Laser written nitrogen vacancy centers in diamond integrated with transfer print GaN solid immersion lenses

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ABSTRACT

Laser-written nitrogen vacancy (NV^-) centers are combined with transfer-printed GaN micro-lenses to increase fluorescent light collection by reducing total internal reflection at the planar diamond interface. We find a 2x improvement of fluorescent light collection using a 0.95 NA air objective at room temperature, in agreement with FDTD simulations. The nature of the transfer print micro-lenses leads to better performance with lower numerical aperture (NA) collection, as confirmed by results with a 0.5NA air objective which show improvement greater than 5x. The approach is attractive for scalable integrated quantum technologies.

Keywords: Solid state defects, laser writing, NV centers, diamond, additive GaN micro-optics, transfer printing, quantum systems

1. INTRODUCTION

In contemporary quantum technology research, solid-state quantum defects in bulk substrates have emerged as pivotal components, especially in fields such as quantum information science,[1,2] quantum sensing,[3] and quantum imaging. The nitrogen-vacancy (NV^-) centers, exemplary of solid-state defects, are increasingly becoming a focal point of research interest in the field of quantum material science, primarily attributed to their remarkable room-temperature stability, promising spin coherence time and potential for scalable fabrication. However, the collection efficiency of photons emitted by quantum emitters such as NV^- centers is a major limitation for many quantum applications. The high refractive index mismatch between diamond and medium results in a considerable proportion of emitted photons being reflected internally, which causes a significant loss of total number of photons emitted from an NV^- center. To address this limitation, several techniques have been developed to minimize the effect of refractive index mismatch on emitters inside diamonds, including surface treatments,[4] cavity coupling[5,6] and nanostructuring of diamond.[7,8] Here we expand upon our recent work using GaN micro lenses as SILs (Solid immersion lens) to minimize the effect caused by the high refractive index mismatch.[9] These micro lenses are back-end integrated to known NV^- centers by transfer-printing. Comparing to traditional NV^- centers fabrication techniques such as ion implantation[10,11] and electron beam irradiation,[12,13] direct laser writing[14,15] has a unique advantage of precise three-dimensional positioning due to the nonlinear light-matter interaction mechanism which makes this the ideal technique to integrate with these micro lenses.

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2. FABRICATION AND LENS INTEGRATION

The diamond used is commercially available electronic-grade single crystal grown by chemical vapour deposition (nitrogen density < 5 ppb). The diamond substrate is patterned with laser written alignment marks comprising 60um squares with 4 quadrants each. All laser written alignment marks are located in regions where with no intrinsic NV^- centers or other colour centers' presence. Each quadrant contains an array of 5x5 laser irradiated sites with 5 μm spacing to the nearest neighbour and well-aligned to the additional extruding surface alignment markers. A single sub-picosecond laser pulse irradiates each site, with the writing-laser pulse energy successively lowered between each row of the array. This results in mainly creating graphitisation points in the upper two quadrants and the statistical creations of nitrogen vacancy centers in the two bottom quadrants after annealing, as described previously.[14,15] After pre-characterization by photoluminescence imaging, specific NV^- centers are selected for integration with GaN micro-lenses.

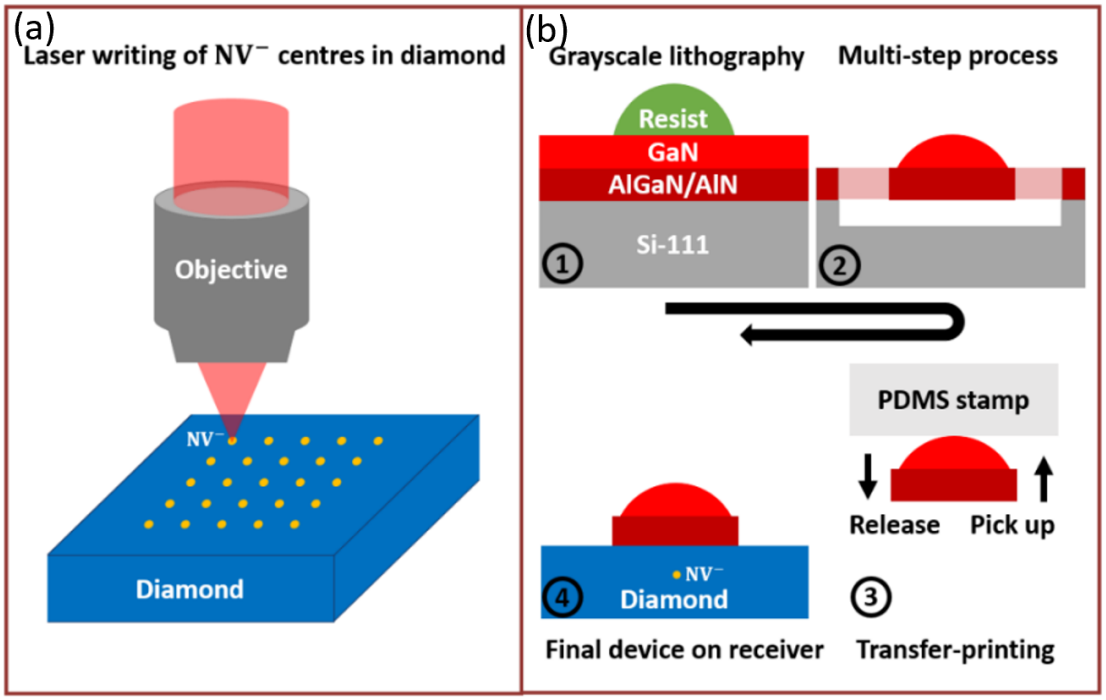

Figure 1. Schematics describing the process flow, a) NV^- defect centers are generated in single crystalline diamond by laser writing and successive annealing, b) suspended GaN solid immersion lenses are fabricated by parallel plasma dry and wet etching techniques and assembled above the defect centers with μ-transferprinting.

Fig. 1 depicts a summarized schematic of the entire process flow, further details are described in previous work.[9] (1) Polymer resist lenses are defined via grayscale lithography and thermal reflow above the GaN lens layer and AlGaN/AlN buffer layer. (2) Multi-step processes involving ICP lens etching and KOH lateral SiOx hard mask wet etching to form GaN and AlGaN/AlN buffer layer SILs. (3) GaN SILs are deterministically transfer-printed to pre-assigned positions by patterned polydimethylsiloxane micro-stamp (6:1 Sylgard 184 PDMS) with sub-micron lateral precision. (4) GaN SILs are back-end integrated with prefabricated NV^- centers. Using different objective lenses (NA = 0.95 and 0.5) we obtain a photoluminescence map of one of the fabricated arrays after lenses integration at room temperature, as shown in Fig. 2. We place a single GaN micro-lens in each quadrant and use the map to adjust the print position according to the most promising emitter location. Thus, we demonstrate three-dimensional deterministic matching of emitter's position and SIL's position and shape.

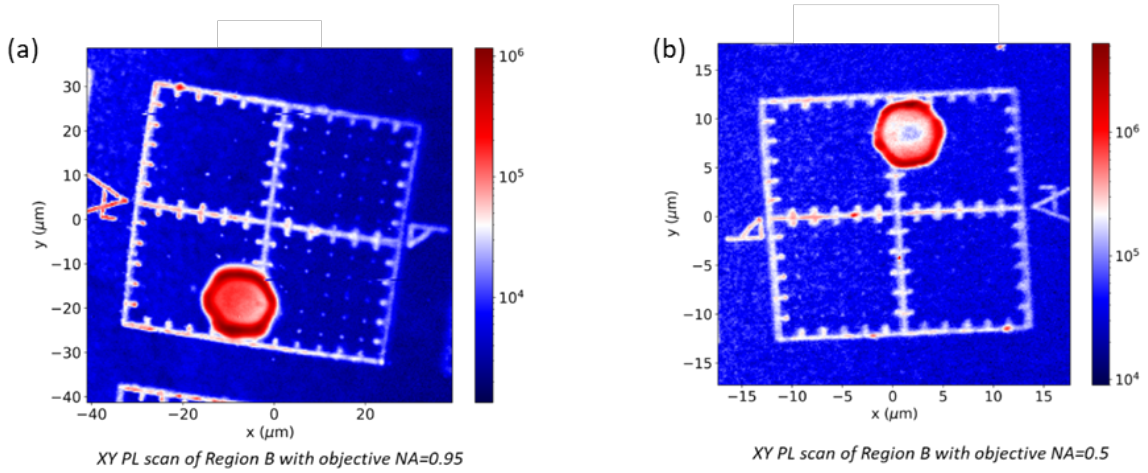

Figure 2. Two-dimensional Photoluminescence mapping of post laser patterned array after lens integration with (a 0.95 NA objective lens and b) 0.5NA objective lens.

3. RESULTS AND SIMULATIONS

A home-built confocal microscope is used to characterize the performance of the integrated GaN SILs. We collected photoluminescence (PL) maps, Hanbury Brown-Twiss (HBT) measurements, spectral measurements, and power saturation measurements of the NV^- center under different objective lenses with varying numerical apertures (NA) (NA = 0.5, 0.95. 1.25) using a 532 nm continuous-wave (CW) laser (GEM 532) as excitation source. The fluorescence signal from the NV^- center is collected back through the same objective lens in epi-fluorescence and spectrally filtering off the excitation laser and Raman signal associated with excitation before detection. Fig. 3a shows the photoluminescence images of one NV^- center doublet (pair of NV^- centers) taken with different objective lenses with and without GaN SIL in place. Results show that the visibility of NV^- centers is lowest with a 0.5 NA objective lens, with the written site being barely visible, as expected due to the limited collection angle. As the NA of the lens is increased to 0.95 and 1.25, the visibility of the NV^- centers increased significantly. Additionally, after the transfer of the GaN SILs on top of the same NV^- sites, the visibility and count rates of the NV^- centers are enhanced. To confirm the GaN SILs' compatibility with measurement in the quantum region, g2 autocorrelation measurement is taken with the SIL in place as shown in Fig. 3c. Since both photon bunching and anti-bunching behavior is observed, this indicates the presence of a metastable state populated from the optically excited state,[16] g2 autocorrelation data is fitted to a three level-system as indicated in Equation (1). Under high excitation intensity, the lifetime of the emitter is estimated to be 10.1 ns from τ_1, in good agreement with literature [16]. The utilization of solid immersion lenses (SILs) enhances the feasibility of autocorrelation measurements for dim emitters with count rates below $10^4\,s^{-1}$. The spectrum in Fig. 3d shows a typical emission spectra of the written defect center which exists a zero-phonon line (ZPL) centered at 637 nm with a phonon sideband of approximately 100 nm, which is characteristic of the NV^- center in diamond.[17]

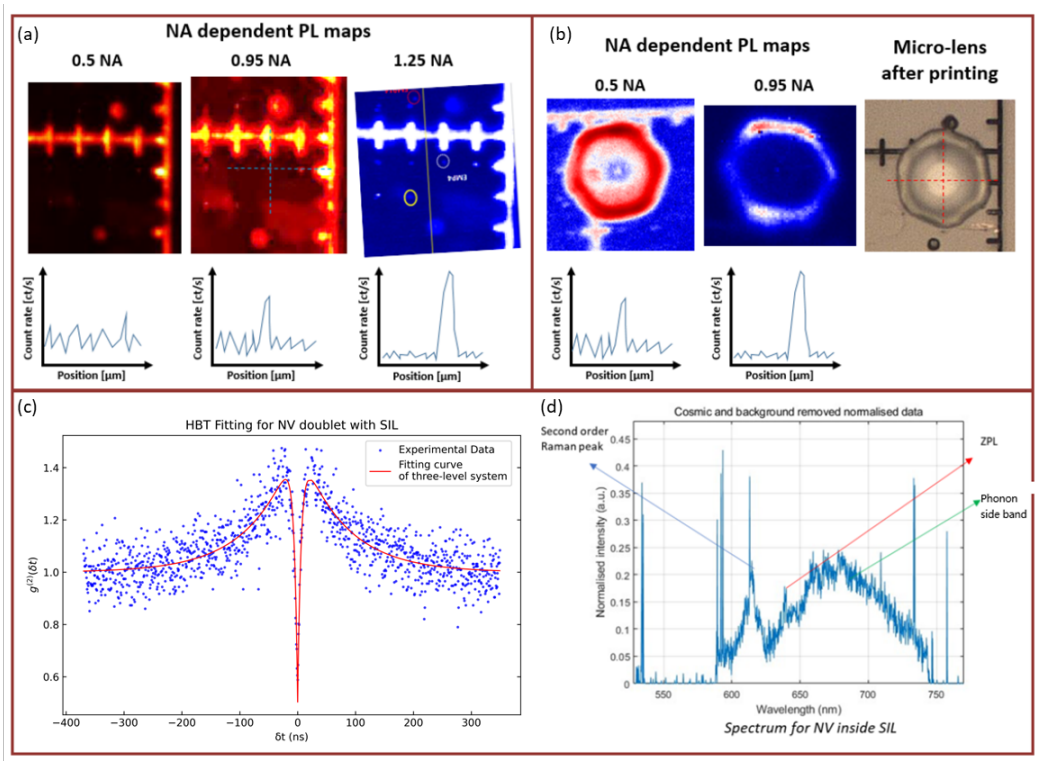

Figure 3. a) Confocal photoluminescence maps focusing on the same emitter after laser writing and annealing and b) after GaN SIL integration in dependence of the numerical aperture of the collecting objective lens system, line profiles are taken along the alignment marks, c) normalised autocorrelation and spectral data from the emitter after SIL integration using the objective with NA = 0.95 and fitting curve of a three-level system, d) room temperature spectrum of a typical NV^- center measured with lens integrated.

$$g2(t) = 1 - a \cdot \exp\left(-\frac{|t|}{\tau_1}\right) + (a-1) \cdot \exp\left(-\frac{|t|}{\tau_2}\right) \tag{1}$$

Further to quantify the enhancement factor of the GaN SILs, power saturations of the laser written NV^- centers are measured (Fig. 4a). Since the NV^- centers are confirmed to be doublet, power saturation curves are fitted to a two-level quantum system indicated in Equation (2). With this, enhancement factors of 2.2x and 1.8x are obtained, in good agreement with the FDTD results displayed in Fig. 3b for objective lens with 0.95NA that predict an enhancement factor of 2.5x.[9] The variations are mostly likely to be aberration and misalignment of the SILs. Even though the lateral mismatch is within submicron level, the vertical mismatch of the GaN micro lens and NV^- center can cause a 20% signal enhancement loss[8]. The nature of the transfer print SILs leads to better performance with lower NA collection. Light collection enhancement factor for 0.5NA objective is estimated to be larger than 5x by comparing the count rates of the same NV^- center with the same pumping power. At higher light cone angles (essentially at higher numerical aperture), the light extraction enhancements are higher with a monolithic SIL, fabricated via a focused ion beam (FIB) removal of the diamond material, compared to GaN lens, since GaN/AlGaN lens structure can not maintain the emitted light wavefront without strong curvature towards the surface normal due to refraction at higher collection angles.

$$I(P) = \frac{I_{\text{sat1}} \cdot P}{P + P_{\text{sat1}}} + \frac{I_{\text{sat2}} \cdot P}{P + P_{\text{sat2}}} \tag{2}$$

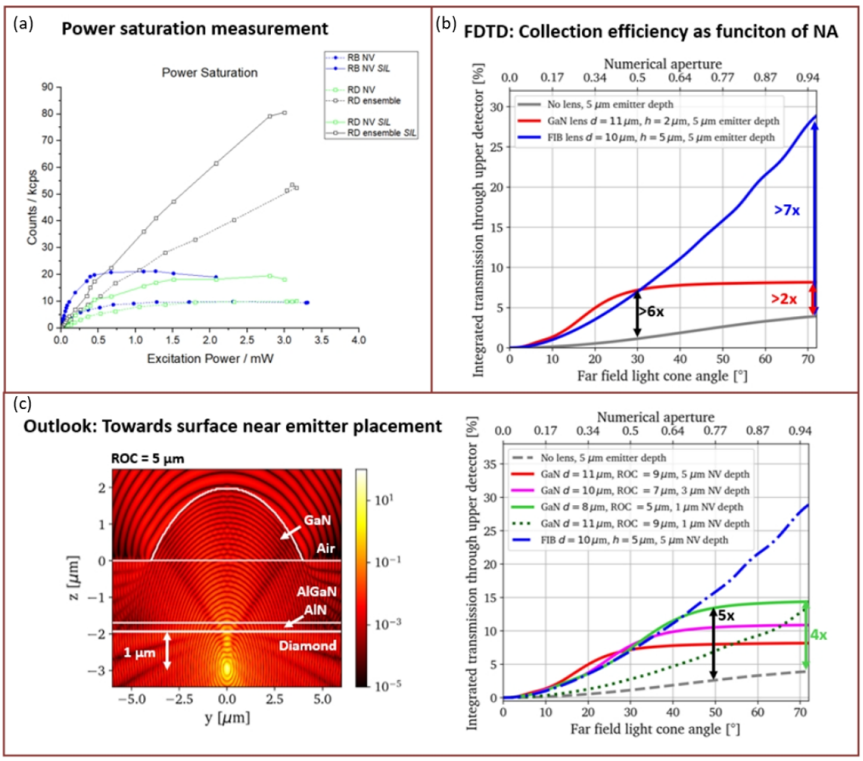

Figure 4. a) Fitted power saturation measurements of two doublet NV^- centers with and without GaN SIL using an objective lens with NA of 0.95, b) expected free space collection improvement in dependence of the objective lens' NA, c) cross section though a simulation region with a GaN SIL matched to a dipole emitter in 1 μm depth at $\lambda = 700$ nm wavelength and the expected corresponding free space collection improvement in comparison to a matched lens fabricated with focused ion beam milling.

4. CONCLUSIONS AND OUTLOOK

Building upon prior research,[9] here we advance the discussion on the application of transfer printing GaN microlenses for the enhancement of photon collection efficiency from solid-state emitters, exemplified by laser-written negatively charged nitrogen vacancy centers. The enhancement factor is in good agreement with the FDTD simulation results and the emitter's photophysics properties are preserved after GaN SIL integration. Even though the enhancement performance of the transfer printed GaN SILs may not match that of the FIB-fabricated monolithic SILs for higher numerical aperture objective lenses, transfer printed SILs still have their unique advantages of faster fabrication speed, lower residual damage to the substrate and non-invasive reversible integration nature. Further increasing the enhancement factor of the GaN SILs can be achieved by integrating with emitters closer to the substrate surface and increase the GaN epilayer thickness, since these enable the lens aperture to cover more angular space above the emitter and at the diamond-air interface so that its enhancement is comparable to that of monolithic SILs. Future works of laser writing of emitters after GaN lens printing is ongoing, this approach holds the exciting promise of deterministically creating surface quantum defects. Combining the approach of multi-stamp head and automation of transfer printing and laser writing process, rapid large scale on-chip device arrays fabrication is conceivable.

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