NON-STATIONARY PHOTOCONDUCTIVITY OF GaN NANOCOMPOSITES IN ARTIFICIAL OPAL MATRIX

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Abstract

It was recently proposed to use synthetic opals as a host matrix for obtaining 3D arrays of electronic nanodevices [1]. In the present work the opal matrices were infiltrated with GaN. We study electronic properties of opal-GaN, by means of transient photoconductivity (TPC) measurements using 5 ns laser pulses at wavelengths above (266 nm) and below (532 nm) the GaN bandgap (3.4 eV). A broad plateau is observed in the photocurrent decay covering several orders of magnitude. We compare the results with measurements in conventional GaN.

Introduction

Artificial opal was recently proposed as matrix for obtaining 3D arrays of electronic nanodevices, which allow to produce a six orders of magnitude higher density of active elements when compared to planar technology [1]. A successful use would enable to reach a working area of the junctions per unit volume in group-III-nitride-based light emitting diodes (LEDs) as high as 10 m^2/cm^3 and to reduce the current density by 3-4 orders of magnitude as compared with conventional planar systems.
A recent work demonstrated the ability to infiltrate GaN into the opal void sublattice, and the structural properties of the material was characterised by X-ray diffraction, Raman spectroscopy and atomic force microscopy [2].
Here, we study for the first time electronic properties of opal-GaN, by means of transient photoconductivity (TPC) measurements using laser pulses of 5 ns duration from a Nd:YAG system at wavelengths above (266 nm) and below (532 nm) the GaN bandgap of 3.4 eV. The time domain covered extends over 10 orders of magnitude. We compare the results with the power law decay observed in conventional GaN.

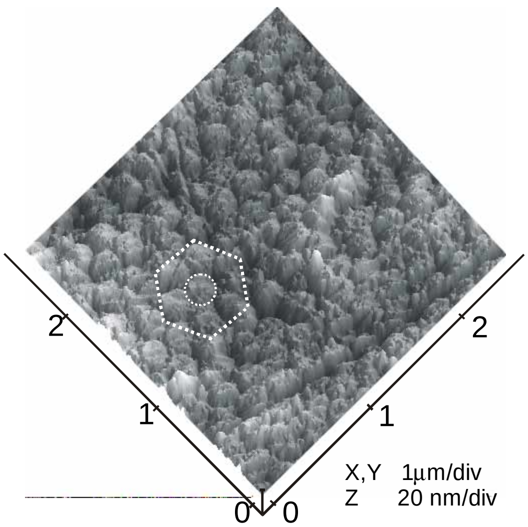

Figure 1: AFM image of opal infiltrated with GaN, fill factor 60 %; the dashed line indicates the hexagonal structure.

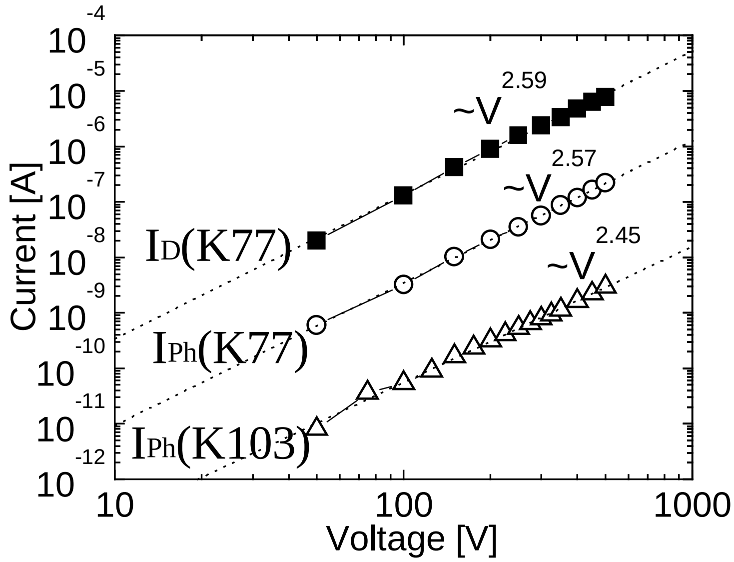

Figure 2: I-V curves for K77 under dark (squares) and photo (circles) conditions, and for K103 (triangles) under illumination. All curves follow a power law with index about 2.5, a signature of space-charge limited currents: $I \propto U^2$ (Mott-Guerney equation [3]).

Sample preparation

The host material is a single crystal of synthetic opal, consisting of close-packed amorphous silica (SiO_2) spheres with diameters of approximately 220 nm. The voids between the spheres have diameters around 45-90 nm, and up to 26 % is accessible by guest materials. Macroscopically, the 1 mm thick samples are porous and fragile.

To introduce GaN into the opal matrix we used heterogeneous chemical reactions of NH_3 with solid precursors containing Ga. In a first step, an oxide of Ga was deposited onto the internal void interfaces by thermal decomposition of Ga salts from a water/ethanol solution (chemical bath deposition technique). The second step, the nitration, consisted of annealing the sample in ammonia atmosphere, for around 30h at temperatures around 500-1000 °C. TEM, XRD, AFM, and Raman measurements were carried out to determine the structural properties of opal-GaN composites [2]. Hexagonal wurtzite structure with good coverage of the opal surfaces was confirmed by AFM (Figure 1). Optical reflection measurements confirm the redshift of the photonic pseudogap with increasing filling factor [2].
In this work, we study three samples:

- K77 is filled with GaN, has a fill factor of 60 % and is nanocrystalline.
- K103 is filled with GaN with 5 at.% of Mg, and has a fill factor of 30 %.
- K105 is filled with GaN and Pt, has fill factors of 30 % and 10 % for GaN and Pt, respectively, and the phases are separated.

The addition of magnesium and platinum in K103 and K105, respectively, had the intention to achieve p-type doping in the first sample and internal Schottky barriers in the second one.

Dark- and steady-state photocurrent – voltage characteristics

For the electrical characterisation, we used coplanar aluminium contacts, at a distance of 0.5 mm. The resistance for low voltages was some 100 kΩ for K77, and around 10 GΩ for K103 and K105. So, the samples show highly insulating properties. Efficient transport paths of strongly n-type doped GaN are not present, as could have been expected from planar GaN technology. However, the filling factor shows some influence: for K77, the filling factor is twice the value of K103 and K105, and the resistivity is orders of magnitude lower.

Figure 2 shows in a double logarithmic representation the dark- and steady-state photocurrent-voltage characteristics, and the respective linear fits. Only the less resistive K77 permitted to measure the dark current-voltage characteristics. The linear fit results in a power law index around 2.5. The steady-state photocurrent – voltage characteristics of K77 and K103 are shown in the same figure. The excitation source was a cw 633nm He-Ne line chopped at 30 Hz and read out with lock-in technique. The lines are again linear fits to the data in the double logarithmic representation.

Though there are orders of magnitude differences in the magnitude of the currents, the three curves show approximately the same analytic relation between the current and the voltage. For different samples, in dark and under illumination, we find the same power law index of 2.5.

For K105 it was not possible to obtain I-V transfer curves using this technique, since the signal-to-noise ratio was too small.

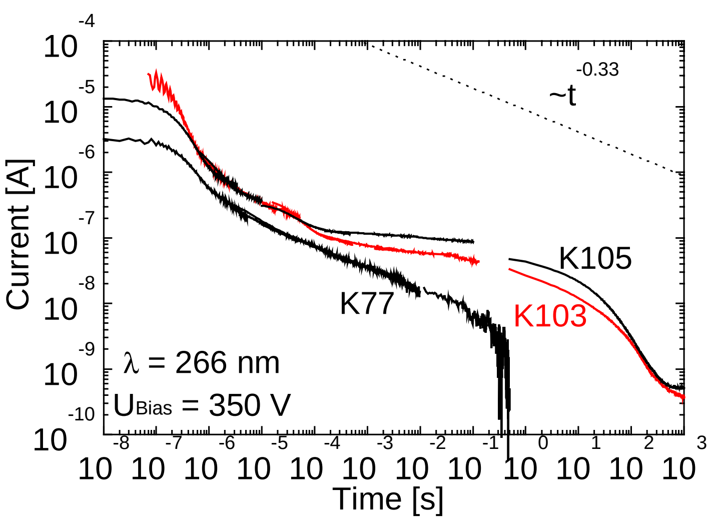

***Figure 3**: Photocurrent transients after UV pulse. Planar GaN behavior is indicated by the dashed line.*

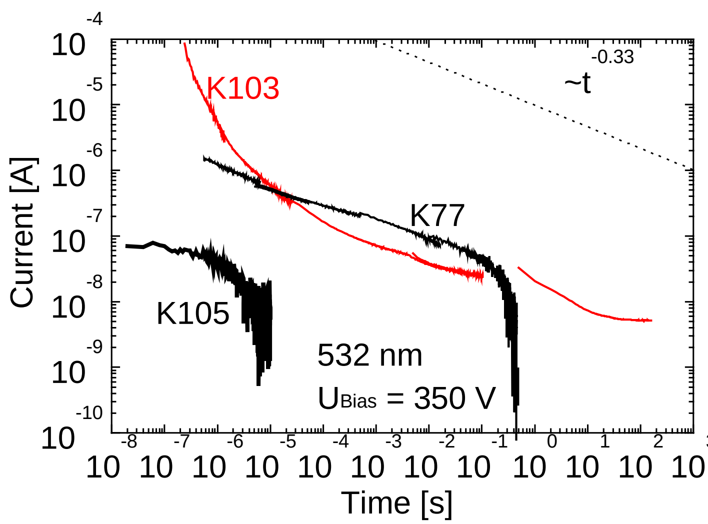

***Figure 4:** Photocurrent transients after VIS pulse. Planar GaN behavior is indicated by the dashed line.*

Photocurrent Transients

Non-stationary photoconductivity (PC) was studied with Q-switched Nd:YAG laser radiation. Strong 5 ns pulses of 532 nm and 266 nm laser lines yielded measurable transient photocurrents between milli- and nanoamperes at an applied bias voltage of 350V. Transients faster than tenths of milliseconds were measured with a Tektronix 200 MHz oscilloscope; transients slower than seconds with a Keithley amperemeter. The signal-to-noise ratio depended sensitively on the sample and the time regime. However, generally we could measure down to some nanoamperes with the Tektronix. The resulting composed current decay is shown in figures 3 and 4 for UV and visible radiation, respectively.

Figure 3 shows that for UV pulses, K77, K103 and K105 behave similarly up to about 100 ns. At this point, K103 and K105 reach a long plateau, maintaining the photocurrent level over orders of magnitudes in time. Just at about 1s a fast decay sets in, and the photocurrents fall rapidly below the detection limit.
In contrast to this, K77, decays obeying a power law with index of about -0.33, which is a typical feature of GaN [4]. The sudden decrease at about 0.3 s could be due to an onset of recombination.

Figure 4 shows the results for illumination with green light pulses. K77 behaves similarly to the UV case, with a power law decay up to about 0.3 s. This sample definitely shows the TPC characteristics of GaN [4], with its characteristic power law index of –0.33.
K103 and K105, however, behave in a very different way, compared with UV illumination. K103 shows a very high photoconductivity at 100 ns, decays “convexly”, reaching finally, at about some ms, a power law regime with index of –0.3, as GaN. This may be a sign that in this time regime (>0.1 ms) transport in the GaN gets dominant.
In contrast to this, the K105 sample shows only a very small initial signal, decays rapidly and shows even a slight quenching effect at longer times (not shown here).

Discussion

The complex opal structure infiltrated with GaN makes a quantitative photocurrent analysis difficult. Instead of one or more planar interfaces, one deals with inumerous curved internal interfaces. For optical measurements, three materials have to be taken into account: SiO_2, air and GaN (eventually additional materials). The proportion between air and GaN is determined by the macroscopic fill factor. In contrast to planar technology, for example, UV radiation could penetrate quite deeply into the sample by travelling inside the SiO_2, with multiple reflections at the internal interfaces. Additionally, the geometric structure of the opal combined with the filling factor leads to new effects as, for example, the photonic pseudogap and the filling induced redshift [2].

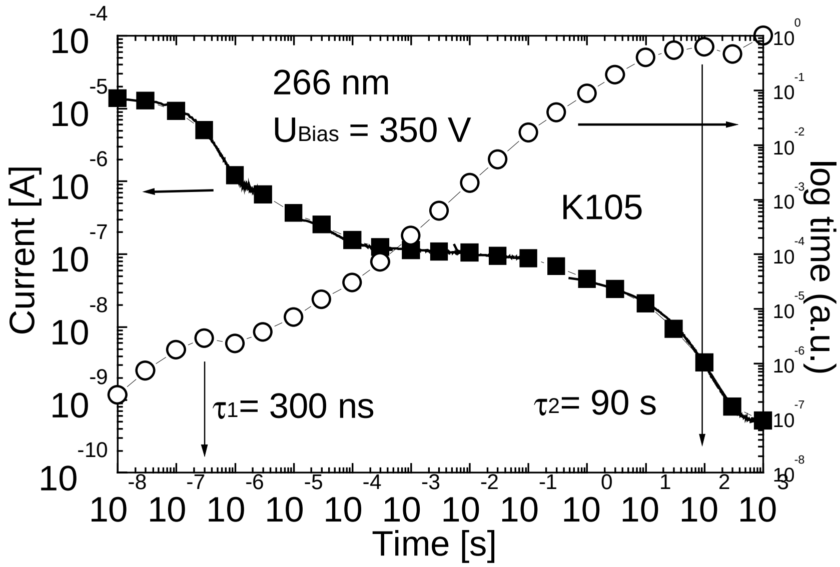

***Figure 5**: The TPC decay (solid squares) can be interpreted in terms of an apparent distribution g(τ) of carrier lifetimes (open circles) [5]:*

$$g(\tau) \propto I(t) \times t$$

Two broad peaks with mean values τ_i are obtained.

With well connected GaN covered interfaces, one should expect a reasonable conductivity: the GaN should be strongly n-type doped as known from planar technology. But the large resistivities observed show that there are no efficient transport paths present.
The finding of the same power law relation between current and voltage, for different samples and for dark- and photocurrent, can be explained by means of the existence of space-charge limited currents. For this case, theory predicts a parabolic relation between current and voltage $I \propto U^2$ (Mott-Guerney equation [3]). However, trapping and detrapping may vary the theoretical index of 2 over a significant range. This signature of trapping and detrapping may be related to the existence of numerous structural defects at the internal interfaces.

K77 shows definitively GaN typical photocurrent decay characteristics: for UV as well as for visible illumination, the decay obeys dominantly the power law with index –0.3. The relatively high filling factor of 60% possibly makes the difference to the other samples, where no good connection between the GaN covered interfaces exists.
The photocurrent decay of K103 and K105 after UV pulses is quite astonishing: it extends over 10 orders of magnitude in time, and shows a long unusual plateau. The TPC decay can be interpreted in terms of an apparent distribution g(τ) of carrier lifetimes [5]:

$$g(\tau) \propto I(t) \times t$$

Figure 5 shows the calculated apparent distribution of lifetimes for K105 after UV illumination. We find two lifetime maxima: τ_1=300 ns and τ_2=90 s, each being the mean value of a broad distribution. Assuming multiple trapping, the photocurrent can be deconvoluted into an effective density of defect states in the bandgap. According to

$$E = kT \ln(\nu t)$$

(where E is the energy of the defect level, k the Boltzmann constant, T the absolute temperature, and ν the escape frequency, $\nu=10^{12}$ Hz), the two mean lifetimes indicate defect levels at E_1=0.32 eV and E_2= 0.80 eV, respectively.

Conclusion

GaN can be infiltrated into the void sublattice of an opal host. The I-V curves are characterised by a power law with index 2.5, indicating space charge limited currents. Pure GaN (K77) as guest material leads to a power law decay with index –0.3 in the TPC data for UV and for VIS illumination, as is known from GaN. The TPC of K103 and K105 leads to a broad distribution of carrier lifetimes in the respective materials.

Acknowledgements

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