Plasmon-enhanced Quantum Emission from Spin Defects in Two-dimensional Hexagonal Boron Nitride

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Abstract: We demonstrate 120-fold photoluminescence enhancement of V_B^- spin defects in hBN by coupling them to nanopatch antennas. Since the laser spot is 6.25 times larger than the antenna area, the actual enhancement is 750-fold.

1. Introduction

The field of atomically thin layered materials has expanded rapidly following the discovery of graphene. This transition from bulk crystals to two-dimensional (2D) layers has enabled the study of previously inaccessible optical, mechanical, electrical, and magnetic properties [1]. Furthermore, the facile integration of 2D materials into heterostructures using fast pick-up techniques allows the study of light confinement at extreme scales, magnetic proximity effects, spin-charge coupling, etc. In recent years, solid-state quantum emitters (QEs) in 2D hexagonal boron nitride (hBN) have been studied extensively [2, 3]. The 2D nature of few-layer hBN, combined with its chemical and physical robustness, offers unparalleled advantages for integrating these QEs with plasmonic and photonic structures for hybrid quantum devices.

Among the various type of quantum emitters reported in 2D hBN, ensembles of negatively charged boron vacancy centers (V_B^-) have attracted attention as optically active spin defects. Previous results have shown that these defects can be used to map external stimuli such as temperature, pressure, and magnetic fields with capabilities comparable to color centers in 3D systems such as diamond and silicon carbide [4]. The most significant advantage of spin defects in few-layer hBN lies in the possibility of placing shallow implanted spin defects within nanometer distance from samples of interest, thereby enabling advanced quantum sensing applications. One limitation of this system is the low quantum efficiency or brightness of V_B- centers, which has adverse effects on their sensitivity as quantum sensors. To address this challenge, we couple these emitters with a silver nano-patch antenna (NPA) structure, acting as a plasmonic cavity (Fig. 1a). By choosing hBN flakes with an optimized thickness of ~6 nm, we observe the photoluminescence (PL) intensity of V_B^- centers enhanced by up to 120 times when coupled to the NPA, which is nearly an order of magnitude higher enhancement than in previous reports [5]. Our result marks an important step towards using V_B^- defects for practical quantum sensing.

2. Results

Homogeneously distributed V_B^- spin defects are obtained by low-energy helium ion implantation of hBN flakes. An ion energy of 300 eV is used to achieve an implantation depth of ~3.5 nm [5]. Thin hBN flakes are first identified and characterized on Si/SiO_2 substrate, and then transferred onto an epitaxial silver film substrate coated by a 3 nm-thick alumina (Al_2O_3) spacer layer. Finally, single-crystalline silver nanocubes (NCs) with dimensions around 100 nm are drop-cast onto the same substrate to form NPA structures (Fig. 1a). The hBN layer thickness of 6 nm that yields the highest Purcell enhancement is obtained from the full-wave finite element simulations (COMSOL Multiphysics). Fig. 1b shows an AFM image of the hBN flake on silicon, with a nearly optimized thickness (6.5 nm), chosen for the study. Fig. 1c shows the AFM map of the system after NPA assembly, where the bright spots are silver nanocubes. Photoluminescence (PL) spectra of a number of NPA structures are collected under 532 nm continuous-wave laser excitation at a power of 150 μW. Specifically, the spectrum of the NPA circled in red in Fig. 1c shows a peak intensity that is four times larger than that measured from the same flake on a silicon substrate with a laser power of 7 mW (Fig. 1d), indicating PL enhancement by orders of magnitude. The slightly blue-shifted emission peak for the NPA is likely induced by the coupling of boron vacancy defects to the gap plasmon cavity mode.

To better quantify the PL enhancement factor, PL intensities of V_B^- defects at different laser powers are recorded for the same hBN flake on silicon and in the NPA structure. As demonstrated in Fig. 1e, the brightness of V_B^- defects in NPA structures are enhanced by 120 times at over a range of laser excitation powers. Note that due to a laser spot size (~250 nm) larger than the NPA dimension (~100 nm), the actual PL enhancement by the NPA is about $120\times(250/100)^2 = 750$ times. The potential of V_B^- defects for spin-based sensing could be evaluated by their spin contrast (C_{T1}), defined as the relative difference between the fluorescence rates emitted by the $m_s = 0$ and $m_s = \pm1$ states. A spin contrast of 8% is measured for V_B^- defects on silicon substrates (Fig. 1f). After being coupled with NPAs, a reduced but non-zero C_{T1} of ~ 2% is observed.

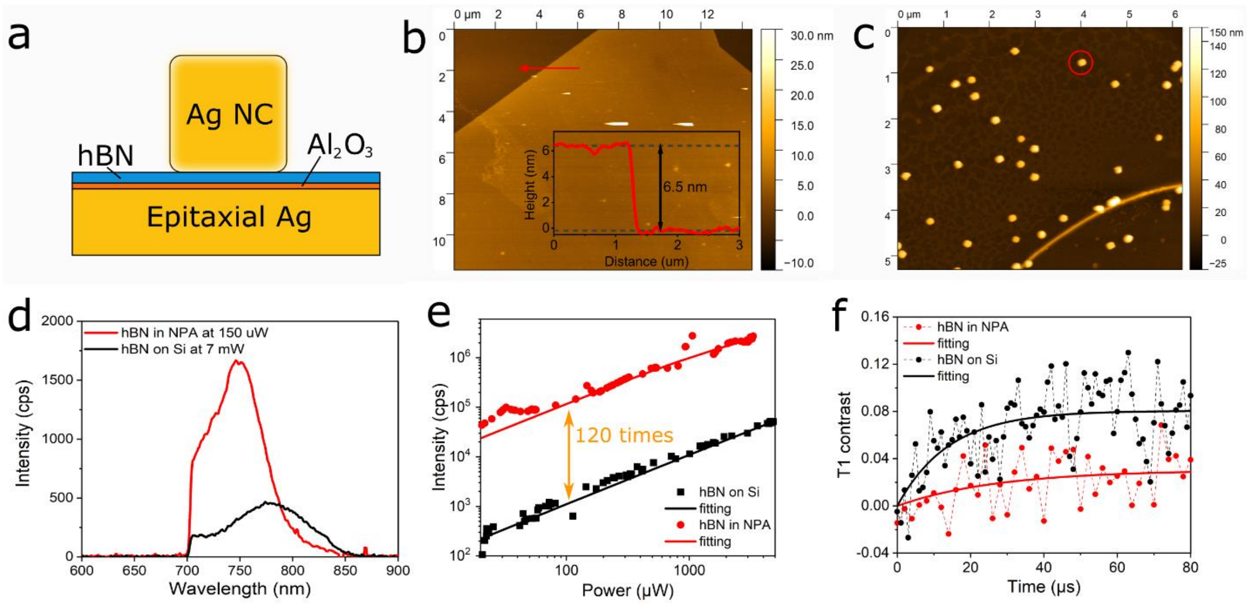

Figure 1. (a) A schematic of the NPA structure fabricated in this work. (b) An AFM scan of the hBN flake placed on silicon. The flake is transferred to the silver film for NPA assembly. Inset: a height profile along the red arrow in the image, showing a flake thickness of 6.5 nm. (c) An AFM scan of part 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 (black curve. Laser excitation power: 7mW, 700nm long-pass filter) and coupled with the NPA circled in red in (c) (red curve. Laser excitation power: 150 μW). (e) Power-dependent photoluminescence (PL) intensity of the hBN flake placed on the silicon substrate (black dots) and coupled with the NPA circled in red in (c) (red dots). Solid lines are fitting curves. Intensity enhancement of ~120 times is measured over the tested laser power range. (f) Spin contrast of boron vacancy defects in the hBN flake placed on silicon (black dots) and coupled with the NPA structure (red dots). Solid lines are fitting curves.

3. Conclusion

We have integrated shallow boron vacancy spin defects in hBN with NPA structures to improve the quantum efficiency of such spin defects. A 120-fold photoluminescence intensity enhancement is observed, which is an order of magnitude higher than previous reports. This result sets a new record for plasmon-enhanced emission from hBN spin defects. Boron vacancy defects coupled to NPA structures still show detectable spin contrasts, making them promising candidates for spin-based quantum sensors.

4. References

[1] S.-J. Liang, B. Cheng, X.Y. Cui, F. Miao, Van der Waals Heterostructures for High-Performance Device Applications: Challenges and Opportunities. Adv. Mater. **32**, 1903800 (2020).

[2] J.D. Caldwell, I. Aharonovich, G. Cassabois *et al*. Photonics with hexagonal boron nitride. Nat. Rev. Mater. 2020, **4,** 552–567 (2019).

[3] X. Xu, Z.O. Martin, D. Sychev *et al*, Creating Quantum Emitters in Hexagonal Boron Nitride Deterministically on Chip-Compatible Substrates, Nano Lett. 21, 8182–8189 (2021)

[4] A. Gottscholl, M. Diez, V. Soltamov *et al*. Spin defects in hBN as promising temperature, pressure and magnetic field quantum sensors. Nat. Commun. **12,** 4480 (2021).

[5] X. Gao, B. Jiang, A.E.L.Allcca *et al*, High-contrast plasmonic-enhanced shallow spin defects in hexagonal boron nitride for quantum sensing Nano Lett. **21**, 7708-7714 (2021).

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