Realization of a High-Performance GaN UV Detector by Nanoplasmonic Enhancement

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As a result of the huge progress made on GaN-based light-emitting diodes (LEDs) and laser diodes,[1,2] another kind of GaN-based optoelectronic devices, GaN-based UV detectors, has attracted increasing interest. This is because of their potential application in flame monitoring and detection, biomedicine, environmental monitoring and UV astronomy, etc.[3] In addition, GaN-based UV detectors have the advantage of being solid-state and of small size, with good chemical and thermal stability, thereby saving energy and having a long lifetime. A lot of encouraging advances on GaN-based detectors have been reported in terms of their low dark current, high responsivity and high sensitivity in a variety of detector structures such as Schottky type,[4] metal-semiconductor-metal (MSM) type[5] and p–i–n type detectors.[6] In spite of these advantages, the performance of GaN-based UV detectors is still lower than expected and need much lower dark current and much higher responsivity in order to replace widely-used photomultiplier tubes to detect very weak UV signals. Currently, there are two main approaches to improve responsivity, namely photoconductive gain and avalanche multiplication. However, photodetectors with photoconductive gain usually have slow and noisy photoresponses[6] and avalanche-type GaN-based detectors can obtain high responsivity but at the expense of increased noise. Moreover, the avalanche-type detector has much more stringent requirements on the crystalline quality of GaN, the device structure and processing techniques. Recently, nanoplasmonic (NP) enhancement by nanoparticles has offered new opportunities to engineer and improve the performance of optoelectronic devices. In particular, NP enhancement has demonstrated significant potential for improving the performance of GaN-based LEDs and solar cells.[7–9] For example, Ag nanoparticles deposited on top of GaN/InGaN solar cells have been exploited to achieve greater optical absorption in the photoactive layer via nanoparticles scattering[8] and similar behavior has been obtained in Si and GaAs solar cells.[10–12] Through excitation of the surface plasmon, the incident light resonantly interacts with nanoparticles, greatly enhancing the scattering cross section.[13] This phenomenon has been exploited in many near-field NP-applications, such as nanoresolution optical imaging,[14] surface enhanced Raman scattering[15] and NP interference lithography.[16] In the application of NP enhancement, Au nanoparticles are also frequently used.[17–19] However, because of parasitic absorption, i.e. unwanted losses arising from the resonant coupling of the incident sunlight with the nanoparticles,[20,21] Ag nanoparticles are considered to be better than other metallic materials, exhibiting lower parasitic absorption and higher scattering. It has been reported that this resonant scattering effect can reach as high as 80% for Ag for wavelengths, λ > 400 nm.[22] Nanoparticles’ plasmonic scattering efficiency is attributed to surface charges and is determined by the geometry of the plasmonic structure.[13] It follows that by changing shapes of the metallic nanoparticles, the distribution of the surface charges can be modified. This allows one to tune the surface plasmon resonance frequency,[23–25] and manage scattering from the nanoparticles over a broad wavelength range.[11,26] However, up to now, research on the semiconductor optoelectronic devices that have been enhanced by metallic nanoparticles mainly focus on LEDs and solar cells working at visible frequencies and direct evidence of NP enhancement in UV detectors is still lacking.

In this paper, our purpose is to explore a new approach, based on nanoplasmonic enhancement, to improve UV detectors and specifically to study the effect of nanoplasmonics on the performance of GaN-based UV detectors. By fabricating Ag nanoparticles on a GaN surface, high-performance GaN UV detectors have been realized. The mechanism of Ag nanoparticle enhancement on the performance of GaN UV detectors is discussed in detail. The results reported here demonstrate that nanoplasmonic enhancement could be applied to the UV region, which will support the improvement and practical application of UV optoelectronic devices.

Figure 1a and b show typical tapping-mode atomic force (AFM) topographies of the GaN epilayer (without Ag, Sample A), the GaN with deposited Ag before annealing (i.e., as-deposited Ag, Sample B) and then after annealing (Annealed Ag, Sample C). The corresponding cross-sectional profiles of the three samples are also shown (Figure 1c). The GaN epilayer has a smooth surface with atomic steps that can be seen, and the RMS roughness of the GaN epilayer over a 2 μm × 2 μm area is 0.138 nm. The image of the as-deposited Ag on the GaN surface, using electron beam evaporation, shows nano-islands, where there is a high density Ag nanostructures. Following thermal annealing, the hemiellipsoid-shaped Ag nanoparticles are formed as a result of surface tension and recrystallization.
being about 16 times compared with the detector having no metallic nanoparticles.

Three possible reasons are proposed as the cause the enhanced responsivities of the detectors with Ag nanoparticles: i) the depletion width at the metal-semiconductor interface was widened by the higher Schottky barrier height (SBH) arising from the Ag nanoparticle between the GaN epilayer and Ni/Au Schottky metal contacts; ii) photoconduction gain was generated due to trapped holes caused by the surface state at the interface associated with Ag nanoparticles; iii) the number of photogenerated electron-hole pairs increased because of increased light absorption in the GaN epilayer resulting from the nanoplasmonic scattering effect from the Ag nanoparticles coupling with the incident light. To investigate which one of these is the most significant in improving the responsivity, a series of experiments were carried out.

We measured the reverse-bias leakages of the three samples and calculated their SBH using a thermionic emission model (Figure 1b). The density of the annealed Ag nanoparticles is about $1.8 \times 10^9$ cm$^{-3}$ and the half lengths of the main axes of the ellipsoid a, b, and c are about 70 nm, 78 nm, and 42 nm, respectively.

Figure 2 illustrates the room-temperature spectral response of three GaN UV MSM-type detectors without Ag, with as-deposited Ag and with annealed Ag on the GaN surfaces; all of which were measured at 5 V bias. The spectra show the peak responsivities around the band gap edge of GaN (365 nm), but the responsivity of the three detectors exhibited dramatic differences. Their responsivities are approximately 4000 mA W$^{-1}$, 2458 mA W$^{-1}$ and 148 mA W$^{-1}$ for the respective cases of annealed Ag nanoparticles, as-deposited Ag and without Ag. This indicates that the responsivity of the detector with annealed Ag nanoparticles was enhanced nearly 30 times over that without Ag nanoparticles. This enhancement phenomenon also occurs for the detector with as-deposited Ag, but the enhancement factor was lower than that with annealed Ag nanoparticles, being about 16 times compared with the detector having no metallic nanoparticles.

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Figure 3. The calculated SBHs for the three samples using the thermionic emission model, Ni/Au as the Schottky contact and In as the Ohmic contact for measurement. The horizontal line is simply to illustrate the relationship.

Figure 3, since the SBHs of the detectors with Ag are a little bit lower than that without Ag, the existence of Ag nanoparticles narrowed the depletion width of the Schottky contact instead of widening the width. Therefore, a widening of the depletion layer as a result of the presence of the Ag nanoparticles causing the enhancement of the responsivity could be neglected. Since the GaN MSM detectors consist of two Schottky barriers connected back to back on a coplanar surface, the depletion width could also be deduced qualitatively from the dark current of the three detectors. The larger the dark current is, the narrower the depletion width is.

Figure 4 displays the dark currents of the three detectors. This shows that the dark current of detectors with Ag nanoparticles has increased or no change compared to the one without Ag nanoparticles, further indicating that the enhanced responsivity of the GaN UV detector with Ag nanoparticles did not arise from the widened depletion width.

Moreover, as shown in Figure 4, the dark currents of the three detectors are symmetrical. Under a certain applied voltage, if the holes were trapped by the surface state at the interface, this would mean that the $p$ value would increase. From Equation 1, an additional electric field would be created due to this additional positive charge, and thus the SBH would be reduced,

$$\Delta V = \frac{q p d}{\varepsilon_0 \varepsilon_s}$$

where $q$ is the elementary charge, $p$ is the trapped sheet hole concentration, $d$ is the depletion width at the metal-semiconductor interface, $\varepsilon_0$ is the permittivity of free space and $\varepsilon_s$ is the relative dielectric permittivity. If the SBH is to be reduced by the trapped holes, the dark current curve would become asymmetric in the positive voltage range, which is not consistent with the measured dark current of the GaN UV detectors with Ag nanoparticles as illustrated in Figure 4. Therefore, the greatly enhanced responsivity of the detectors by Ag nanoparticles excludes this trapped holes effect.

To confirm this hypothesis, the temporal responses of the three detectors were measured by shutting on/off the light at the wavelength of 360 nm under a 5 V applied bias (Figure 5).
The three detectors exhibited nearly equally fast responses. It is well known that photogenerated electrons will circulate through the external circuit many times before recombining with holes if those photogenerated holes were trapped by the surface state at the interface. Consequently, the temporal photocurrent would not drop sharply when the UV light was turned off leading to a lower temporal response being observed. Thus, we can conclude that the enhancement of the responsivity in the presence of Ag nanoparticles is not a result of holes being trapped at the interface.

According to the above analysis, the only appropriate explanation for the enhancement of the responsivity of the detectors with Ag nanoparticles is a nanoplasmonic enhancement mechanism. The mechanism for the plasmonic scattering effect has been described in the literature. Figure 6 shows a schematic analysis of the Ag nanoparticles' scattering effect on the GaN surface. The surface plasmon peak of Ag has been proved to be around 360 nm and greatly depend on particle size and environment. It is therefore reasonable to deduce that plasmonic scattering by Ag nanostructures is a likely mechanism in the UV region for the responsivity enhancement. For the GaN UV detectors, when light is incident on the Ag nanoparticles, collective oscillations of the conducting electrons takes place because of the oscillating electromagnetic field of the light. The subsequent polarization effects and restoring forces lead to a resonance behavior. As a result, larger scattering cross sections are observed because of this plasmonic scattering effect, leading to more electron-hole pairs generated and thus a higher responsivity.

Using Mie theory, the scattering cross sections $C_{sca}$ can be calculated from Equation 2:

$$ C_{sca} = \frac{1}{6\pi} \left( \frac{2\pi}{\lambda} \right)^2 |q|^2 $$

where $\alpha$ is the polarizability parameter, both for the spherical and ellipsoidal metallic nanoparticles, which is proportional to square of the volume ($V$) of the nanoparticles. Thus, at certain wavelengths, the scattering cross section both of spherical and ellipsoidal Ag nanoparticles satisfies Equation 3, i.e., $C_{sca}$ is proportional to square of the nanoparticles' volume $V$.

$$ C_{sca} \propto V^2 $$

According to Equation 3, larger-size nanoparticles have higher scattering efficiency than smaller ones. The as-deposited Ag nanostructures grow to larger size Ag nanoparticles after annealing (as shown in Figure 1), and hence the scattering effect is correspondingly larger from these annealed Ag nanoparticles. This explains why the responsivity of the detectors with annealed Ag nanoparticles was further enhanced (as illustrated in Figure 2). Furthermore, the surface plasmon resonant frequency of Ag nanoparticles can be tuned by altering the shape of the Ag nanoparticles. The density and shape of the Ag nanoparticles also changed after annealing making it possible that the plasmon resonant frequency of the annealed Ag nanoparticles is tuned closer to the response frequency of the GaN UV detectors. This in turn would lead to stronger coupling between the response frequency and the plasmon frequency which is what appears to have happened. This thus provides another explanation for the higher responsivity of the detectors with annealed Ag nanoparticles.

In summary, high-responsivity GaN UV detectors have been realized by exploiting Ag nanoparticles’ plasmon enhancement. The highest responsivity of these detectors is increased by about 30 times with annealed Ag nanoparticles on the GaN surface. The high responsivity can be attributed to the plasmonic scattering effect of these Ag nanoparticles. Considering that achieving a high responsivity has been one of the most important challenges for the application of UV detectors, the results reported here are very significant for advancing the application of GaN-based UV or even deep UV optoelectronic devices.

**Experimental Section**

The ~3 μm thick undoped GaN epilayer was grown on (0001) sapphire substrate by metal-organic chemical vapor deposition (MOCVD) at 1050 °C. Prior to depositing the GaN layer, the sapphire was thermally cleaned under H$_2$ for 10 min at 1100 °C and then the temperature was decreased to 550 °C and a ~25 nm thick GaN buffer layer was deposited. Trimethylgallium and ammonia were used as Ga and N precursors, respectively. Before fabricating GaN UV detectors, Ag metal was deposited on the surface of the GaN epilayer by electron beam evaporation under a pressure of about 5.4 × 10$^{-6}$ mbar. After the Ag was deposited, some of the samples were subsequently annealed at 800 °C for 5 min in order to form Ag nanoparticles and N$_2$ was used to

![Figure 5](https://www.advmat.de) Temporal response of the three detectors measured by shutting on/off the 360 nm light under a 5V applied bias.

![Figure 6](https://www.MaterialsViews.com) Schematic depicting the scattering effect arising from the Ag nanoparticles at the GaN surface.
prevent oxidation of the Ag nanoparticles. The Ni/Au (20 nm/40 nm) Schottky contacts were fabricated by vacuum evaporation both for the SBH measurements and the GaN MSM interdigitated contacts and then the samples were treated by rapid thermal annealing at 500 °C for 300 s. For the MSM structure studied here, the fingers were 100 μm long and 10 μm wide with a spacing of 10 μm. The devices were fabricated by using standard photolithography to pattern photoresist for subsequent metallization and liftoff to form metal Schottky contacts on the GaN.

The morphology of the samples was characterized by the multi-mode AFM and Hitachi S4800 field-emission scanning electron microscope. For the measurement of the photo response, a 150W Xe arc lamp, a mechanical chopper and a lock-in amplifier were employed. The system was calibrated with a UV-enhanced Si detector. A Keithley 237 electrometer was used to evaluate the current–voltage (I–V) performance of the GaN UV detectors. All the measurements were carried out at room temperature.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements
This work was partly supported by the National Key Basic Research Program of China (973 Program, Grant No. 2011CB301901), the National Natural Science Foundation of China (Grants Nos. 51072195 and 51072196) and the National Natural Science Foundation of China (Grant Nos. 51072195 and 51072196) and the National High-tech R&D Program (863 Program, National Natural Science Foundation of China (973 Program, Grant No. 2011CB301901), the National Key Basic Research Program of China (973 Program, Grant No. 2011CB301901), and the National High-tech R&D Program (863 Program, Grants No. 2011AA03A111).

Received: July 6, 2011
Published online: January 2, 2012