compared to our previous results in binary nitrides.

Requested shifts: 12 shifts, (split into two runs over one year)

1. Introduction

Due to their wide and direct band gaps, group-III nitrides are promising semiconductor materials for a wide range of technological applications in optoelectronics, as well as high power- high frequency- and high temperature electronic devices[i,ii]. These devices usually consist of heterostructures composed of binary (AlN, GaN, InN), ternary (InGaN, AlGaN, AlInN) or even quaternary layers. Despite the successful commercialization in particular of InGaN based photonic devices many basic questions on the mechanisms of light emission remain unanswered. A subject of intensive scientific discussion is the fact that the intense luminescence from InGaN based LEDs and laser diodes is relatively insensitive to the large density of defects, mainly threading dislocations[iii]. A widely accepted explanation for this behaviour is the localization of excitons at regions with minima in the potential caused for example by compositional fluctuations. This effect seems to be especially effective in In-containing III-nitride alloys, however, the nature of the exciton trap is still unknown[iv].

2. Project results

The present project, running since 2009, aims to understand the role of In in III-nitride alloys by investigating its immediate lattice surroundings in different III-nitride hosts. The Perturbed Angular Correlation (PAC) technique provides a powerful tool to examine the lattice environment of a substitutional In probe on a microscopic scale. Previous studies with the PAC-probe ^{111}In(^{111}Cd) in GaN and AlN revealed an unexpected, reversible behaviour of its local structure with temperature[v,vi,vii]. After implantation and annealing GaN and AlN at 1000ºC only a fraction of the In probes was found in undisturbed substitutional cation sites while a large fraction is subjected to a strong electric field gradient typical for a point defect trapped at the probe atom. However, when the measurements were performed at higher temperature the undisturbed substitutional fraction increased until all probes are found in regular sites. Surprisingly, this behaviour is fully reversible when the temperature is decreased again.

These results suggested that a large fraction of In impurities in GaN and AlN form a stable complex with a nearest neighbour nitrogen vacancy in the <0001> position. However, at the start of the present project it was not clear if the fact that the actual PAC measurement is performed at the Cd daughter nucleus and if after effects[*] influence the PAC measurements using the ^{111}In(^{111}Cd) probe. Therefore, PAC measurements using the ^{111m}Cd(^{111}Cd) and ^{117}Cd(^{117}In) probes were performed in GaN and AlN. It was shown that the defect complex is absent for the ^{117}Cd(^{117}In) and ^{111m}Cd(^{111}Cd) probes. The defect complex is therefore a peculiarity of In in III-nitride alloys. Furthermore, by performing e^--γ PAC measurements using the ^{111m}Cd probe it was shown that after effects are not likely to influence the measurements at the analysed temperature down to 15 K[viii].

A third objective of the project could not be accomplished due to technical difficulties. The aim was to determine the *sign* of the quadrupole interaction by means of a $\beta-\gamma$ measurement using the ^{115}Cd or the ^{111}Ag probes. In a first try, the sample implanted at ISOLDE with ^{115}Cd could not be shipped to the University of Bonn in due time to perform the measurements. Therefore, in a second tentative the longer lived ^{111}Ag isotope was implanted. This isotope, however, was found to diffuse out of the sample during the necessary post-implant annealing treatment.

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[*] Disturbances of the probe's electronic shell are caused by the decay of ^{111}In to ^{111}Cd via electron capture. The hole in the electron shell will be filled by x-ray and Auger processes leaving further holes and excited states that cause fluctuating field gradients. In some insulators, where the recombination processes are slow due to the low electron mobility, this was seen to cause the loss of angular correlation[*], however, in semiconductors usually the holes are filled in a short time interval within the time resolution of the spectrometer and therefore do not interfere with the measurement.

In this addendum to the present project we ask for beam time to repeat the ^{115}Cd implantation for $\beta-\gamma$ measurement. Furthermore we would like to extend the project from binary alloys to ternary AlGaN alloys. Preliminary measurements of this material showed that PAC is a useful technique to study the effect of implantation on crystal quality and to investigate alloy disorder[ix].

3. Proposed work

3.1 $\beta-\gamma$ measurements using ^{115}Cd(^{115}In) in GaN

The use of the probe ^{115}Cd(^{115}In) will allow the determination of the sign of the quadrupole interaction by means of a $\beta-\gamma$ measurement. Although the half life of the intermediate state of this nucleus is much shorter than that of the comparable states in ^{111}Cd and ^{117}In, with the information gained by the previous studies, it should be possible to extract the necessary information. This would be very important since, at present, density functional calculations of the electric field gradient (EFG) in the nitride semiconductors are hampered by the fact that the value of the u-parameter (u=b/c where b is the bond-length in the c-direction and c the lattice parameter) is not known with sufficient precision neither in GaN nor in AlN. Variations of the u-parameter within the experimental errors cause even the sign of the numerical EFG result to change, rendering any calculations uncertain at present.

A mobile beta detector has been set-up in Bonn for these measurements which can be moved to ISOLDE to perform the ^{115}Cd measurements in order to avoid the shipping procedure.

3.2 $\gamma-\gamma$ measurements using ^{111m}Cd(^{111}Cd) and ^{117}Cd(^{117}In) in AlGaN alloys

PAC measurements with the probes ^{111m}Cd(^{111}Cd) and ^{117}Cd(^{117}In) will be performed in $Al_xGa_{1-x}N$ alloys within the entire compositional range ($0\leq x\leq 1$). Residual implantation damage and alloy disorder will cause a damping of the observed PAC frequency. Furthermore, complementary measurements with the ^{111}In probe will be performed to check if the typical In-defect complex seen in AlN and GaN is also present in the ternary and to study how the corresponding PAC frequency and the fraction of probes trapping a defect change with composition.

Summary of requested shifts:

We estimate the total amount of ISOLDE beam time needed to accomplish the above-described tasks to be 12 shifts, distributed according to table I:

Table I: Beam time request

<table>
<tr><th>Required isotope</th><th>Implanted beam</th><th>PAC experiment</th><th>Intensity [at/μC]</th><th>Target / Ion source</th><th>Comments</th><th>nº of shifts</th></tr>
<tr><td>^{111m}Cd</td><td>^{111m}Cd</td><td>$\gamma-\gamma$, $e^--\gamma$</td><td>10^8</td><td>molten Sn, plasma</td><td></td><td>6</td></tr>
<tr><td>^{117}Cd</td><td>^{117}Ag</td><td>$\gamma-\gamma$</td><td>10^8</td><td rowspan="2">UC_2, RILIS (Ag)</td><td rowspan="2">Nb or Ta ion source cavity to decrease In surface ionization contamination</td><td>3</td></tr>
<tr><td>^{115}Cd</td><td>^{115}Ag</td><td>$\beta-\gamma$</td><td>10^8</td><td>3</td></tr>
</table>

All of our beam times consist of collections to be measured off-line and can in this way be easily shared with other users. We stress the particular case of the ^{111m}Cd beam time, where collections should run day and night with a period of about 4-5 hours between collections that usually last for 15-30 min. There are actually four PAC setups co-shared during beam times and the samples can be implanted on the same collective sample holder used with other users which are also doing PAC experiments.

For these PAC experiments, the number of implanted atoms per sample range from $5 \cdot 10^8$ up to 10^{11}, depending on half-lives, coincidence efficiency and on the fluence limit for proper recovery of the implantation damage. All isotopes will be collected in the general-purpose implantation chambers at GLM and/or High Voltage Platform at the ISOLDE hall, building 170. All γ–γ PAC measurements will be performed off-line, outside the ISOLDE hall, in the new Solid State Laboratory in building 115.

Several furnace systems exist already at ISOLDE for annealing treatments under vacuum or gas flow at atmospheric pressure at the new SSP lab.

References:

[i] B. Gil, ed., in Group III nitride semiconductor compounds, physics and applications; Series on Semiconductor Science and Technology 6 (Oxford Science Publications, Oxford , 1998).

[ii] S. Nakamura, G. Fasol, The blue laser diode – GaN based light emitters and lasers, Springer Verlag (1997).

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[iv] S. F. Chichibu et al., nature materials 5, 810 (2006).

[v] K. Lorenz, F. Ruske, and R. Vianden, Appl. Phys. Lett. 80, 4531 (2002).

[vi] K. Lorenz and R. Vianden, Hyp. Int. 158, 273 (2004).

[vii] J. Schmitz, J. Penner, K. Lorenz, E. Alves, and R. Vianden, physica status solidi (a) 205, 93 (2008).

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises: *(name the fixed-ISOLDE installations, as well as flexible elements of the experiment)*

Part of the Choose an item.	Availability	Design and manufacturing
SSP-GLM chamber	☒ Existing	☒ To be used without any modification
Existing equipment on the solid state labs in building 115 - 6 detector PAC standard setups - annealing furnaces - glove boxes	☒ Existing	☒ To be used without any modification ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT

(if using fixed installation) Hazards named in the document relevant for the fixed SSP-GLM chamber and building 115 installations.

Additional hazards:

Hazards	*SSP-GLM*	*Building 115*	*[Part 3 of the experiment/equipment]*
Thermodynamic and fluidic			
Pressure	[pressure][Bar], [volume][l]		
Vacuum	10-6 mbar at SSP chamber 10 during collections		
Temperature	295 K, room temperature collections		
Heat transfer	-		
Thermal properties of materials	-		
Cryogenic fluid		Liquid nitrogen, 1 Bar, few litres used during the PAC measurements on appropriate dewar	
Electrical and electromagnetic			
Electricity	[voltage] [V], [current][A]		
Static electricity			
Magnetic field	[magnetic field] [T]		
Batteries	☐		
Capacitors	☐		
Ionizing radiation			
Target material	[material]		
Beam particle type (e, p, ions, etc)			

Beam intensity			
Beam energy			
Cooling liquids	[liquid]		
Gases	[gas]		
Calibration sources:	☐		
• Open source	☒ Produced at ISOLDE 111mCd (48m) 117Cd(2.5h) 115Cd(53.5h)	Sources to be measured at 115	
• Sealed source	☒	22Na sources provided by RP services at CERN, used at 115	
• Isotope	111mCd (48m) 117Cd(2.5h) 115Cd(53.5h)		
• Activity	111mCd (48m) < 3 e 7 Bq 117Cd(2.5h) < 8 e 6 Bq 115Cd(53.5h) < 4 e 5 Bq		
Use of activated material:	none		
• Description	☐		
• Dose rate on contact and in 10 cm distance	[dose][mSV]		
• Isotope			
• Activity			
Non-ionizing radiation			
Laser	none		
UV light	none		
Microwaves (300MHz-30 GHz)	none		
Radiofrequency (1-300MHz)	none		
Chemical			
Toxic	[chemical agent], [quantity]		
Harmful		Acetone (ICSC: 0087), ethanol (ICSC: 0044) and methanol (ICSC: 0057). Less than few centilitres per chemical, used on cleaning samples on ventilated fume hood on building 115. The respective ICSC forms have been printed and will be handled during preparation and experiments.	
CMR (carcinogens, mutagens and substances toxic to reproduction)	[chemical agent], [quantity]		
Corrosive	[chemical agent], [quantity]		
Irritant	[chemical agent], [quantity]		
Flammable	[chemical agent], [quantity]		
Oxidizing	[chemical agent], [quantity]		
Explosiveness	[chemical agent], [quantity]		
Asphyxiant	[chemical agent], [quantity]		
Dangerous for the environment	[chemical agent], [quantity]		
Mechanical			
Physical impact or mechanical energy (moving parts)	[none]		
Mechanical properties	[none]		

(Sharp, rough, slippery)			
Vibration	[none]		
Vehicles and Means of Transport	[none]		
Noise			
Frequency	[frequency],[Hz] Ambient noise at the ISOLDE Hall, building 170		
Intensity	Ambient noise at the ISOLDE Hall, building 170		
Physical			
Confined spaces	[none]		
High workplaces	[none]		
Access to high workplaces	[none]		
Obstructions in passageways	[none]		
Manual handling	All samples and sample holders are manually handled either by long tweezers to insert and extract the sample holder into and out of the SSP implantation chamber at GLM, or when manipulating the samples and sample holders inside glove boxes or fume houses on building 115 r-007	All samples and sample holders are manually handled either by long tweezers to insert and extract the sample holder into and out of the SSP implantation chamber at GLM, or when manipulating the samples and sample holders inside glove boxes or fume houses on building 115 r-007	
Poor ergonomics	[none]		

0.1 Hazard identification

3.2 Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above): *(make a rough estimate of the total power consumption of the additional equipment used in the experiment)*

There is no additional equipment with relevant power consumption on these small-scale experiments.