High-Contrast Plasmonic-Enhanced Shallow Spin Defects in Hexagonal Boron Nitride for Quantum Sensing

Xingyu Gao, Boyang Jiang, Andres E. Llacsahuanga Allcca, Kunhong Shen, Mohammad A. Sadi, Abhishek B. Solanki, Peng Ju, Zhujing Xu, Pramey Upadhyaya, Yong P. Chen, Sunil A. Bhave, and Tongcang Li*

ABSTRACT: The recently discovered spin defects in hexagonal boron nitride (hBN), a layered van der Waals material, have great potential in quantum sensing. However, the photoluminescence and the contrast of the optically detected magnetic resonance (ODMR) of hBN spin defects are relatively low so far, which limits their sensitivity. Here we report a record-high ODMR contrast of 46% at room temperature and simultaneous enhancement of the photoluminescence of hBN spin defects by up to 17-fold by the surface plasmon of a gold film microwave waveguide. Our results are obtained with shallow boron vacancy spin defects in hBN nanosheets created by low-energy He+ ion implantation and a gold film microwave waveguide fabricated by photolithography. We also explore the effects of microwave and laser powers on the ODMR and improve the sensitivity of hBN spin defects for magnetic field detection. Our results support the promising potential of hBN spin defects for nanoscale quantum sensing.

KEYWORDS: spin defects, hexagonal boron nitride, quantum sensing, optically detected magnetic resonance

Optically active spin defects in wide-band-gap materials have shown great potential for a wide range of emerging technologies, from quantum information processing2,7 to high-resolution sensing of magnetic and electric fields.3–6 Color centers in bulk semiconductors such as diamond7,8 and silicon carbide9,10 are prime examples that reveal optically detected magnetic resonance (ODMR). Recently, atomic defects in layered van der Waals materials such as hexagonal boron nitride (hBN) are attracting increasing attention as alternative candidates for studying light–matter interaction, nanophotonics, and nanoscale sensing.11–14 Atomic defects in hBN are stable in nanosheets as thin as a monolayer and are readily accessible for device integration and top down nanofabrication.15,16 Furthermore, recent experiments discovered that some defects in hBN could be spin addressable at room temperature.17–22 The negatively charged boron vacancy (V_{B}^{-}) spin defect is the most studied one among these defects.20–24 It has spin s = 1, and its orientation is out of plane.25 V_{B}^{-} defects can be generated by neutron irradiation,17,26 ion implantation,27 femtosecond laser writing,28 and electron irradiation.29 Spin defects in thin hBN nanosheets will be useful for quantum sensing and spin optomechanics.30,31 However, so far, the V_{B}^{-} spin defects have relatively low brightness and ODMR contrast, which limits their sensitivity in quantum sensing.22

Here we report high-contrast plasmonic-enhanced shallow V_{B}^{-} spin defects in hBN nanosheets for quantum sensing. We fabricate a gold film coplanar microwave waveguide by photolithography to optimize the homogeneity and local intensity of the microwave for spin control. The hBN nanosheets with spin defects are transferred onto the microwave waveguide and are in contact with the gold surface for both plasmonic emission enhancement and spin control. The surface plasmons32–34 provide broadband emission enhancement covering the wide range of the photoluminescence (PL) of V_{B}^{-} defects from 750 to 950 nm.17 Our method does not require complex nanofabrication or cause adverse effects on quantum sensing. The microwave magnetic field generated by the gold waveguide is parallel to the surface and perpendicular to the orientation of V_{B}^{-} electron spins, which is crucial to achieve high ODMR contrast. With these, we find that the ODMR contrast can reach 46% at room temperature.
Finally, we perform coherent spin control and double Lorentzian model. Here we acquire the basic spin properties of the gold (hBN) flakes. The defects are excited by a 532 nm laser with an NA = 0.9 objective lens, which also collects the PL of defects. We use low-energy \( \text{He}^+ \) ions (200 eV to 3 keV) to implant hBN nanosheets, which creates high-quality \( V_{\text{hBN}}^− \) defects with average depths ranging from 3 to 30 nm and avoids introducing undesired defects. After ion implantation, the hBN flakes are transferred onto a gold microwave stripline on a sapphire substrate and characterized using an ODMR setup (Figure 1a). CW ODMR measurements are performed to probe the basic spin properties of the \( V_{\text{hBN}}^− \) defects using a home-built ion implanter for doping. We use low-power microwave driving. We also observe a similar high contrast with a thick hBN flake placed inside a \( \Omega \)-shaped waveguide, we observe a 6-fold lower ODMR contrast with the same microwave power (Supporting Information, Figure S6). The decrease of the ODMR contrast is due to the misalignment of the driving microwave magnetic field, which is nearly out of plane inside a \( \Omega \)-shaped ring.

Figure 1c shows measured high-contrast ODMR spectra of \( V_{\text{hBN}}^− \) defects at room temperature. Strikingly, with strong microwave driving, we find that these defects can exhibit up to 46% ODMR contrast, which is 1 order of magnitude higher than the best contrast of hBN spin defects reported previously. This value is even larger than the ODMR contrast of diamond nitrogen-vacancy centers at room temperature. The ODMR contrast can readily reach around 20% without significant power broadening using low-power microwave driving. We also observe a similar high contrast with a thick hBN flake on the gold stripline which does not show PL enhancement (Supporting Information, Figure S7). It is noted that such a high ODMR contrast is due to the good alignment of the microwave magnetic driving field and the strong driving field strength. Since the hBN flakes are placed directly on the gold stripline, the driving magnetic field is parallel to its surface and perpendicular to the \( V_{\text{hBN}}^− \) spins, which yields the maximum ODMR contrast. In comparison, for a hBN flake placed inside a \( \Omega \)-shaped waveguide, we observe a 6-fold lower ODMR contrast with the same microwave power (Supporting Information, Figure S6). The decrease of the ODMR contrast is due to the misalignment of the driving microwave magnetic field, which is nearly out of plane inside a \( \Omega \)-shaped ring.

Figure 1. (a) An illustration of the experimental setup for ODMR measurements. An ion implanted hBN nanosheet is placed on top of a gold film microwave stripline. A microwave is delivered through the stripline for spin manipulation. The surface plasmon of the gold surface provides magnetic field sensitivity to be about \( 8 \mu \text{T/} \sqrt{ \text{Hz}} \). Finally, we study the laser power and microwave power dependence of the continuous-wave (CW) ODMR and optimize the magnetic field sensitivity to be about \( 8 \mu \text{T/} \sqrt{ \text{Hz}} \). (b) The energy diagram of a \( V_{\text{hBN}}^− \) defect and the optical pumping cycle between the ground state (GS), the excited state (ES), and the metastable state (MS). A magnetic field induces Zeeman shifts of the spin sublevels. (c) Measured CW ODMR spectra under 100 mW (light blue dots) and 2 W (red squares) microwave driving. With a high-power microwave (2 W), the ODMR contrast can reach 46%. The laser excitation power is 5 mW. The \( V_{\text{hBN}}^− \) defects are generated by 2.5 keV \( \text{He}^+ \) ion implantation. (d) ODMR spectra in different external magnetic fields. A clear splitting of 346 and 560 MHz is observed in 5.9 and 9.8 mT magnetic fields, respectively. Solid curves are fittings with a double Lorentzian model.
ν₀ = D₀/h = 3.47 GHz, where h is the Planck constant. And the splitting between υ₁ and υ₂ is due to the nonzero off-axial ZFS parameter E₂/h = 50 MHz. Figure 1d presents ODMR spectra in different external static magnetic fields. We use a permanent magnet to apply a static magnetic field perpendicular to the nanosheet surface. A translation stage is used to change the position of the magnet and tune the magnetic field strength. With an external static magnetic field B, υ₁ and υ₂ will be split further owing to the Zeeman effect, 

\[ υ₁,₂ = D₀/h ± \sqrt{E₂/h + (gυB)^2}/h, \] 

where g = 2 is the Landé g-factor. The splitting (υ₂ − υ₁) is 560 MHz (346 MHz) at 9.8 mT (5.9 mT).

It is highly desirable to create spin defects as close to the surface as possible without degrading the spin properties for nanoscale quantum sensing. This can decrease the ultimate distance between a sample and the sensor, which can significantly improve the signal. In Figure 2, we study the formation of shallow \( V_{B}^- \) defects by using He⁺ ions. First, we use the Stopping and Range of Ions in Matter (SRIM) software to calculate the densities of vacancies created with different ion energies from 200 eV to 3 keV (Figure 2a,b). The most probable depth is 3.5, 6.4, 15, and 25 nm when the He⁺ ion energy is 300 eV, 600 eV, 1.5 keV, and 2.5 keV, respectively. Then, we perform the CW ODMR measurements on the samples with different doping depths. Here we use weak microwave driving to avoid the power broadening. Therefore, we can extract the nature linewidth of \( V_{B}^- \) defects (Figure 2c). All of the ODMR spectra display similar line widths as well as the contrasts, indicating the hBN spin properties are nearly the same at different doping depths.

For sensing applications, the PL brightness is an important factor that directly affects the sensitivity. Former theoretical studies indicate that the near-infrared optical transition of \( V_{B}^- \) defects is not an electric-dipole-allowed transition and is hence relatively dark. In this context, improving their brightness is a crucial task. Here we utilize surface plasmons of a metallic film to enhance the brightness of the \( V_{B}^- \). Surface plasmons are collective oscillations of coupled electromagnetic waves and free electrons on metallic surfaces. They have large localized electric and magnetic fields which can speed up both radiative and non-radiative decays. We chose plasmonic enhancement because it can cover the whole broad PL spectral range of \( V_{B}^- \) defects. In addition, this method can utilize the metallic surface of our microwave waveguide and does not require complex nanofabrication. Our microwave waveguide is made of a 300 nm thick gold film prepared by electron-beam physical vapor deposition on top of a sapphire wafer. The width of the center microstrip is 50 μm. The hBN flakes with \( V_{B}^- \) defects are transferred onto both the gold film and the sapphire substrate for comparison (Figure 3a). \( V_{B}^- \) defects on both gold and sapphire surfaces display broad PL emission spectra around 810 nm (Figure 3b). Remarkably, the \( V_{B}^- \) defects on the gold film show an order of magnitude higher PL intensity than those on the sapphire substrate under the same laser excitation. Parts c and d of Figure 3 present the PL intensities of these two samples and their PL ratio at different laser excitation powers. The experimental data in Figure 3c are fit to \( I = I_{sat}/(1 + P_{sat}/P_{laser}) \), where \( I \) is the PL intensity of the \( V_{B}^- \), \( I_{sat} \) is the saturation PL intensity, \( P_{laser} \) is the excitation laser power, and \( P_{sat} \) is the saturation laser power. On a gold film, the \( V_{B}^- \) defects show up to 17-fold enhancement of PL intensities under low-power excitation. We also observe a strong modification of the saturation behavior (Figure 3c). The laser saturation power \( P_{sat} \) is reduced by a factor of \( S \), and the saturation PL count rate \( I_{sat} \) is increased by around 3.5 times. These indicate that a gold film can improve the quantum efficiency of \( V_{B}^- \) defects significantly.

We also study the effect of the separation between the gold film and \( V_{B}^- \) defects on the brightness enhancement. We transfer hBN flakes with different thickness onto the gold microstrip, so that we can get various distances between the \( V_{B}^- \) defects and the gold film. The thickness of hBN nanosheets is measured by an atomic force microscope (AFM). The PL intensities are measured before and after transfer. Here we use 600 eV He⁺ ions to generate shallow \( V_{B}^- \) with a most probable depth of 6.4 nm. Therefore, the average separation between the gold film and \( V_{B}^- \) defects is equal to the thickness of hBN nanosheets subtracted by 6.4 nm. As a result, we observe a strong thickness dependence of the PL enhancement. The highest enhancement is obtained when the hBN flake thickness is around 32 nm (Figure 3e). Our result is consistent with the former result on plasmonic enhancement of quantum dots on a gold surface. When a hBN nanosheet is too thin, the non-radiative decay dominates. There is also little brightness enhancement when the hBN nanosheet is too thick because the surface plasmonic modes decay exponentially away from the surface. Thus, there is an optimal thickness for plasmonic enhancement. In addition, we characterize the electron spin initialization time of \( V_{B}^- \) defects on both gold and sapphire surfaces as a function of the power of the 532 nm excitation laser (see the Supporting Information for more details). We find that the required spin initialization time can be about 100 ns under high-power excitation (Figure 3f). Thus, the ground-state recovery time should be less than 100 ns, which is much shorter than the former theoretical prediction. Our results may be helpful for future theoretical works on \( V_{B}^- \) spin defects. The gold film reduces the spin initialization time (Figure 3f), which also speeds up quantum sensing.

Sensitivity is the most important parameter to determine the performance of a sensor. To measure an external static magnetic field with spin defects, a common way is to use CW ODMR to detect the Zeeman shifts of the spin sublevels caused by the magnetic field. The precision to determine the magnetic field is directly affected by the photon count rate R,
the ODMR contrast \( \eta_C \), and the linewidth \( \Delta\nu \), following the equation \(^{17}\)

\[
\eta_C = A \times \frac{h}{g\mu_B} \times \frac{\Delta\nu}{C\sqrt{R}}
\]

(1)

where \( \mu_B \) is the Bohr magneton. In this expression, \( A \) is a numerical parameter related to the specific line shape function.

For a Lorentzian profile, \( A \approx 0.77 \). The values of \( C, \Delta\nu, \) and \( R \) are further related to the microwave power and laser power \(^{17}\) (see the Supporting Information). To improve the detection sensitivity, it is crucial to increase the count rate and contrast as high as possible without significant power broadening of the linewidth.

Here we perform a group of CW ODMR measurements with various microwave powers and laser powers to find the

Figure 3. (a) Illustrations of hBN nanosheets with spin defects on top of a 300 nm-thick gold film and on top of a bare sapphire wafer for comparison. (b) A comparison of the PL spectra of \( V_{\text{B}}^- \) defects on a gold film and on a sapphire substrate. The gold film enhances the PL count rate substantially. The \( V_{\text{B}}^- \) defects are generated by 2.5 keV He\(^+\) ions. (c) PL intensities of \( V_{\text{B}}^- \) defects on a gold film and a sapphire substrate as functions of the laser power. (d) PL enhancement at different laser powers. The enhancement is obtained by calculating the ratio of PL intensities of the \( V_{\text{B}}^- \) defects on the gold film and the sapphire substrate. The enhancement is up to 17 when the laser power is low. (e) Dependence of the PL enhancement on the thickness of the hBN nanosheet. The highest PL enhancement is obtained when the thickness is around 35 nm. The \( V_{\text{B}}^- \) defects are generated by 600 eV He\(^+\) ions and are near the top surface of the hBN nanosheet. (f) The required time for optically polarizing \( V_{\text{B}}^- \) electron spins as a function of the laser power.

Figure 4. (a) Typical CW ODMR spectra of \( V_{\text{B}}^- \) spin defects at different microwave powers. The laser excitation power is 1 mW. No external magnetic field is applied. (b) ODMR peak contrast as a function of the microwave power. (c) Microwave power dependence of the ODMR linewidth. (d) Magnetic field sensitivity as a function of the microwave power. The orange dots and blue squares are experimental data. Solid curves are fittings with theoretical models.
optimal conditions for magnetic field sensing (Figure 4). Figure 4a presents three typical CW ODMR spectra at different microwave powers. Under low-power 40 mW microwave driving, we obtain an ODMR contrast of ~10%. Such a low microwave power does not induce significant spectral power broadening. A natural linewidth can be extracted as ~110 MHz. With an increasing microwave power, we first observe a significant improvement of the contrast without much spectral broadening. When we increase the microwave power further, the linewidth broadening becomes severe. Parts b–d of Figure 4 present the quantitative measurements of the ODMR contrast, linewidth, and sensitivity as functions of the microwave power. The experimental results fit well with theoretical models, as discussed in the Supporting Information. Here we perform the experiments at two different laser powers (1 and 5 mW). The 5 mW laser excitation gives a broader linewidth but a higher saturation ODMR contrast compared to those with 1 mW laser excitation. As a result, if we increase the microwave power, the sensitivity is first improved owing to the increase of the ODMR contrast and then becomes worse when the spectral power broadening dominates. The best sensitivity that we have achieved is about 8 μT/√Hz. This sensitivity is 10 times better than the former result with hBN spin defects (our system also has a better spatial resolution).\(^6\) This sensitivity will be enough for studying many interesting phenomena in magnetic materials. For example, the magnetic field generated by a monolayer CrI\(_3\) (a 2D van der Waals magnet) is on the order of 200 μT.\(^6\)

Finally, we perform pulsed ODMR measurements to determine the spin–lattice relaxation time \(T_1\) and the spin coherence time \(T_2\) of the shallow \(V_{\text{B}}^\text{−}\) defects generated by ion implantation. \(T_1\) and \(T_2\) of hBN spin defects have only been measured for neutron irradiated samples before.\(^{19,26}\) It will be useful to know their values for our shallow spin defects created by ion implantation. In addition, pulsed ODMR measurements are inevitable steps for realizing more complex sensing protocols. A pulsed ODMR measurement consists of optical initialization of the ground state, coherent manipulation of the spin state with microwave pulses, and optical readout of the final spin state. Here we add an external magnetic field of 13 mT parallel to the hBN nanosheet to split the spin sublevels. \(m_I = 1, 0\) states are used as the two-level spin system to carry out the spin coherent control. Figure 5a shows the Rabi oscillation as a function of the microwave power. The data is fit using \(A + B_1 \exp(-\pi T_2^*) \cos(2\pi f_T/2 + \phi_1) + B_2 \exp(-\pi/T_2^*) \cos(2\pi f_T/2 + \phi_2)\). We observe an oscillation with two Rabi frequencies, which are in the tens of megahertz range. The exponential decay of one oscillation component gives the spin-dephasing time of \(T_2^* = 120\) ns. To gain more insight into the spin properties of the \(V_{\text{B}}^\text{−}\) defects at different depths, we measure the spin–lattice relaxation times \(T_1\) and spin–spin relaxation times \(T_2\) of the \(V_{\text{B}}^\text{−}\) defects created with different ion implantation energies. The pulse sequences are shown as insets in the left panels of Figure 5b,c. By fitting the results, \(T_1\) and \(T_2\) are obtained as \(\sim 17\) and \(\sim 1.1\) μs, respectively. As shown in Figure 5b,c, both \(T_1\) and \(T_2\) are independent of the ion energy, indicating the spin properties of the \(V_{\text{B}}^\text{−}\) defects are nearly independent of the depth. The depth-independent \(T_2\) suggests that the coherence times are not limited by the surface. They are likely dominated by nuclear spin noise but may also be limited by the inhomogeneity, as we use an ensemble of spin defects instead of a single spin defect.\(^38\)

In conclusion, we have realized a significant improvement of the ODMR contrast and the brightness of hBN \(V_{\text{B}}^\text{−}\) spin defects. We observe a record-high ODMR contrast of 46%, which is 1 order of magnitude higher than the best former result with hBN. We use low-energy He\(^+\) ion implantation to create \(V_{\text{B}}^\text{−}\) defects as shallow as 3 nm in hBN nanosheets. Moreover, both CW and pulsed ODMR measurements display that their spin properties are nearly independent of the ion implantation energy. This result confirms the feasibility to create high-quality \(V_{\text{B}}^\text{−}\) defects proximal to the hBN surface. In addition, we utilize the gold film surface plasmon to enhance the brightness of \(V_{\text{B}}^\text{−}\) defects and obtain an up to 17-fold enhancement of the PL intensity. We also explore the effects of laser power and microwave power on the CW ODMR contrast and linewidth. With these, we achieve a CW ODMR sensitivity around 8 μT/√Hz. We expect that the PL can be enhanced further with plasmonic nanoantennas,\(^{39}\) and the magnetic field sensitivity can be improved by using more powerful pulsed sensing protocols. In addition, during the preparation of this manuscript, we became aware of a related work\(^{30}\) that reported a PL enhancement of hBN spin defects with a photonic cavity by a factor of ~6 at high NA collection and an ODMR contrast of about 5%. The surface plasmon may be combined with a photonic cavity to obtain better results in the future. Our work strongly supports the promising potential of \(V_{\text{B}}^\text{−}\) defects as a nanoscale sensor in a 2D material platform.
ASSOCIATED CONTENT

Supporting Information
The Supporting Information is available at https://pubs.acs.org/doi/10.1021/acs.nanolett.1c02495.

Additional experimental details, methods, analysis of results, and figures of photoluminescence intensities and ODMR contrasts (PDF)

AUTHOR INFORMATION

Corresponding Author
Tongcang Li — Department of Physics and Astronomy, Purdue University, West Lafayette, Indiana 47907, United States; Elmore Family School of Electrical and Computer Engineering, Birck Nanotechnology Center, and Purdue Quantum Science and Engineering Institute, Purdue University, West Lafayette, Indiana 47907, United States; orcid.org/0000-0003-3308-8718; Email: tcli@purdue.edu

Authors
Xingyu Gao — Department of Physics and Astronomy, Purdue University, West Lafayette, Indiana 47907, United States
Boyang Jiang — Elmore Family School of Electrical and Computer Engineering, Purdue University, West Lafayette, Indiana 47907, United States
Andres E. Llacsahunga Alcca — Department of Physics and Astronomy, Purdue University, West Lafayette, Indiana 47907, United States
Kunhong Shen — Department of Physics and Astronomy, Purdue University, West Lafayette, Indiana 47907, United States
Mohammad A. Sadi — Elmore Family School of Electrical and Computer Engineering, Purdue University, West Lafayette, Indiana 47907, United States
Abhishek B. Solanki — Elmore Family School of Electrical and Computer Engineering, Purdue University, West Lafayette, Indiana 47907, United States
Peng Ju — Department of Physics and Astronomy, Purdue University, West Lafayette, Indiana 47907, United States
Zhijing Xu — Department of Physics and Astronomy, Purdue University, West Lafayette, Indiana 47907, United States
Pramey Upadhyaya — Elmore Family School of Electrical and Computer Engineering, Purdue University, West Lafayette, Indiana 47907, United States
Yong P. Chen — Department of Physics and Astronomy, Purdue University, West Lafayette, Indiana 47907, United States; Elmore Family School of Electrical and Computer Engineering, Birck Nanotechnology Center, and Purdue Quantum Science and Engineering Institute, Purdue University, West Lafayette, Indiana 47907, United States
Sunil A. Bhave — Elmore Family School of Electrical and Computer Engineering and Birck Nanotechnology Center, Purdue University, West Lafayette, Indiana 47907, United States; orcid.org/0000-0001-7193-2241

Complete contact information is available at: https://pubs.acs.org/doi/10.1021/acs.nanolett.1c02495

Notes
The authors declare no competing financial interest.
(15) Tran, T. T.; Bray, K.; Ford, M. J.; Toth, M.; Aharonovich, I. Quantum emission from hexagonal boron nitride monolayers. Nat. Nanotechnol. 2016, 11, 37–41.

(16) Palombo Blascetta, N.; Liebel, M.; Lu, X.; Taniguchi, T.; Watanabe, K.; Efetov, D. K.; van Hulst, N. F. Nanoscale imaging and control of hexagonal boron nitride single photon emitters by a resonant nanoantenna. Nano Lett. 2020, 20, 1992–1999.

(17) Gottscholl, A.; Kianinia, M.; Soltamov, V.; Orlinski, S.; Mamin, G.; Bradac, C.; Kasper, C.; Krambok, K.; Sperlich, A.; Toth, M.; et al. Initialization and read-out of intrinsic spin defects in a van der Waals crystal at room temperature. Nat. Mater. 2020, 19, 540–545.

(18) Mendelson, N.; Chugh, D.; Reimers, J. R.; Cheng, T. S.; Gottscholl, A.; Long, H.; Mellor, C. J.; Zettl, A.; Dyakonov, V.; Beton, P. H.; et al. Identifying carbon as the source of visible single-photon emission from hexagonal boron nitride. Nat. Mater. 2021, 20, 321–328.

(19) Gottscholl, A.; Diez, M.; Soltamov, V.; Kasper, C.; Sperlich, A.; Kianinia, M.; Bradac, A.; Aharonovich, I.; Dyakonov, V. Room temperature coherent control of spin defects in hexagonal boron nitride. Science Advances 2021, 7, No. eaab3630.

(20) Chejanovsky, N.; Mukherjee, A.; Geng, J.; Chen, Y.-C.; Kim, Y.; Denisenko, A.; Finkler, A.; Taniguchi, T.; Watanabe, K.; Dasari, D. B. R.; et al. Single-spin resonance in a van der Waals embedded paramagnetic defect. Nat. Mater. 2021, 20, 1079–1084.

(21) Stern, H. L.; Jarman, J.; Gu, Q.; Barker, S. E.; Mendelson, N.; Chugh, D.; Schott, S.; Tan, H. H.; Sirringhaus, H.; Aharonovich, I.; et al. Room-temperature optically detected magnetic resonance of single defects in hexagonal boron nitride. 2021, arXiv:2103.16494. arXiv.org e-Print archive. https://arxiv.org/abs/2103.16494 (accessed August 12, 2021).

(22) Gottscholl, A.; Diez, M.; Soltamov, V.; Kasper, C.; Kraufh, D.; Sperlich, A.; Kianinia, M.; Bradac, C.; Aharonovich, I.; Dyakonov, V. Spin defects in hBN as promising temperature, pressure and magnetic field quantum sensors. Nat. Commun. 2021, 12, 4480.

(23) Abdí, M.; Chou, J.-P.; Gali, A.; Plenio, M. B. Color centers in hexagonal boron nitride monolayers: a group theory and ab initio analysis. ACS Photonics 2018, 5, 1967–1976.

(24) Ivády, V.; Barca, G.; Thiering, G.; Li, S.; Hamdi, H.; Chou, J.-P.; Legeza, Ö.; Gali, A. Ab initio theory of the negatively charged boron vacancy qubit in hexagonal boron nitride. npj Computational Materials 2020, 6, 41.

(25) Reimers, J. R.; Shen, J.; Kianinia, M.; Bradac, C.; Aharonovich, I.; Ford, M. J.; Piecuch, P. Photoluminescence, photophysics, and photochemistry of the $V_{\text{B}}^-$ defect in hexagonal boron nitride. Phys. Rev. B: Condens. Matter Mater. Phys. 2020, 102, 144105.

(26) Liu, W.; et al. Rabi oscillation of $V_{\text{B}}^-$ spin in hexagonal boron nitride. 2021, arXiv:2101.11220. arXiv.org e-Print archive. https://arxiv.org/abs/2101.11220 (accessed August 12, 2021).

(27) Kianinia, M.; White, S.; Froch, J. E.; Bradac, C.; Aharonovich, I. Generation of spin defects in hexagonal boron nitride. ACS Photonics 2020, 7, 2147–2152.

(28) Gao, X.; Pandey, S.; Kianinia, M.; Ahn, J.; Ju, P.; Aharonovich, I.; Shivaram, N.; Li, T. Femtosecond Laser Writing of Spin Defects in Hexagonal Boron Nitride. ACS Photonics 2021, 8, 994–1000.

(29) Murzakhano, F. F.; Yavkin, B. V.; Mamin, G. V.; Orlinski, S. B.; Mumdzhii, I. E.; Gracheva, I. N.; Gabbasov, B. F.; Smirnov, A. N.; Davydov, V. Y.; Soltamov, V. A. Creation of Negatively Charged Boron Vacancies in Hexagonal Boron Nitride Crystal by Electron Irradiation and Mechanism of Inhomogeneous Broadening of Boron Vacancy-Related Spin Resonance Lines. Nanomaterials 2021, 11, 1373.

(30) Abdí, M.; Hwang, M.-J.; Aghtar, M.; Plenio, M. B. Spin-Mechanical Scheme with Color Centers in Hexagonal Boron Nitride Membranes. Phys. Rev. Lett. 2017, 119, 233602.

(31) Li, P.-B.; Zhou, Y.; Gao, W.-B.; Nori, F. Enhancing Spin-Photon and Spin-Spin Interactions Using Linear Resources in a Hybrid Quantum System. Phys. Rev. Lett. 2020, 125, 153602.