

Enhancing Quantum Emission from Spin Defects in Hexagonal Boron Nitride with a Plasmonic Nanocavity

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Abstract: We report a 250-fold photoluminescence enhancement of V_B^- spin-defects in hBN by coupling them to nanopatch antennas (NPA). Considering the relative size of the NPAs and laser-spot, an actual enhancement of 1695 times is determined. © 2023 The Author(s)

1. Introduction

The field of solid-state quantum emitters (QEs) in two-dimensional materials like transition metal dichalcogenides (TMDCs) and hexagonal boron nitride (hBN) has evolved rapidly following the discovery of single photon emitters in monolayer hBN [1]. Unlike their bulk counterparts, QEs in two-dimensional host materials offer significant advantages for integration in photonic and plasmonic structures. Previous reports have also demonstrated that the properties of QEs in hBN can be controlled via strain and electric field tunable optical constants, photoluminescence ranging from ultraviolet (UV) to near-infrared emission (NIR), and optically active spin defects [1,2].

The negatively charged boron-vacancy center V_B^- in hBN has attracted significant interest for quantum sensing applications. It has been shown that the ground-state of the defect is a spin-triplet state with a ground-state splitting $D_{GS}/h \sim 3.5$ GHz and a spin-coherence time $T_2 \sim 1$ μ s [2,3]. Interestingly, the spin coherence time is preserved for ultrathin hBN with a shallow implantation depth of ~ 3 nm [3]. These attractive features, combined with its facile integration with other Van der Waals materials [4], can potentially enable ultrathin quantum sensors that can be placed at $\sim$ nm distances from materials of interest. Despite their great potential, the practical applications of V_B^- defects are limited by their poor quantum efficiency and low photoluminescence (PL) signal strength. Therefore, improving the signal strength by coupling the defects with plasmonic structures is critical. Previous works have demonstrated this effect by coupling the defects with gold films and nano-patch antenna (NPA) cavities [3] to demonstrate a 17-fold PL enhancement. In this work [5], we show that the overall PL-strength can be significantly improved by coupling the defects with NPA cavities by optimizing the plasmonic cavity structure and employing low-loss optical materials. We achieve an overall intensity enhancement factor ~ 250 times while retaining an ODMR contrast of 6%, which corresponds to an actual intensity factor ~ 1695 , considering the relative size of the probe laser with respect to the size of the cavity.

2. Results

We optimized two parameters in the design of the nano-patch antenna cavity using finite element method COMSOL simulations: (i) The implantation depth of the defects in the hBN flake, which determines the location of the defects in the cavity structure (ii) the thickness of the hBN flakes and the alumina spacer layer. Based on these considerations, we determined that the highest PL enhancement can be obtained for 6 nm hBN flake with defects implanted at a depth of 3 nm from the surface. Suitable hBN flakes were exfoliated on standard SiO_2/Si substrates, identified using optical contrast, and irradiated uniformly using helium ion implantation. The resulting flakes were characterized using a 532nm pump laser in a confocal microscope setup. We measured the saturation curve, spectrum and ODMR on the silicon substrate. The flake was then transferred to an epitaxial silver substrate with a 3 nm alumina spacer layer using standard dry-transfer methods. The silver substrate is lithographically patterned to form a microwave waveguide suitable for ODMR measurements in 3-5 GHz range (Figure 1a). Following this, we drop-casted silver nanocubes with an edge length of 100nm on the flakes to form nano-patch antennas.

We measured the PL spectrum, saturation, and ODMR from defects coupled with the NPA cavities (shown in Figures 1d, e, f), such as the spot circled in the AFM map in Figure 1b and PL map in Figure 1c. Comparing the saturation

curves for the defects measured on a silicon substrate and the NPA cavity, we determine that the overall PL enhancement is nearly 250 times (Figure 1e). We also measure the saturation curve for defects on the silver film but outside the nano-cavity. The actual PL enhancement for spin-defects under the Nano-patch antenna cavity of ~ 1695 times was retrieved from these results. We measured a 6% ODMR spin contrast for defects coupled with NPA cavity using the microwave waveguide at 40 mW microwave driving power and 30 μ W laser excitation power (Figure 1f). We estimate that the sensitivity of these defects to external magnetic fields is $\sim 138 \mu T/\sqrt{\text{Hz}}$ in CW-ODMR measurements. The sensitivity can be significantly improved by optimizing the laser driving power and microwave power to yield a higher spin contrast.

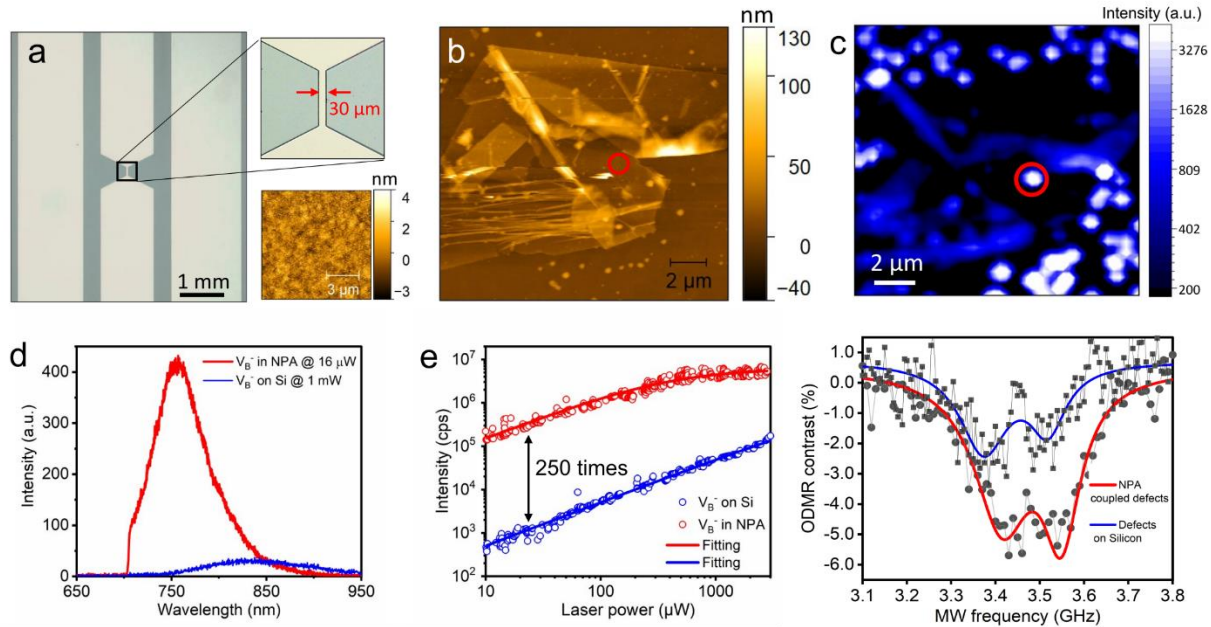


Figure 1. (a) A schematic for silver waveguide used in the experiment. Enlarged optical view of the central neck region of the waveguide, showing a neck width of 30 μm . (bottom right) AFM scan from the neck region of the waveguide. A surface roughness of 0.91 nm is obtained. (b) An AFM scan of the hBN flake placed on silver. The wrinkles were formed by the transfer process for thin flakes. (c) A PL scan of the hBN flake shown in (b) after random NPA structures are formed. (d) Spectra of the hBN flake shown in (b) when it is on the silicon substrate (blue curve. Laser excitation power: 1 mW, 700 nm long-pass filter) and coupled with the NPA circled in red in (c) (red curve. Laser excitation power: 16 μW). (e) Power-dependent photoluminescence (PL) intensity of the hBN flake placed on the silicon substrate (blue dots) and coupled with the NPA circled in red in (c) (red dots). Solid lines are fitting curves. Intensity enhancement of ~ 250 times is measured over the tested laser power range. (f) Spin contrast of boron vacancy defects in the hBN flake placed on silicon (blue fit measured at 1 mW laser power) and coupled with the NPA structure (red fit measured at 30 μW laser power). Solid lines are fitting curves.

3. Conclusion

We demonstrate that the PL intensity of the spin defects in hBN can be enhanced significantly by coupling them with NPA cavity structures and driving the defects with microwave waveguides. Our results demonstrate an order of magnitude improvement over the previous results while preserving a 6% ODMR contrast paving the way for practical quantum sensors with spin defects in hBN.

4. References

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This work is supported by the U.S. Department of Energy (DOE), Office of Science through the Quantum Science Center (QSC), and the National Science Foundation Award 2015025-ECCS, DMR-1747426.