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Enhanced light collection from a gallium nitride color center using a near index-matched solid immersion lens

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Among the wide-bandgap compound semiconductors, gallium nitride is the most widely available material due to its prevalence in the solid state lighting and high-speed/high-power electronics industries. It is now known that GaN is one of only a handful of materials to host color centers that emit quantum light at room temperature. In this paper, we report on a bright color center in a semi-polar gallium nitride substrate, emitting at room temperature in the near-infrared. We show that a hemispherical solid immersion lens, near index matched to the semiconductor, can be used to enhance the photon collection efficiency by a factor of 4.3 ± 0.1, whilst improving the lateral resolution by a factor equal to the refractive index of the lens.

Color centers in semiconductor materials are promising sources of non-classical photon states. Owing to their localised energy levels, which are embedded deep within the bulk electronic band gaps of their host, they combine the optical properties of single atoms with the scalability of a solid-state environment. Indeed, enhanced electronic confinement arising from mid-gap energy levels in wide-bandgap semiconductors enables single photon emission at room temperature and above. A number of such color centers have been discovered, including in diamond, silicon carbide (SiC), aluminium nitride (AlN), gallium nitride (GaN), and hexagonal-boron nitride (h-BN). Discovery of these color centers has lead to impressive demonstrations of quantum technologies, including: nanoscale magnetic sensing, nanoscale quantum thermometry, quantum repeaters, coherent control of isolated quantum spin states, a room temperature quantum light emitting diode, and the first room temperature continuous maser. Due to the recent discovery of color centers in III-nitride materials, there is renewed interest in the nitrides as a platform for quantum optics.

GaN is arguably the most commercially important wide-bandgap semiconductor due to its prevalence in solid state lighting and high power electronics industries. Such industrial interest has accelerated research into GaN, resulting in considerable expertise in epitaxial deposition of complex structures, mature substrate processing and purity, integration of silicon and an understanding of material properties. However, it wasn’t until recently that color centers in GaN were explored as quantum light sources, with emission energies spanning the visible spectrum and the near infrared. The large spread in emission energy, as well as differing spectra, suggests these color centers originate from different complexes. Although previously attributed to nanoscale cubic inclusions in the wurtzite host, a report from Nguyen et al. compared various growth regimes and concluded that the emission was likely due to a point defect or impurity, similar to color centers in diamond and silicon carbide. Theoretical studies of 2-site color centers, such as a substitutional nitrogen atom adjacent to a vacancy, predicted emission in the 1.5 eV to 2 eV range would be observed from more than a dozen configurations. The physical properties of these color centers have yet to be fully understood.

Due to the high refractive index of gallium nitride \((n_1 = 2.35 \text{ at } 815 \text{ nm})\) single photon extraction is limited by refraction and total internal reflection (TIR) at the interface between the semiconductor and free space. Therefore, a number of enhancement schemes have been investigated and successfully exploited to enhance the extraction efficiency. One of the most straight-forward methods is to exploit the geometry and high refractive index of a solid immersion lens (SIL). A SIL is a truncated sphere, commonly of either a hemispherical or Weierstrass geometry, that provides aberration free imaging at one of two aplanatic points. The hemispherical SIL maps the full numerical aperture (NA) of the imaging system into the high index material without refraction at the high-to-low index interface. In addition, the enhancement is inherently broadband and can be applied after pre-selection of a certain color center, with coarse alignment of position. Here, we report the use of a zirconium dioxide \((\text{ZrO}_2)\) hemispherical SIL, which is almost index matched to GaN \((n_{\text{SIL}} = 2.13 \text{ at } 815 \text{ nm})\), to show enhanced imaging and photon collection from a color center in GaN.

The sample under investigation in this report is a semi-polar gallium nitride on sapphire substrate. The crystal structure of c-plane GaN is shown in Fig.1(a), where the shaded plane represents the [1122] plane which forms the sample surface. The sample structure, illustrated in Fig.1(b), is as follows; Half a micron of m-plane GaN is grown on a m-plane sapphire substrate via metal-organic chemical vapour deposition. A silicon dioxide \((\text{SiO}_2)\) layer is deposited and subsequently patterned, forming circular islands in the m-plane GaN. The sample is then overgrown with GaN. Growth conditions are chosen so the [0001] plane overgrows the [1120] plane, which results in the semi-polar [112\overline{2}] surface.

A room temperature scanned confocal fluorescence map of the sample, taken with an excitation wavelength \(\lambda_{\text{exc}} = 532 \text{ nm}\), is shown in Fig.1(c). An optical long pass
filter along with the absorption of the silicon avalanche photodiode (APD) detector used limits the detection window of the collection path of the microscope to between 800 and 1000 nm respectively. The confocal scan map reveals the presence of point-like color centers, whose spatial extent in the image is consistent with a point-like emitter convolved with the diffraction-limited point-spread function of the microscope. This sample displays a much lower density of color centers than c-plane GaN samples that have previously been reported, assisting in the isolation of a single color center. Strongly localised emission below the bandgap suggests the emission originates from one color center. In addition to the color center, the periodic arrangement of embedded disks can be seen in the scan image, due to laser scattering from the SiO$_2$ disks and/or fluorescence from other photo-active defects.

Point-like color centers can be observed in multiple locations in the scan map, both near to and away from the SiO$_2$ disks, suggesting the origin of the emission lies within the semiconductor and not the SiO$_2$. There is often emission between the disks, which display photo-induced bleaching and no antibunching: we do not study these areas further and believe the origin of this emission is different to that of the point like emitters in this report.

We focus in this paper on one representative emitter at the center of the scan that is easy to relocate and is bright as a result of its nanosecond timescale radiative lifetime. The highlighted emitter is used for all measurements presented in this paper. Repeated measurements of the color center over several months have shown no change in its properties.

A polarization resolved measurement, shown in Fig.1(d), where a thin film polarizer in the collection path of the microscope was rotated in the plane of the sample, shows dipole-like emission from the color center. We hypothesise that the non-perfect extinction of the polarisation suggests an out of plane component arising from a single dipole aligned at an angle of $\phi = 29.2 \pm 0.7^\circ$ to the sample plane. We note that the non-perfect extinction may also be a result of multiple emission dipoles similar to what is seen for the NV color center. The azimuthal angle $\phi$ is determined from the fit using Eq.1, where the emitted intensity $I$ is a function of the in-plane dipole angle $\theta$ and $\phi$, with $n$ as a normalisation factor.

$$I(\theta, \phi) \propto n[1 - \sin^2(\theta)\cos^2(\phi)]$$

A spectral measurement of the color center, taken under 532 nm excitation, is presented in Fig.1(e). The spectral measurement is fit using two peaks. A clear zero-phonon line
(ZPL) centred at 815 nm can be identified, which arises from
the optical excited-to-ground state transition of the defect
without coupling to high energy phonons. The symmetric
broadening around the ZPL is consistent with coupling to
low energy acoustic phonons. The linewidth of the ZPL
is measured as 5.75 ± 0.03 nm. The ZPL is accompanied
by a red-shifted phonon side band (PSB). The ZPL and the
contributions from phonon assisted transitions overlap in
the spectrum at room temperature, consistent with other
color centers in the sample. The portion of the intensity
situated within the ZPL, including the symmetric broadening
due to acoustic phonons, is 39%. The overall spectrum is
significantly narrower than that observed from the NV center
in diamond and AlN color centers, in which the phonon
sideband dominates the spectrum with a room temperature
broadening greater than 100 nm.

The photon statistics of the light are measured in the
second-order correlation measurement presented in Fig.1(f),
taken at 110% saturation power for the emitter. Strong anti-
bunching occurs on a 0.90 ± 0.02 ns timescale while bunching
can also be observed on a 37.2 ± 0.9 ns timescale. The insert
shows the antibunching in more detail. A three-level energy
model is used to fit the data, where the proposed third level
represents a metastable or "shelving" state, consistent with
other works on single room temperature color centers.2,11,12
The finite value of the correlation function at zero delay can
be accounted for due to imperfect filtering of laser scatter
and sapphire fluorescence beyond 750 nm. We do not correct
for background fluorescence in the measurement. This
poissonian background light could be reduced by narrowing
the range of the spectral filtering.

To enhance the light collection from the emitter studied
in Fig. 1, we deterministically position a 1 mm diameter
hemispherical SIL directly on top of the emitter presented in
Fig.1 using a micromanipulator. This sample was patterned
with a regular array of labelled crosses in a thin metallic
layer, to allow the position to be accurately determined during
placement, with an accuracy of ±10 µm. A high refractive
index mounting media (Cargille Meltmount™, n = 1.704)
fills the gap between the sample and SIL.

The increased NA within the semiconductor allows smaller
features to be resolved in the lateral direction,25 where
the Rayleigh limit is proportional to 1/NA. Similarly, the
increased NA reduces the Rayleigh range of the focused
beam, allowing smaller features to be resolved in the axial
direction. In addition, an index-matched SIL increases the
magnification by a factor of nSIL in the lateral direction. The
magnification within the SIL can be seen in the optical image
in Fig.2(b), by comparing the apparent sizes of the patterned
crosses visible under the SIL with those in the bottom left
hand side of the image.

For an aberration free confocal imaging system, the diffrac-
tion limited resolution is given by the Rayleigh criterion;

\[ \Theta_{FWHM} = 0.5014 \frac{\lambda_c}{NA} \]  

where \( \lambda_c \) is the collection wavelength, in this case 815 nm.
The increased resolution that can be achieved with a SIL,
over a sample with a flat surface, is a result of the increased
numerical aperture of the light coupled into the semiconduc-
tor, \( NA_{SIL} = n_2 \times NA_{Imaging} \).

The enhanced resolution is illustrated in Fig.3 which
includes two scans over the same angular range, with and
without the SIL. The embedded SiO₂ disks can be seen in
both scan maps and are highlighted with the dashed blue lines.
Clear magnification of the image can be seen evidenced by
the increased disk size, by a factor of \( n_{SIL} \). The magnification
is also made apparent in the optical image shown in Fig.2(b),
in which one can compare the angular dimensions of identical
lithographic features beneath the SIL to those on the flat sur-
face. In the two scans in Fig.3 the increase in magnification
of the imaging system with the SIL cancels the increased
resolution, resulting the emitter having the same angular size.
X and Y slices across the emitters are fitted using a Gaussian
function and the FWHM compared to the expected spot size
is presented in Table I. The uneven sample surface and the
chromatic behaviour of the objective prevents our imaging
system reaching the resolution limit, but nevertheless,
The extraction of light from a high refractive index material close to that of the lens as all light exits the SIL normal to the SIL-to-air interface, therefore increasing the number of photons collected.

For a dipole source orientated in-plane it is possible to determine the collection efficiency into a lens of numerical aperture NA=0.75 using an analytical expression, as presented by Barnes et al.:

\[
\eta = \frac{1}{32} \left[ 15 \left( 1 - \sqrt{1 - \sin(\theta)^2} \right) + \left( 1 - \cos(3\theta) \right) \right] \theta_a \tag{3}
\]

where, for a color center in bulk GaN;

\[
\theta = \arcsin\left( \frac{NA}{n_1} \right) \quad \text{and} \quad \theta_a = \frac{4n_1n_3}{(n_1 + n_3)^2}
\]

\[\therefore \eta_{\text{bulk}} \approx 3.2\%\]

and with the SIL;

\[
\theta = \arcsin\left( \frac{n_{\text{SIL}}NA}{n_1n_3} \right) \quad \text{and} \quad \theta_a = \frac{16n_1n_{\text{SIL}}^2n_3}{(n_1 + n_{\text{SIL}})^2(n_{\text{SIL}} + n_3)^2}
\]

\[\therefore \eta_{\text{SIL}} \approx 15.3\%\]

\(T_a\) is an approximation of the transmission due to Fresnel reflections at the GaN-to-air and SIL-to-air interfaces respectively. Therefore, we predict an improvement in the collection efficiency with the SIL equal to \(\times 4.8\). The collection efficiency as a function of the numerical aperture for a dipole in air (grey, dashed), in GaN (green) and with the SIL (blue) is illustrated in Fig.4(a). The significant enhancement of the collection efficiency with the SIL is apparent as the blue plot approaches that of the ideal case of a dipole in air.

To determine the expected collection efficiency enhancement for a dipole with a finite azimuthal angle we turn to Finite Difference Time Domain (FDTD) simulations using commercial software (Lumerical). The GaN epilayer is considered to be a homogeneous dielectric layer without the SiO\(_2\) disks and no absorption. The electric field profile is measured in three dimensions around the dipole. The near-field electric field profile above the surface in the XY plane is projected into the far-field and integrated across the half angle of the collection optics (NA=0.75). Due to the geometry of the SIL, the near field electric field profile is collected in the ZrO\(_2\) dielectric and projected into the far field. The reflection at the SIL-to-air interface is accounted for using \(T_a\) in Eq.3. Due to the unknown thickness of the adhesion layer used in the experiment, we do not account for the Meltmount layer in the simulation. The results therefore present an upper bound for the collection efficiency enhancement for an emitter with an azimuthal angle of 29.2 ± 0.7° when imaging from the surface.

As illustrated in Fig.4(b), where the dipole is in the plane of the sample (\(\phi = 0\)), the CE agrees with the analytically determined value in Eq.3 with and without the SIL, with a CE of 16.3 and 3.4% respectively. The expected enhancement at the determined azimuthal angle \(\phi = 29.2 ± 0.7°\) is slightly increased from the in-plane case, with an enhancement of \(\times 5.2\).
The measured collection efficiency enhancement is shown in (c), where the power dependent intensity of the color center is presented with and without the SIL. The data is fit with a saturation function, \( I(P) = \frac{P}{P_{\text{sat}}} \), where \( I_\infty \) is the count rate at infinite power and \( P_{\text{sat}} \) is the power required to saturate the emitter. \( I_\infty \) is used to quantify the enhancement, where \( I_\infty \) with and without the SIL is 520 ± 10 kcps and 121 ± 3 kcps respectively, an improvement equal to \( \times 4.3 \pm 0.1 \). The discrepancy with the predicted value may be a result of the buried SiO\(_2\) disks and rough sample/SIL surface.

We investigated the properties of color centers in semi-polar GaN, demonstrating room temperature quantum light in the near infrared from a color center with a dipole-like emission pattern. Enhanced imaging of a single emitter in a semi-polar GaN sample was achieved by spatially aligning a SIL on top of a color center. The use of the hemispherical SIL also increased the photon collection efficiency by a factor of 4.3 ± 0.1, such that the efficiency approaches that of a dipole in free space. The combination of existing expertise in GaN processing and epitaxy, with enhanced photon extraction enabled by this easy-to-implement SIL, can lead to efficient off-the-shelf quantum light sources operating without cryogenic cooling, which will be highly beneficial for future quantum technologies.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are openly available in the Cardiff University Research Portal at http://doi.org/[doi], reference number[reference number].

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