Lattice Location of Implanted ^{147}Nd and ^{147*}Pm in GaN using Emission Channeling

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The lattice location of ^{147}Nd and ^{147*}Pm in thin-film, single-crystalline hexagonal GaN was studied by means of the emission channeling technique. The angular emission yields of β^- particles and conversion electrons emitted by the radioactive isotopes ^{147}Nd and ^{147*}Pm were measured using a position-sensitive detector following 60 keV room temperature implantation at a dose of 1×10^{13} cm^{-2} and annealing at 900°C. The emission patterns around the [0001], [$\underline{1}$102], [$\underline{1}$101], and [$\underline{2}$113] crystal axes give direct evidence that the majority (≈70%) of Nd and Pm atoms occupy substitutional Ga sites.

Related to the fact that temperature-stable and narrow-line width electroluminescence can be generated from atomic transitions of rare earth (RE) atoms, there are perspectives to realize optoelectronic devices operating in the visible or infrared region based on RE doped GaN [1,2]. RE dopants that have been reported in that respect are Er, Tm, Pr, Eu, Dy, Sm, Ho and Nd. Since the local crystal environment strongly influences the intensity and splitting of optical transitions, a prerequisite for optical activity is to incorporate the dopant in optically active lattice sites. In GaN, these are generally considered to be the substitutional Ga sites. Consequently, knowledge of the lattice position of the rare earth atoms is helpful for a detailed understanding of their luminescence properties.

Neodymium (Nd), which is widely applied in commercial Nd^{3+} YAG lasers operating at a wavelength of 1064 nm, has so far not been extensively studied as an optical dopant in semiconductors. At present the only investigation on Nd doped GaN is a photoluminescence (PL) and photoluminescence excitation (PLE) study performed by KIM *et al.* [3]. While their results showed the existence of several distinct Nd defect complexes of trigonal symmetry, the PL and PLE data did not provide any direct indication of the actual lattice sites occupied by Nd.

In this article we present a lattice location study of ^{147}Nd and its daughter nucleus ^{147*}Pm in GaN using the emission channeling technique [4,5]. This experimental method makes use of the fact that charged particles emitted by radioactive isotopes in a single crystal or epitaxial thin film experience channeling or blocking effects along crystallographic axes and

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planes. The resulting anistropic emission patterns depend characteristically on the occupied lattice sites of the probe atoms.

The sample was a 1-2 μm thin film of single-crystalline, hexagonal GaN epitaxially grown on a sapphire substrate by metalorganic chemical vapor deposition (MOCVD). Implantation of 60 keV ^{147}Nd ions was performed at the ISOLDE facility at CERN at an angle of 7° from the surface normal up to a dose of 1×10^{13} cm^{-2} into a 1 mm diameter beam spot. The radioactive isotope ^{147}Nd ($t_{1/2}$ = 10.98 d) decays via emission of β^- particles (endpoint energy 804.9 keV) to the excited state ^{147*}Pm, which decays with a half-life of 2.5 ns to the ground state of ^{147}Pm. This transition is accompanied by the emission of 45.9 keV conversion electrons (49%) or a γ-ray (51%). Using a position-sensitive detection system, we measured the angular distributions of β^- particles and conversion electrons (CE) around 4 crystal axes both in the as-implanted state as after vacuum annealing at 900°C. While the β^- patterns give direct information on the lattice location of ^{147}Nd prior to the β^- decay, the CE patterns allow lattice site determination of ^{147*}Pm right after the radioactive decay. Note that during the β^- decay of ^{147}Nd the ^{147*}Pm atom receives a nuclear recoil around 2.4 eV only, so it is very likely that ^{147*}Pm inherits the lattice location of ^{147}Nd.

One of the known problems in electron emission channeling experiments is the backscattering of electrons from the sample and other components of the setup into the detector, resulting in an isotropic background. Corrections related to this backscattering were quantified by means of Monte Carlo simulations using the GEANT3 code [6], taking into account the elemental composition and geometrical features of the GaN film, the Al_2O_3 substrate, the Mo sample holder, and the stainless steel vacuum chamber.

In order to identify the lattice sites of ^{147}Nd and ^{147*}Pm, the experimental channeling yields were compared to simulated patterns for the emitting atoms on several possible lattice sites: substitutional Ga (S_{Ga}) and substitutional N (S_N) sites with various root mean square (rms) displacements, bond-centered sites within and off the c-axis (abbreviated as BC-*c* and BC-*o*), antibonding sites AG-*c*, AN-*c*, AG-*o*, and AN-*o*, the "hexagonal" sites HG and HN, the so-called T and O sites [7], as well as various displacements between these sites. More information on the simulations and on the lattice sites can be found elsewhere [8].

Figure 1(a) shows the β^- [0001] emission channeling pattern. The channeling effect along the c-axis and major crystal planes intersecting the c-axis is evidence of ^{147}Nd occupying sites located within the atomic rows along this direction. The best two-fraction fit of a theoretical pattern to the experimental yield is shown in Figure 1(e). This corresponds to a fraction of 83% of Nd atoms along the c-axis rows with an rms displacement of u_1 = 0.20 Å perpendicular to these rows. Note that this does not yet prove that the Nd atoms are on substitutional Ga sites. To discriminate between the different interstitial and substitutional sites located along the c-axis, the following axes where also measured: [$\underline{1}$102], [$\underline{1}$101], and [$\underline{2}$113] [Figs. 1(b)-1(d)]. The fit of theoretical patterns [Figs. 1(f)-1(h)] to the experimental data provided direct evidence that the majority of Nd atoms occupy substitutional Ga sites. The conversion electron emission channeling patterns, which give information on the lattice location of ^{147*}Pm just after the β^- decay, were measured around the same 4 axes [Figs. 2(a)-2(d)], with very similar results. Including additional lattice sites in the fits did not significantly improve the quality of the fits, although small fractions (around 5%) of Nd and Pm atoms on S_N, O, HG, AG-*c* or AG-*o* sites would still be in accordance with the experimental data.

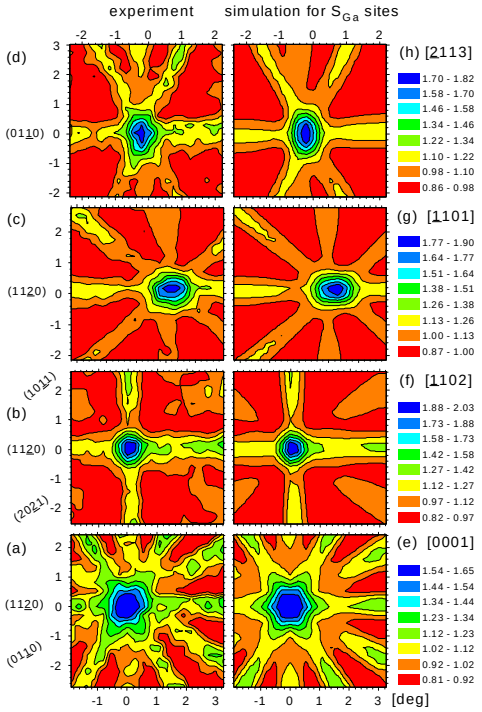

Fig. 1: Angular-dependent β^- emission yields from ^{147}Nd in GaN around the crystalline axes [0001] (a), [1102] (b), [1101] (c) and [2113] (d) following 10 min vacuum annealing at 900°C. Channeling patterns were extracted from the β^- energy window above 55 keV up to the endpoint energy. Panels (e)-(h) represent the best two-fraction fit of theoretical patterns to the experimental data, corresponding to 83%, 69%, 56%, and 62% of Nd atoms on substitutional Ga sites S_{Ga}, with rms displacements perpendicular to the corresponding axes of $u_1 = 0.20$ Å, 0.14 Å, 0.10 Å, and 0.16 Å, respectively. Note that the remaining fractions of ^{147}Nd atoms are considered to occupy random (R) sites, giving an isotropic contribution to the emission yield in each pattern.

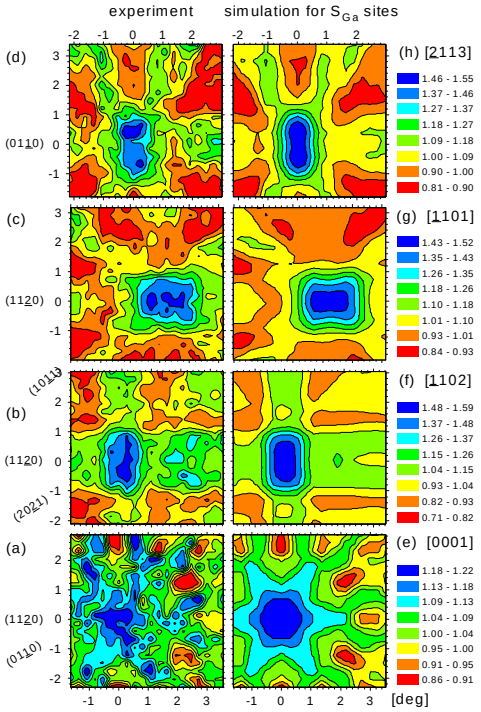

Fig. 2: Angular-dependent emission yields of conversion electrons from ^{147*}Pm in GaN around the crystalline axes [0001] (a), [1102] (b), [1101] (c) and [2113] (d) following 10 min vacuum annealing at 900°C. Channeling patterns were extracted from the CE energy window from 15 keV to 55 keV. Panels (e)-(h) represent the best two-fraction fit of theoretical patterns to the experimental data, corresponding to 67%, 65%, 53%, and 80% Pm atoms on substitutional Ga sites S_{Ga} with rms displacements perpendicular to the corresponding axes of $u_1 = 0.19$ Å, 0.22 Å, 0.20 Å, and 0.26 Å, respectively. The remaining fractions of ^{147*}Pm atoms are considered to occupy random (R) sites. Note that due to the 49% branching ratio for the conversion electron emission the CE patterns show worse statistics than those of Fig. 1.

Figure 3 contains the summary of results for the ^{147}Nd and ^{147*}Pm measurements in the as-implanted state and after 900°C annealing. As can be seen from Figs. 3(a) and 3(b), annealing the sample did not substantially change the substitutional fraction of Nd and Pm atoms. Another observation is that roughly the same substitutional fractions are found for both atoms. This proves that the recoil energy of 2.4 eV imported to the daughter during the β^- decay did not result in the occupation of fundamentally different lattice sites. The rms dis-

placements of Nd atoms from the perfect substitutional Ga sites in the as-implanted state and after 900°C annealing are shown in Fig. 3(c). These displacements are for all axes clearly larger than the thermal vibration amplitude of Ga, which is 0.074 Å. Also a small decrease is visible after annealing, which is possibly due to the removal of crystal defects close to the rare earth atoms, resulting in a more perfect incorporation into substitutional lattice sites. The rms displacements of the Pm atoms [Figure 3(d)] are somewhat larger than those of Nd, which could be caused by the recoil energy which ^{147*}Pm receives during the β^- decay.

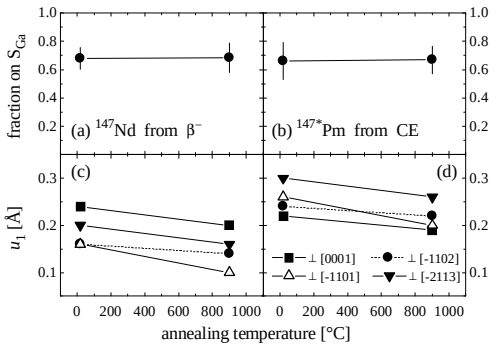

Fig. 3: Fractions of ^{147}Nd (a) and ^{147*}Pm (b) atoms on substitutional Ga sites and their 1-dimensionally projected root mean square displacements u_1 perpendicular to the indicated crystal directions for the as-implanted state and after annealing at 900°C. Note that in all cases the measurements were done at room temperature. The room temperature rms displacements of Ga and N atoms are u_1(Ga) = 0.074 Å and u_1(N) = 0.081 Å, respectively.

Summarizing, we have given direct evidence that the majority of Nd and Pm atoms occupy substitutional Ga sites already following implantation at room temperature. Annealing the implanted sample at 900°C did not significantly affect the substitutional fraction, although the rms displacements of the Nd and Pm atoms decreased slightly. The substitutional fractions for Nd and Pm and their rms displacements are very similar to those previously reported by us for implanted ^{143}Pr [8].

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